

'Hybrid' non-destructive imaging techniques for engineering materials applications

Nikolaos Baimpas

Trinity College



Supervisor: **Professor Alexander M. Korsunsky**

Thesis submitted for the degree of Doctor of Philosophy
Department of Engineering Science, University of Oxford

Trinity Term 2014

Abstract

The combination of X-ray imaging and diffraction techniques provides a unique tool for structural and mechanical analysis of engineering components. A variety of modes can be employed in terms of the spatial resolution (length-scale), time resolution (frequency), and the nature of the physical quantity being interrogated. This thesis describes my contributions towards the development of novel X-ray “rich” imaging experimental techniques and data interpretation. The experimental findings have been validated via comparison with other experimental methods and numerical modelling.

The combination of fast acquisition rate and high penetration properties of X-ray beams allows the collection of high-resolution 3-D tomographic data sets at submicron resolution during *in situ* deformation experiments. Digital Volume Correlation analysis tools developed in this study help understand crack propagation mechanisms in quasi-brittle materials and elasto-plastic deformation in co-sprayed composites.

For the cases of crystalline specimens where the knowledge of “live” or residual elastic strain distributions is required, diffraction techniques have been advanced. *Diffraction Strain Tomography* (DST) allows non-destructive reconstruction of the 2-D (in-plane) variation of the out-of-plane strain component. Another diffraction modality dubbed *Laue Orientation Tomography* (LOT), a grain mapping approach has been proposed and developed based on the translate-rotate tomographic acquisition strategy. It allows the reconstruction of grain shape and orientation within polycrystalline samples, and provides information about intragranular lattice strain and distortion. The implications of this method have been thoroughly investigated.

State-of-the-art engineering characterisation techniques evolve towards scrutinising submicron scale structural features and strain variation using the complementarity of X-ray imaging and diffraction. The first successful feasibility study is reported of *in operando* stress analysis in an internal combustion engine.

Finally, further advancement of ‘rich’ imaging techniques is illustrated via the first successful application of Time-of-Flight Neutron Diffraction Strain (TOF-NDST) tomography for non-destructive reconstruction of the complete strain tensor using an inverse eigenstrain formulation.

Acknowledgements

The work described in this thesis was carried out in the Department of Engineering Science at the University of Oxford, between May 2010 and May 2014, under the supervision of Prof. Alexander M. Korsunsky.

First and foremost, I would like to express my deepest gratitude to my supervisor Prof. Alexander M. Korsunsky. The sum of knowledge, continuous support and guidance, together with an honest approach into the pure principles of scientific research has been Alexander's greatest contribution towards successful completions of my research work and thesis. His kind attitude and inspirational discussions allowed me to explore new possibilities and overcome difficulties during my DPhil student time. I don't think I could achieve what I have today without his selfless help.

I am very grateful to the members of my family, my father Georgios, my mother Aikaterini and my sister Christina for coping with me and supporting all the way. A great thanks to all my dear friends in Oxford, London and Greece for all the nice moments we had together.

A number of collaborators and colleagues in Oxford and elsewhere have contributed to this projects. Many thanks to: Mengyin Xie, Alexander J. G. Lunt, Tan Sui, Felix Hoffmann, Brian Abbey, Xu Song, Mehmet Kartal, Kalin Dragnevski, Mahmoud Mostafavi, James T. Marrow, Igor Dolbnya, Andrew Malandain, Jean-Sébastien Micha, Christian Mocuta, Michael Drakopoulos, Christina Reinhard, Thomas Connolley, Leigh Connor, Richard Duffin, Sophie Eve, Eric Le Bourhis, Dominique Thiaudière and Chris Hardie.

My thanks also go to the staff and students of Trinity College. It has been a delightful place to live, study and work.

Finally, I would like to acknowledge the funding I received from the EPSRC and the beamtime allocations from the DLS, Soleil and the ESRF.

Common abbreviations

AMPW	Asymmetric Multipole Wiggler
BCC	Body Centered Cubic (Crystal Structure)
CCD	Charge-coupled Device
CF	Carbon Fibre
CP	Commercially Pure
CT	Computerised Tomography
DAXM	Differential Aperture X-ray Microscopy
DCM	Double Crystal Monochromator
DCT	Diffraction Contrast Tomography
DIC	Digital Image Correlation
DLD	Direct Laser Deposition
DLS	Diamond Light Source, Didcot, UK
DMLS	Direct Metal Laser Sintering
DST	Diffraction Strain Tomography
DVC	Digital Volume Correlation
EDXRD	Energy Dispersive X-ray Diffraction
EBSD	Electron Backscatter Diffraction
ESRF	European Synchrotron Radiation Facility, Grenoble, France
FCC	Face Centered Cubic (Crystal Structure)
FEM	Finite Element Modelling
FWHM	Full Width at Half Maximum
KB	Kirkpatrick-Baez (Focussing Mirrors)

LOT	Laue Orientation Tomography
RSM	Reciprocal Space Mapping
SAXS	Small Angle X-ray Scattering (X-ray diffraction)
SEM	Scanning Electron Microscopy
SF	Safety Factors
SLS	Swiss Light Source, Villigen, Switzerland
SSD	Statistically Stored Dislocations
TEM	Transmission Electron Microscopy
TERM	Tomographic Eigenstrain Reconstruction Method
TOF-NDST	Time-of-Flight Neutron Diffraction Strain Tomography
WAXS	Wide Angle X-ray Scattering (X-ray diffraction)

Contents

1. INTRODUCTION	1
1.1. HISTORICAL SKETCH OF ENGINEERING AND DIFFRACTION	8
1.2. X-RAY LIGHT TO CONDENSED MATTER INTERACTION MECHANISMS	10
1.3. X-RAY TRANSMISSION IMAGING AND TOMOGRAPHY	14
1.4. PROBING MATERIAL DEFORMATION AND STRUCTURE USING X-RAY LIGHT	16
2. SCIENTIFIC BACKGROUND AND METHODS	22
2.1. X-RAY IMAGING TECHNIQUES	22
2.2. X-RAY AND NEUTRON DIFFRACTION TECHNIQUES	26
2.3. MICRO-MECHANICAL DEFORMATION MEASURING TECHNIQUES.....	37
2.4. EIGENSTRAIN THEORY	44
2.5. CONCLUSIONS AND REMARKS.....	48
3. TOMOGRAPHY-DIGITAL IMAGE CORRELATION.....	50
3.1. SYNCHROTRON-BASED TOMOGRAPHIC DATA RECONSTRUCTION ALGORITHM	52
3.2. DIGITAL VOLUME CORRELATION (DVC) OF SYNCHROTRON X-RAY TOMOGRAPHY DATASETS.....	57
3.3. THREE-DIMENSIONAL CRACK OBSERVATION, QUANTIFICATION AND SIMULATION IN QUASI-BRITTLE MATERIALS ..	60
3.3.1. <i>Experimental Setup - Data Processing</i>	62
3.3.2. <i>Finite element simulation</i>	69
3.3.3. <i>Conclusions</i>	74
3.4. THREE-DIMENSIONAL MULTI-SCALE <i>IN SITU</i> DEFORMATION ANALYSIS OF (Al-12%Si)-12%Ti METAL-METAL COMPOSITE.....	76
3.4.1. <i>Experimental Setup - Data Processing</i>	78
3.4.2. <i>Finite element modelling</i>	82
3.4.3. <i>Results - Discussion</i>	83
3.5. CONCLUSIONS AND REMARKS.....	90
4. X-RAY DIFFRACTION ‘RICH’ TOMOGRAPHY TECHNIQUES.....	92
4.1. TOMOGRAPHIC ACQUISITION PRINCIPLES	94
4.2. POWDER DIFFRACTION STRAIN TOMOGRAPHY (DST)	96
4.3. DST ON Ni SUPERALLOY DIRECT LASER DEPOSITION (DLD) TURBINE BLADE.....	109
4.3.1. <i>Materials and applications</i>	109
4.3.2. <i>Diffraction and imaging measurements and interpretation</i>	111
4.3.3. <i>Contour method</i>	116
4.3.4. <i>Results and conclusions</i>	117
4.4. MULTIPHASE DST ON HUMAN DENTAL TISSUE	120
4.4.1. <i>Materials and applications</i>	120
4.4.2. <i>Multi-phase DST implementation and tomographic imaging</i>	122
4.4.3. <i>FIB-DIC</i>	127
4.4.4. <i>Results and conclusions</i>	129
4.5. <i>IN SITU</i> LOADING DST OF YTTRIA-STABILISED ZIRCONIA FIXED PROSTHETIC DENTURE.....	132
4.5.1. <i>Materials and applications</i>	132
4.5.2. <i>FPD shape analysis and modelling</i>	135

4.5.3. Diffraction measurements and interpretation	137
4.5.4. Results and conclusions	139
4.6. X-RAY LAUE ORIENTATION TOMOGRAPHY (LOT)	141
4.6.1. Introduction - Principles.....	142
4.6.2. Laue pattern prediction from rotation matrices (R)	144
4.6.3. Laue diffraction data collection and implementation	146
4.6.4. Rodriguez Vector formulation	153
4.6.5. Results and discussion	154
4.7. CONCLUSIONS ON 'RICH' TOMOGRAPHY TECHNIQUES	160
5. IN SITU 'HYBRID' STRAIN MAPPING TECHNIQUES.....	163
5.1. IN OPERANDO MECHANICAL ANALYSIS OF INTERNAL COMBUSTION ENGINE BY SYNCHROTRON TIME RESOLVED X-RAY DIFFRACTION AND IMAGING	166
5.1.1. Experimental Setup – EH2 I12, DLS.....	171
5.1.2. Compound radiography inside an Internal Combustion Engine	176
5.1.3. In operando stress quantification inside Internal Combustion Engines	178
5.1.4. Conclusions	181
5.2. STRESS EVALUATION IN THIN FILMS: MICRO-FOCUS SYNCHROTRON X-RAY DIFFRACTION COMBINED WITH FOCUSED ION BEAM PATTERNING	181
5.2.1. Experimental Setup – DIFFABS, Soleil Synchrotron.....	185
5.2.2. Results and discussion	189
5.2.3. Conclusions and remarks	191
5.3. NANO-SCALE MAPPING OF LATTICE STRAIN AND ORIENTATION INSIDE CARBON CORE SiC FIBRES	191
5.3.1. Experimental Setup – B16, DLS.....	192
5.3.2. Focused-Ion-Beam (FIB) – Digital-Image-Correlation (DIC) for microscopic strain analysis	195
5.3.3. Inverse Eigenstrain Finite Element Analysis formulation	196
5.3.4. Results and discussion	197
5.4. CONCLUSIONS AND REMARKS.....	204
6. NEUTRON DIFFRACTION TIME-OF-FLIGHT 'RICH' TOMOGRAPHY TECHNIQUE.....	206
6.1. EXPERIMENTAL SETUP FOR TRANSMISSION BRAGG-EDGE DIFFRACTION TOMOGRAPHY, ENGIN-X, ISIS	207
6.2. DIRECT PROBLEM: DETERMINATION OF RESIDUAL ELASTIC STRAIN FROM GIVEN EIGENSTRAIN	208
6.3. INVERSE EIGENSTRAIN ANALYSIS PROBLEM	211
6.4. MODELLING STEP-WISE CONSTANT EIGENSTRAIN REPRESENTATION	213
6.5. OPTIMISING RECONSTRUCTION USING THE CONSTRAINED MODEL APPROACH.....	216
6.6. RESULTS AND DISCUSSION	219
6.7. CONCLUSIONS	223
7. CONCLUSIONS AND OVERVIEW	225
8. REFERENCES	234

Chapter 1

Introduction

Non-destructive, quantitative and analytical determination of internal strain distributions within engineering components and materials can be only accomplished by a few experimental techniques. The well-established framework of penetrating radiation (X-ray and neutron beams) in diffraction and imaging modes plays a pivotal role to the investigation of the mechanical behaviour of structural materials under external load application. The in-depth understanding of the inter-relationship between material structure and properties is of fundamental importance across a wide range of applications in engineering sciences. The ultimate design goal for most engineering components is the optimisation of its performance and durability in service. The factors that can directly influence the mode of response and longevity of engineering structures are related to the processing history from the raw material to the final manufactured product. The mode of deformation response to loading can be separated into elastic (i.e. fully recoverable along the same path) and inelastic, with the latter being further classified into time-independent and time-dependent plasticity, etc. A combined multi-scale approach to in-depth characterization of the structure and deformation behaviour of engineering materials offers the prospect of advancing the capabilities of engineering design and development beyond the current state-of-the-art.

In modern practice, the demands of novel engineering applications require careful tailoring of the mechanical properties and deformation behaviour of new materials. High performance

applications progressively require careful tuning of the physical and chemical properties of materials to develop designs that are lightweight and high-strength, yet reliable and safe. This must rely on material property characterisation techniques that combine high throughput with high reliability. Once the engineering components and assemblies enter service, non-destructive testing and structural health monitoring are required to ensure safety of operation. The availability of high sensitivity, high resolution measurement techniques ensures better insight into damage initiation and growth, which in turn underpins safe extension of service time, together with the reduction in size and weight, and the amelioration of the related costs and risks.

In applied science and engineering context, the use of suitable experimental techniques develops in combination with flexible and diverse sample environments that are capable of simulating the in-service conditions to a high degree of fidelity. In this global context, the discovery of X-rays in late 1895 by Wilhelm Röntgen has played a pivotal role in natural sciences, particularly in physics, chemistry, biology and engineering. Rapid evolution of field applications and systematic measuring techniques has established X-rays as a ubiquitous tool for research in almost all branches of scientific endeavour [1], from the determination of internal architecture in biological cells, the chemical decomposition, structural materials investigation and provenance of non-destructive imaging within the bulk of non-transparent archaeological artefacts.

The fundamental electromagnetic properties of X-rays are especially important for investigating the properties of materials. They are able to probe deeply and non-destructively in solid materials with their degree of penetration depending on their energy and on the electron density (hence the atomic weights and spatial distribution within atoms) of the sample under investigation [2]. In addition, X-ray photons can contain energies of several kiloelectronvolts (keV) and thus have wavelengths comparable to those of typical atomic spacings in solid materials measured in Angstroms ($1\text{\AA} = 10^{-10}\text{m}$). The crystalline lattice structure itself can

therefore act as an interference grating (i.e. regular array of scattering centres) for X-rays, which can therefore be diffracted [3].

The introduction of 3rd generation high-brilliance synchrotron sources, together with the development of purpose-built engineering instruments, provided additional tools for the investigation of a variety of fundamental material processes. Synchrotron research facilities nowadays generate tuneable beams of electromagnetic radiation from the low energy to the hard X-ray regime, with intensities (defined by their "flux" or "brilliance") many orders of magnitude greater than those produced by laboratory based sources [1]. X-ray and neutron beams make it possible to use the superior penetration ability and the scattering from crystal lattices to access information about the state of strain and damage in the interior (bulk) of materials and components. Today, there are more than 70 facilities available world-wide in operation or under construction, providing services for approximately 100.000 users from virtually every discipline of the natural sciences [1].

An important step in the experimental study of engineering materials is to define the "gauge" volume. This term refers to the region of material that is illuminated by the incident beam and contributes to the signal formed on the detector. The gauge volume size is determined not only by the ultimate resolution sought, but also by the specimen size and the choice of length scale: atomistic (Å), microscopic (µm) or macroscopic (mm).

Figure 1.1 demonstrates examples of Carbon Fibre (CF) used in applications ranging from the monocoque of the current McLaren P1 sports car to the monofilament cores of SCS-6 SiC fibres used to reinforce Ti-alloy matrices for aerospace composites. The car monocoque design requires high rigidity, low weight and good impact resistance. The laminate carbon fibre (CF) structure can be examined after the final assembly for its overall stiffness to confirm numerical Finite Element (FE) model calculation. The monocoque test designs are subjected to crash tests to ensure adequate integrity and thus protection offered to the driver. The CF woven structure before tempering shown in Figure 1.1 (b) consists of bundles of several thousands of CFs put

together into tows and subsequently woven to various configurations according to the desired properties. Alternatively, individual carbon 30 μm monofilaments have been used to grow uniaxial SiC ceramic fibres that provide reinforcement for aerospace Ti alloys [4] (Fig. 1.1 (c)). The inherent light weight nature of the CF monofilament and their remarkable tensile strength ($>4\text{GPa}$) has actively assisted the weight reduction in aerospace engine casings at elevated temperatures up to 350°C [5].

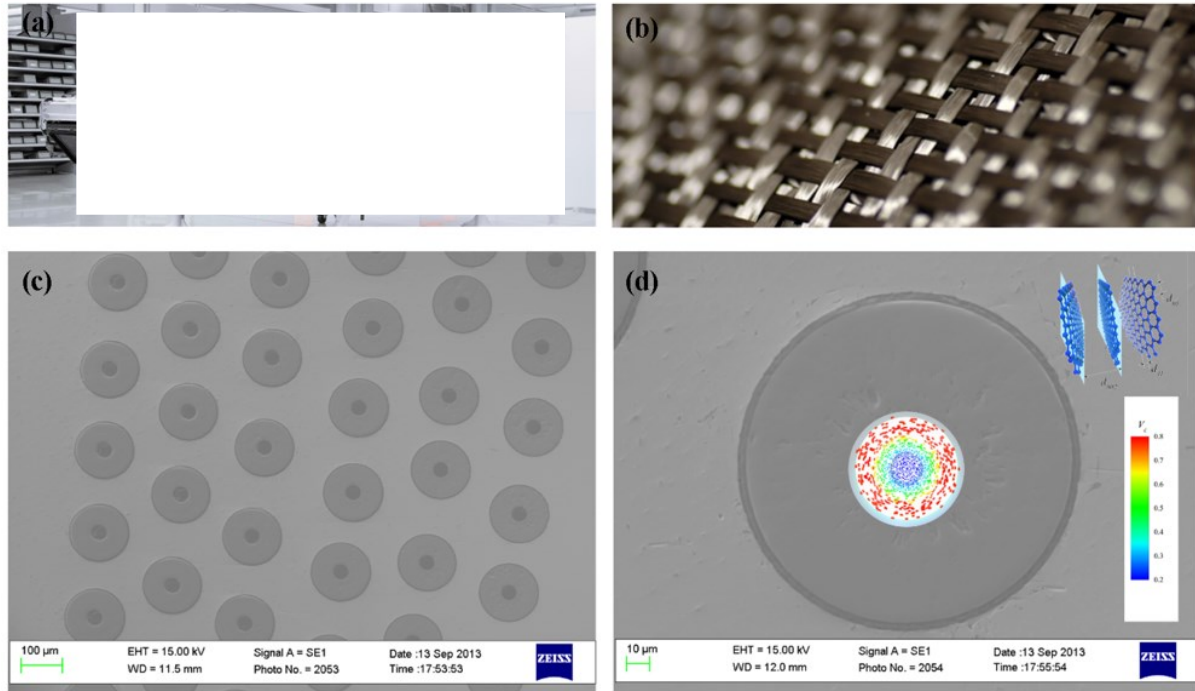


Figure 1.1: The evolution of length scales of study for materials such as Carbon Fibres. From the final product assembly (cm) [6] on (a) to woven carbon fibre bundles (mm) (b), down to carbon monofilaments used as reinforcement (μm) in engineering alloy composites (c) and (d) the study of the structural properties (nm) and lattice deformation ($\text{\AA}$).

The macroscopic examination and testing of final assemblies can provide accurate characterisation of the mechanical response and can be used to inform FE modelling to aid model validation and design optimisation. Profound understanding of the behaviour and the interpretation of the deformation and failure mechanisms require intensive analysis of the relationship between the internal microstructure and the macroscopic component behaviour (Fig. 1.1 (d)). Much of the present understanding of the morphology and texture at the very shortest length scales derives from TEM investigations that reveal the lattice properties and arrangement at the nanoscale [7-9]. Nevertheless, such investigations are by default very local (typical samples are in the order of a few tenths of nanometers), require elaborate sample

preparation and have limited scope for *in situ* sample manipulation and loading, thermal and mechanical. The ability to characterise complex hierarchical structures such as carbon fibres (Fig. 1.1 (d)) or thin films in composite systems across the scales therefore remains a challenge.

Elastic Strain Engineering (ESE)

It is well known that the properties of a crystalline material depend on the lattice parameter/s and the size-shape of the unit cell. A systematic approach towards investigating the micro- and nano-structure can contribute to the understanding of the mechanical behaviour of crystalline materials across scales. One step further would be to manage material properties, by tailoring individual micro-structural characteristics, in such way that materials with enhanced deformation behaviour can be obtained. Typically, elastic strains in traditional materials do not exceed 0.2 – 0.3% before inelastic mechanisms become activated, such as plasticity or fracture. However, some novel nanomaterials have now been identified that can sustain shear or tensile strains in excess of 1% [10]. The mechanical behaviour of these ultra-strength materials is significantly different from their conventional counterparts and mainly is the result of size and dimensional effects and the low density of defects. Elastic strain engineering (ESE) is a recently introduced concept of the system of approaches for enhancing the microstructural characteristics of materials and harnessing the benefits of lattice transformation changes to novel structures and applications.

The strain $\boldsymbol{\varepsilon}$ is a symmetric 3-D tensor of the second rank, with six independent components. The total strain $\boldsymbol{\varepsilon}$ at any given point can be deduced by adding the contributions of elastic strain $\boldsymbol{\varepsilon}_e$ and plastic (inelastic) strain $\boldsymbol{\varepsilon}_{pl}$ [11, 12]:

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}_e + \boldsymbol{\varepsilon}_{pl} \quad (1.1)$$

In the case of crystal lattice strains, the elastic part $\boldsymbol{\varepsilon}_e$ can be thought of in terms of the expansion and distortion of the Bravais lattice unit cell of the crystal at given orientation. The change of size and shape within the crystal can be assessed in terms of the externally applied

load in the elastic regime or as a result of lattice defects within the material each carrying an amount of *inelastic deformation*. The corresponding *plastic strain* ϵ_{pl} in crystals is related to dislocations of either statistically stored character, or of the geometrically necessary kind that accommodate significant deformation gradients [13]. However, inelastic strains may also be associated with phase transformations, the formation of new grains and boundaries, the presence of defects, vacancies, voids and cracks, the effects of thermal expansion, inhomogeneities, temperature gradients, etc.

Local changes in the internal structure are key parameters of mechanical behaviour in materials is elastic strain, which can be classified into *residual elastic strain* and *applied elastic strain* [11]. The latter can be separated into *static* and *dynamic* depending on the time gradient that the loading is applied.

Residual stresses arise as result of a variety of manufacturing processes: rolling, casting, quenching, hot or cold working, shot-peening, solidification during joining and bonding, coating deposition, etc. Upon removal of all external loading sources or heat gradients, residual stress fields allow the material to accommodate non-uniformly distributed inelastic deformation as a consequence of plastic deformation, phase changes, local deformation gradients, etc. [14]. Residual stress or strain fields arise and can be modified at every stage of the engineering component life cycle, from the raw material production to the in-service operation. It is well established that they should be taken into consideration in the design process and the performance assessment of any engineering component, since it is practically impossible to manufacture any component which is completely free of residual stresses. The aim is not only to predict and examine the magnitude of such stresses, but to incorporate them into the design and manufacturing process in order to derive benefits from their existence. For example, introducing residual stress distributions in components may be beneficial when they are specially engineered for particular applications, e.g. compressive residual stresses in surface coatings impede crack initiation and propagation and improve fatigue resistance. Alternatively,

residual stresses (particularly tensile) may lead to various deleterious effects, including sample distortion, film detachment and modifications of the performance of devices in nano/microelectronics or nano/microelectromechanical systems. An approach towards correlating various manufacturing processing conditions directly with the stress distributions that they generate can therefore be attempted.

Applied loads are caused by stresses, deformation and thermal gradients that are to be present during in service operation within the lifetime of the component. Those are evaluated and calculated, according to the structural requirements, at the stage of design. Additional consideration at the product development stage is also given to aspects of the operating environment (acidity, corrosion, temperatures), material availability (cost vs. properties), characteristics (packaging in confined assemblies) and restrictions (hygiene, toxic hazards, etc.). A systematic approach towards estimating the magnitude and direction of the applicable forces is of paramount importance to the reliability and overall integrity of the final component. For example, a cylindrical shaft supporting a monotonically applied force along its axis will be designed in order to withstand the maximum amount of load before buckling. On the other hand a shaft subjected to torque experiences significant shear stress at the outer periphery of the cross-section, and therefore a hollow cylinder shaft design is preferred.

Another important factor is related to the frequency of the applied loads which can cause material failure in the elastic regime after a period of cyclic loading. The characteristic *fatigue* behaviour of materials refers to the progressive localised damage that may occur when the material is subjected to cyclic loading. As a result material failure may take place at stresses significantly inferior to the nominal ultimate tensile stress limit. The effects of cyclic loading can be discriminated into *high-cycle fatigue*, when the component requires more than 10^4 cycles to failure at low deformation increments in the elastic regime, and *low-cycle fatigue* where the stress is enough to cause increments of plastic deformation in each cycle. Because of its nature and its detrimental effects to structures that undergo loads below yielding, high-cycle fatigue is often more relevant to the assessment and prediction of the longevity of components in service.

In summary, *a priori* knowledge and assessment of the inherent residual and applied stress profiles and frequency within components is vital for the integrity of structures during manufacturing stages and in service operation.

1.1. Historical sketch of engineering and diffraction

Leonardo da Vinci (1452-1519) sketched in his notebooks the possible test for determining the tensile strength of a wire. Later on Galileo Galilei (1564-1642) published the *Two New Sciences*, seeking systematic explanations and adopting a scientific approach to practical problems. This landmark publication is regarded by many engineering historians as the foundation of structural analysis. It was soon after that, when the work of Robert Hooke was published in 1678, suggested that the displacement under load was proportional to the force. James Bernoulli (1654-1705) observed that the correct way to describe the magnitude of the load carried by a solid is to calculate the force transmitted across the unit area. The linear relationship between stress σ and ε strain was finally given by Leonard Euler (1707-1783), who amongst his other contributions introduced the notation $\sigma = E\varepsilon$, where the coefficient E was later termed Young's modulus, after Thomas Young had a similar idea in 1807.

The serious breakthrough of mechanical engineering occurred with the introduction of the steam engine in the early 19th century by James Watt (1736-1819), who based his ideas on the original designs of Thomas Savery (1650-1715) and Thomas Newcomen (1663-1729). The advent of the steam engine is widely acknowledged to have been the major driving force behind the industrial revolution, since it changed fundamentally both the manufacturing methods and transport. At this stage in the history of technological development, both engineering design and material selection were based on empirical observations on the materials properties and macroscopic mechanical behaviour. Many of the more intricate aspects of the material behaviour that remained outside the realm of understanding were the response to cyclic loading, local deformation, the conditions for fracture initiation, etc.

From the 1960s, the descriptions of material behaviour began taking into account the effect of microstructural aspects and characteristics. These developments were driven by the increasing needs of the engineering industry for more reliable, cost-effective, stronger components, through the better understanding of the component and material deformation behaviour. The use of *Factors of Safety* (FoS) also known as *Safety Factors* (SF), provides a design margin over the theoretical design capacity to allow for uncertainty in the design process. The uncertainty is related to every stage of the design process including load calculations, material strength, in-service environment, manufacturing quality. Whereas larger SFs increase the safety and reliability of components, they also significantly increase the dimensions, volume and weight. Detailed microstructural component analysis, alongside with the transition to complete FE simulation environments allowed a significant reduction of safety factors [BS 2573-1 (1983)] without compromising the safety and reliability of the final product assembly. Soon after the first computers appeared, novel simulation tools were introduced in the 1970s with the development of FE methods, the modelling and simulation of the response of complex machine elements became possible.

Prior to that, in the year 1912, Max von Laue came up with the idea to send a beam of X-rays through a copper sulphate crystal and record the results on a photographic plate. He performed the tests with the help of practical experimentalists Walter Friedrich and Paul Knipping, colleagues of Wilhelm Conrad Roentgen who discovered the penetrating power of X-rays 17 years earlier. Although physicists suspected that X-rays were a form of electromagnetic radiation, solid evidence of the wave nature remained lacking and needed to be found. Around the same time the interaction of visible light with regular lattices or gratings was being actively studied. Von Laue drew his idea from a discussion with P.P Ewald, during which the idea arose that the interplanar lattice spacings inside crystals may be suitable for causing interference of X-rays of suitable wavelength.

Within a year of Von Laue's discovery of X-ray diffraction, W.L. Bragg proposed a simple and elegant expression for the analysis of the relationship between crystal structure and X-ray beam energy and direction:

$$n\lambda = 2d_{hkl} \sin \theta \quad (1.2)$$

Here λ is the wavelength, n is the order of reflection, d_{hkl} is the interplanar lattice spacing, and θ is the angle of incidence and reflection between the beam and the scattering planes.

The continuous development of particle accelerators together with the evolution in detector technology and purpose-made experimental setups, strain measurement research has reached unprecedented levels of speed, spatial resolution and accuracy, using diffraction and absorption of penetrating radiation, such as neutrons or high-energy X-rays [15]. Parallel to the X-ray Synchrotrons has been the development of neutron beam sources for Engineering stress measurements. The experimental use of neutron beam sources was first established in the 1980's at the Harwell Campus [16] and subsequently at Institute Laue Langevin (ILL) in Grenoble, FR. The first experimental setups used triple-axis spectrometers and two-axis powder diffractometers. Up to date high-energy spallation sources, such as ISIS at Harwell Campus, UK, are using high frequency ($\sim 50\text{Hz}$) protons, accelerated at high energies hitting a target material, and prompting the emission of neutrons beams ($10^{17}\text{n/cm}^2\text{s}$). Alternatively nuclear reactor-based sources, such as ILL, make use of the reaction of ^{235}U to produce an intense flux of neutrons at the heart of the reactor ($10^{15}\text{n/cm}^2\text{s}$), that are then driven through several heat exchanging processes and moderators to beamline stations.

1.2. X-ray light to condensed matter interaction mechanisms

The discussion of X-ray techniques of material characterisation begins with establishing the basic principles of the interaction of X-rays with condensed matter. Familiar examples of relevant phenomena are most often met in the visible region of electromagnetic radiation (visible light) such as refraction and reflection. The speed of light at different mediums is

causing visible light to change direction when crossing the boundary between air and the sea surface, due to the relative change in refractive index. Furthermore, a travelling beam of light impinging on a surface of a transparent material with a lower refractive index will be almost completely reflected, below a certain incidence angle. Visible light spans only a very narrow energy bandwidth from 400nm to 700nm in wavelength (~ 1.75 to 3.2eV) in the spectrum of electromagnetic radiation. However, penetrative high energy photons (i.e. electromagnetic radiation) have the ability to penetrate deeper into condensed matter and therefore interact under various mechanisms that are illustrated on Figure 1.2.

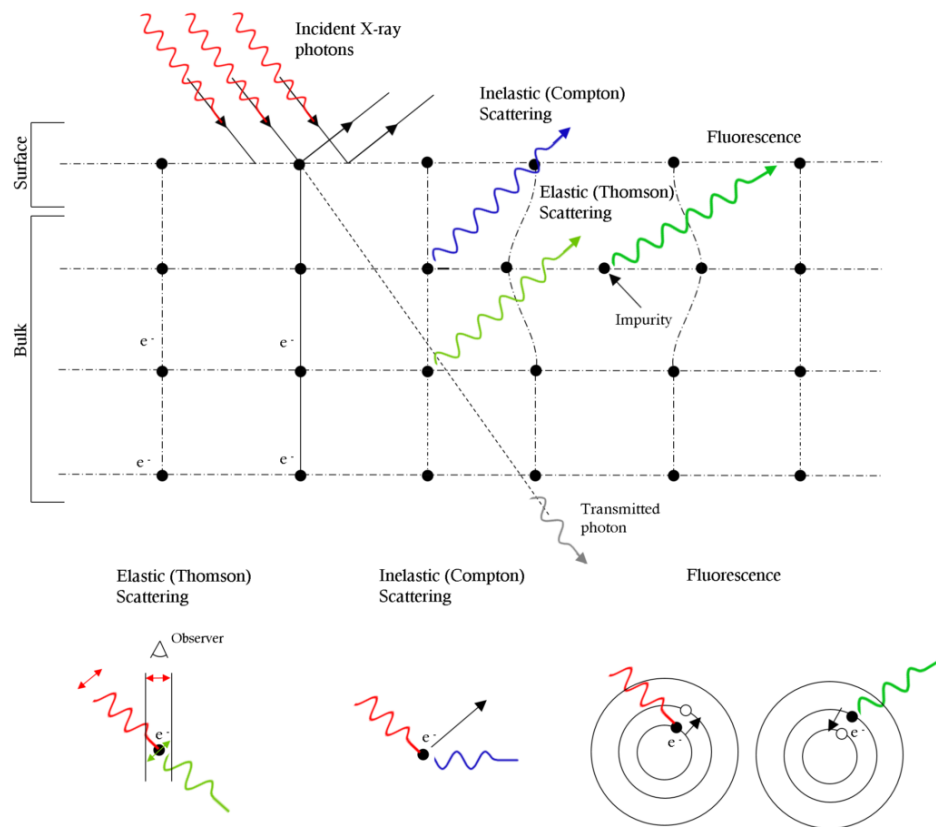


Figure 1.2: Schematic illustration of the interaction mechanisms between high energy penetrating radiation photons with condensed matter.

The most common interactions between X-rays and atoms inside a medium is the electrons surrounding the nuclei [1, 17]. *Elastic (Thomson) scattering* occurs when the electromagnetic field interacts with an electron and causes it to accelerate sinusoidally at the exact same frequency and direction, through an elastic collision process (Fig. 1.2). The re-emission of electromagnetic wave at an observation point (OB) can be deduced by the projection of the oscillatory movement at the angle, defined by the incident wave vector and the direction of

observation. The scattered intensity is proportional to the square of the electric field [3] and effectively $\sim \cos^2\theta$. The ability of an electron to scatter X-rays is quantified by the *Thomson scattering length* (alternatively named 'classic' electron radius) given by:

$$r_0 = \frac{e^2}{4\pi\epsilon_0 mc^2} = 2.82 \times 10^{-5} \text{ \AA} \quad (1.3)$$

where $e = 1.6 \times 10^{-19} \text{ C}$ is the charge of the electron, $m = 9.11 \times 10^{-31} \text{ kg}$ is the electron mass, $c = 3 \times 10^8 \text{ m/s}$ speed of light and $\epsilon_0 = 8.85418782 \times 10^{-12} \text{ m}^{-3} \cdot \text{kg}^{-1} \cdot \text{s}^4 \cdot \text{A}^2$ the permittivity of free space. Finally, the scattering amplitude is proportional to the Thomson scattering length and atomic number in neutral atoms.

Another mode of interaction between an electromagnetic wave and an electron is the *inelastic (Compton) scattering* effect (Fig. 1.2), during which, a part of the kinetic energy of the photon is transferred to the electron, thus the scattered photon contains lower energy [18]. The energy loss can be determined via the conservation of total photon energy ($E = h\nu = hc/\lambda$, where $h = 6.626 \times 10^{-34} \text{ J}\cdot\text{s}$ is Planck's constant, ν is frequency in Hz and λ is the photon wavelength in m) and momentum (in the form of $k=2\pi/\lambda$ wavevector) which gives:

$$\frac{h\nu_0}{h\nu} = \frac{k_0}{k} = \frac{\lambda_0}{\lambda} = 1 + \lambda_C(1 - \cos\psi), \quad (1.4)$$

where ψ of the photon scattering with respect to the incidence, and λ_C is the Compton scattering length ($\lambda_C = h / mc = 2.43 \times 10^{-2} \text{ \AA}$).

Whereas elastic and inelastic scattering occurrences result into the emission of X-ray radiation determined by factors such as the energy of the incoming photon or the angle of deflection of the electron other effects are subsequent to the absorption of high energy X-rays or accelerated electrons at the atomic level. *Fluorescence*, as shown in Figure 1.2 is the result of an outer-shell electron, previously ionised by an incoming energy wave, relaxes to fill inner shell vacancy closer to the nuclei by the emission of a characteristic wavelength radiation. This also causes the incoming wave (or electron) to decelerate according to the amount of energy that has been absorbed by the electron changing its orbital position around the nuclei. The inter-

atomic positions are distinct for every element and the energy difference between the two different elements is well defined, thus allowing the determination of characteristic narrow X-ray emission zones making X-ray fluorescence particularly useful for chemical characterization of materials.

The last dominant interaction between electromagnetic wave and condensed matter interaction is demonstrated by the mass attenuation coefficient μ and is defined as the reciprocal of thickness z of the material which can reduce the intensity of an electromagnetic beam by a factor of $1/e$ [1]. A narrow beam of photons at a specific frequency and an I_0 intensity, penetrating a layer of material with mass thickness z and density ρ , emerges with intensity I given by the Lambert-Beer [19] exponential decay law:

$$I / I_0 = e^{-(\mu/\rho)z} \quad (1.5)$$

where the total mass attenuation coefficient μ contains contributions from photoelectric absorption (μ_{abs}) and from the coherent (μ_{coh} - Thomson) and the incoherent (μ_{incoh} - Compton) scattering phenomena:

$$\mu = \mu_{abs} + \mu_{coh} + \mu_{incoh} \quad (1.6)$$

Equation (1.5) can therefore be rewritten in the form:

$$\mu / \rho = \frac{\ln(I_0 / I)}{z} \quad (1.7)$$

A convenient source for the numerical of μ/ρ values is, for example, the NIST database XCOM [2]. An example of the X-ray photon mass attenuation coefficient μ/ρ is given in Figure 1.3 for copper.

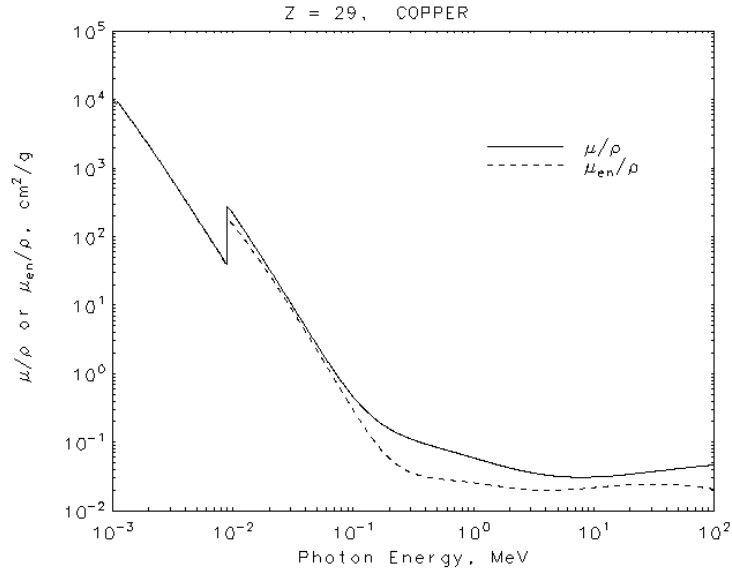


Figure 1.3: X-ray photon mass attenuation coefficient μ/ρ and the mass energy -absorption coefficient against energy (MeV) for Copper ($Z=29$) in logarithmic scale for both x and y axis [2].

It is worth noting that μ is element specific and a function of X-ray energy. It is strongly affected by the atomic number of the element and decreases with increasing photon energy as a function of energy $\sim E^{-4}$. The absorption spectra show sharp increase of μ commonly referred to as *absorption edges*. This phenomenon is related to the X-ray photon energy reaching the minimum energy for ionisation of the atom which manifests itself as sudden increase of absorption in regions on the curve graph.

1.3. X-ray transmission imaging and tomography

An important characterisation method making use of the absorption of penetrating radiation is *X-ray Transmission Imaging and Tomography*. By using detectors to register the transmitted beam through a specimen, information about the structure of the material bulk can be deduced. If the sample dimension is small in the direction of the incident beam, e.g. thin plate, then a single projection may be sufficient to obtain spatially resolved 2-D in-plane mapping. However, if the sample is bulk, a single projection only gives integral information about spatially varying sample properties, but is insufficient for 3-D resolved imaging or mapping.

An approach that offers a way of overcoming this difficulty has been proposed, developed and adopted throughout the 20th century. It has become known by the Greek word “tomography”, meaning “imaging by means of cutting”. Interestingly, historically the first

instance of tomographic imaging in the medical context is indeed associated with an approach that has become known as *serial sectioning*. The story is associated with the name of the Russian surgeon Pirogov who used to walk to his hospital along the banks of the river Moscow, where there were winter market stalls selling frozen joints of meat. Observing the clear sections obtained by the application of sharp knives or axes, he realised that if this operation were to be repeated layer after layer, then after registration (alignment) of the sections a 3-D image would arise. Although this approach could only be applied to cadavers, it had obvious tremendous utility for research and teaching.

Tomography has become one of the most widely used imaging techniques and refers to the cross-sectional imaging of an object from transmission data ("projections") collected by illuminating the object from many different directions [19]. At any given sample rotation angle, the transmission is measured along the path determined by the position of the pixel on the detector, forming a projection. The central idea of tomographic "reconstruction" is that the distribution of absorption coefficient within the entire cross-section can be deduced from the series containing sufficient number of line projections at different angles.

The unique non-destructive nature of these measurement techniques is particularly beneficial in the context of engineering design, since it allows the evaluation of a variety of structural and deformational parameters inside real machine elements, *in situ* and potentially even *in operando*, provided suitable time resolution can be attained. As shown on Figure 1.5 at present significant penetration can be achieved in 3rd generation synchrotron sources, such as the external hutch (EH2) of I12 Joint Engineering Environmental Processes (JEEP) beamline in Diamond Light Source (DLS), UK. It has been demonstrated that the power of the beam is sufficient to penetrate an entire single cylinder engine assembly and perform high-resolution imaging. Furthermore the imaging analysis can be performed in operando even at 2000rpm [20].

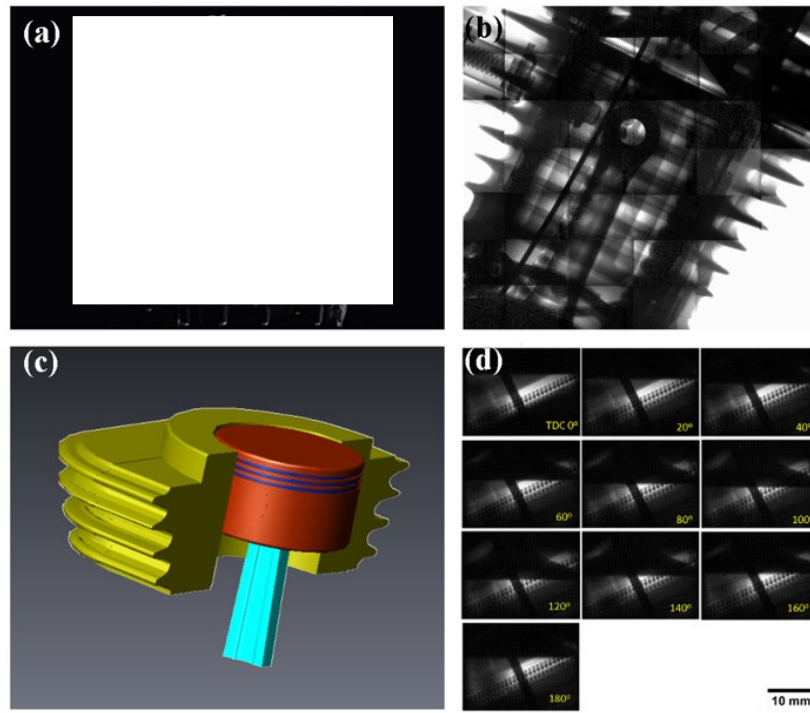


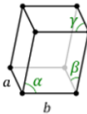
Figure 1.5: *In operando* imaging in internal combustion engine (ICE) components. (a) A final commercial ICE assembly product section [6], (b) Radiographic analysis of a single cylinder ICE engine carried at DLS I12 EH, (c) 3-D CAD representation of the examined volume and (d) *in operando* radiographic imaging of the intake valve opening-closing sequence over 180° [20].

From the mathematical standpoint, the solution to the problem of reconstructing a function from its projections dates back to the paper by Radon in 1917. However, the current urgent interest in tomographic imaging and its further development originated with Hounsfield's invention of the X-ray computed tomographic (CT) scanner for which he received a Nobel prize in 1972. He shared the prize with Allan Cormack who independently discovered some of the algorithms. His invention showed that it is possible to compute high-quality cross-sectional images with an accuracy now reaching one part in a thousand in spite of the fact that the projection data do not strictly satisfy the theoretical criteria underlying the efficiently implementable inversion algorithms.

1.4. Probing material deformation and structure using X-ray light

X-ray diffraction is a technique that allows determining the atomic structure of crystals. The sensitivity of the technique allows minor changes in the lattice formation and interatomic distances to be registered. The relationship between the interplanar lattice spacings and the unit cell lattice parameters for different crystal structures is given in Table 1.1.

Table 1.1: Basic Bravais Lattice structures [17].

Bravais Lattice	Lattice parameter Determination	Crystal Shape
<i>Simple Cubic</i>	$\frac{1}{d_{hkl}^2} = \frac{h^2 + k^2 + l^2}{a^2}$	
<i>Simple Tetragonal</i>	$\frac{1}{d_{hkl}^2} = \frac{h^2 + k^2}{a^2} + \frac{l^2}{c^2}$	
<i>Simple Orthoromphic</i>	$\frac{1}{d_{hkl}^2} = \frac{h^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2}$	
<i>Hexagonal</i>	$\frac{1}{d_{hkl}^2} = \frac{4}{3} \left(\frac{h^2 + hk + k^2}{a^2} \right) + \frac{l^2}{c^2}$	
<i>Simple Monoclinic</i>	$\frac{1}{d_{hkl}^2} = \frac{1}{\sin^2 \beta} \left(\frac{h^2}{a^2} + \frac{k^2 \sin^2 \beta}{a^2} + \frac{l^2}{c^2} - \frac{2hl \cos \beta}{ac} \right)$	 $\alpha \neq 90^\circ, \beta = \gamma = 90^\circ$
<i>Triclinic</i>	$\frac{1}{d_{hkl}^2} = \frac{1}{V^2} \left(\begin{aligned} &h^2 b^2 c^2 \sin^2 \alpha + k^2 a^2 c^2 \sin^2 \beta + l^2 a^2 b^2 \sin^2 \gamma \\ &+ 2hkabc^2 (\cos \alpha \cos \beta - \cos \gamma) \\ &+ 2kla^2 bc (\cos \beta \cos \gamma - \cos \alpha) \\ &+ 2lab^2 c (\cos \gamma \cos \alpha - \cos \beta) \end{aligned} \right)$ where: $V = abc(1 - \cos^2 \alpha - \cos^2 \beta - \cos^2 \gamma + 2 \cos \alpha \cos \beta \cos \gamma)^{1/2}$	 $\alpha, \beta, \gamma \neq 90^\circ$

The apparent elastic variation of the lattice can therefore be deduced as follows:

$$\varepsilon = \frac{a - a_0}{a_0} \quad (1.8)$$

where a is the apparent strained lattice parameter and a_0 is the reference "strain-free" value.

A procedure for determining a_0 is discussed in Chapter 5.2. The processes of lattice strain determination requires the fitting of 1-D diffraction patterns, which represent the amount of constructive interferences for the lattice interaction with the material impinging X-rays formulating peaks in the reciprocal lattice space.

The elegant formulation of Bragg's Law given in Eq. (1.2) provides the criterion for constructive interference between the incident X-ray beam with a highly ordered periodic scatterer, such as crystals that appear on Table 1.1. It can also be re-written in vector formulation with parallel introduction of reciprocal space unit cell in the form of:

$$\mathbf{d}_{hkl}^* = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^* \quad (1.9)$$

The three unit vectors $\mathbf{a}^*$, $\mathbf{b}^*$ and $\mathbf{c}^*$ is a simple geometric transformation of the real space lattice vectors $\mathbf{a}$, $\mathbf{b}$ and $\mathbf{c}$ defined by the direction of their normals according to:

$$\mathbf{a}^* = 2\pi \frac{\mathbf{b} \times \mathbf{c}}{\mathbf{a} \cdot (\mathbf{b} \times \mathbf{c})}, \mathbf{b}^* = 2\pi \frac{\mathbf{a} \times \mathbf{c}}{\mathbf{b} \cdot (\mathbf{a} \times \mathbf{c})} \text{ and } \mathbf{c}^* = 2\pi \frac{\mathbf{a} \times \mathbf{b}}{\mathbf{c} \cdot (\mathbf{a} \times \mathbf{b})} \quad (1.10)$$

By using vector notation $\mathbf{k}_{in}$ is the incident wavevector which ends on 000 on the reciprocal space in Figure 1.4. The construction of Ewald's sphere allows the determination of points within the reciprocal lattice, with respect to the incident beam, where the Bragg law is satisfied and diffraction scattering occurs. For each point that satisfies the Bragg law a vector $\mathbf{k}_{out}$ can be defined. The magnitudes of $\mathbf{k}_{in}$ and $\mathbf{k}_{out}$ are equal to $2\pi/\lambda$. From Figure 1.4 one can deduce that $\mathbf{d}_{hkl}^* = \mathbf{k}_{in} - \mathbf{k}_{out}$ thus. Therefore, the Bragg law can be rewritten:

$$\mathbf{d}_{hkl}^* = \mathbf{k}_{in} - \mathbf{k}_{out} = 2|\mathbf{k}| \sin \theta = \frac{4\pi}{\lambda} \sin \theta \quad (1.11)$$

Throughout the entire thesis the principles, implementation and results of several "hybrid" imaging techniques are discussed. In order to keep the presentation readily accessible, the mathematical formulation details of each method are given in separate appendices at the end.

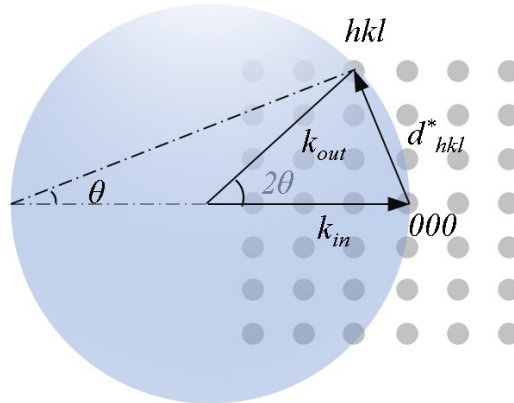


Figure 1.4: Ewald's sphere construction in the reciprocal space.

The insight obtained through the use of several complementary X-ray scanning techniques that we refer to as “hybrid” X-ray imaging here, offers several significant benefits. The combination of information obtained about the internal architecture and the mechanical state of matter brings the experimental data closer to the central theme of materials science and

engineering at large, namely, the elucidation of the structure-property relationships. Particular advantage of the synchrotron X-ray characterisation techniques is their suitability for the development and implementation of *in situ* and *in operando* setups that help understand the components and materials in action. The development of such “hybrid” X-ray imaging techniques goes hand in hand with the establishment of the mathematical framework and practical implementation of “rich” tomographies.

In **Chapter 2**, a bibliographic review of the scientific background and methods used is presented. The specific X-ray and neutron diffraction and imaging techniques that have been used throughout the thesis are described. The methods that were used for complementary validation, are also discussed, such as various microscopy techniques, Focused Ion Beam (FIB) milling with Digital Image Correlation (DIC), Contour Method for Residual Strain evaluation and FE modelling.

In **Chapter 3**, the framework of *in situ* tomographic data acquisition under incremental loading is discussed in the context of synchrotron experimentation. Then the Digital Volume Correlation (DVC) techniques for reconstructing the 3-D deformation fields are described. The DVC technique provides the most direct link between structural and mechanical characterisation, as it relies on the use of a sequence of images collected during load application to extract deformation parameters, such as displacements, and consequently the macro-, meso- and micro-strain fields. It is important to note, however, that DVC interpretation of tomography datasets gives the data for the *total* strain field, and is unable to distinguish between the elastic and plastic parts. Therefore, stress evaluation is not possible using this approach directly. Indirectly, some progress can be made by matching a deformation model to the observations, and using the elastic strain part from the model to assess the stress distribution.

Alternatively, a technique such as diffraction can be used that is sensitive to the elastic crystal lattice strain only.

In **Chapter 4** the implementation of the Diffraction Strain Tomography (DST) method is described. The principles of the method were first introduced by Korsunsky et al. from 2006 [21], whereas several applications, method development and experimental results verification have been the main focus in the present thesis. DST has been used for non-destructive, quantitative reconstruction of internal elastic strain distributions within a cross-section of a loaded [22] or residually stressed object [23]. The results were compared to FE models and the Contour Method respectively. In the second part of the Chapter, the Laue Orientation Tomography (LOT) method is presented. This is another example of a "rich" tomography technique, introduced by Hofmann et al. 2011 [24] that is further developed and implemented to reconstruct relative mis-orientation angles between and *within* individual grains in a polycrystal. The results of LOT are presented and discussed.

In **Chapter 5**, further results pertaining to the development of complex “Hybrid” imaging setups are presented, namely:

Combined use of FIB, X-ray diffraction and imaging for stress relaxation of W-Cu nano-multi-layer coatings used in electronics applications (DIFFABS, SOLEIL, FR)

Nano-scale mapping and inverse eigenstrain modelling of lattice strain and orientation inside carbon core SiC fibres by synchrotron X-ray diffraction and imaging (B16, DLS, UK)

Dynamic stress analysis inside a running internal combustion engine (ICE) using synchrotron X-ray diffraction and imaging (I12, DLS, UK)

The engineering measurement approach applied to different length scales is illustrated using three case studies of *in operando* and *in situ* synchrotron experiments. These span the range from the nano- to the macro-scale in terms of the mechanical and structural spatial resolution. In each case, the development of a bespoke setup is described, and a combination of different techniques (“hybrid”) incorporating imaging and diffraction is presented. Additionally, time-resolved acquisition under dynamic load application is introduced in the case of running internal combustion engine.

In **Chapter 6**, the application of Time-of-Flight Neutron Diffraction Strain Tomography (TOF-NDST) is presented. This is the latest evolution in terms of non-destructive high-resolution reconstruction of non-axisymmetric 2-D strain distributions in the bulk of engineering materials. The results are presented and validated by comparison with the FEM, and the implications of the method are discussed.

Finally, **Chapter 7** presents a critical overview of the thesis results along with discussion of the directions for future work.

Chapter 2

Scientific background and Methods

The present Chapter contains a bibliographic review of the relevant scientific background and methods used in the thesis. The approach covers the physical interaction mechanisms between X-ray electromagnetic radiation and condensed matter together with the application of X-rays in the context of engineering characterisation. X-ray imaging techniques are presented in terms of the mechanisms of radiation interaction with matter, radiography acquisition methods and reconstruction strategies. Specific X-ray and neutron (Bragg-edge) diffraction methods for strain determination in engineering components are discussed. Furthermore, the other techniques used for complementary validation of X-ray results are overviewed, namely, FIB milling with DIC for microscopic residual strain evaluation, the Contour Method for destructive residual strain evaluation by surface profilometry of cut surfaces and numerical modelling, and the inverse FE modelling eigenstrain framework.

2.1. X-ray Imaging Techniques

As explained in Chapter 1, the fundamental interaction of X-rays with matter involves the attenuation of the wave travelling through a non-transparent opaque volume.

In radiography with partially coherent parallel X-rays, both phase and absorption effects play an important role in the formation of a transmission image [25, 26]. Four regions of the imaging distance z can be distinguished on Figure 2.1:

Absorption: The intensity distribution is a pure absorption image.

Near-field: Contrast arises from sharp changes in the refractive index, i.e. at interfaces.

Fresnel region: The image is formed by the combination of refraction and interference, and begins to lose resemblance to the object.

Fraunhofer region: The image intensity is the Fourier transform of the object transmission function.

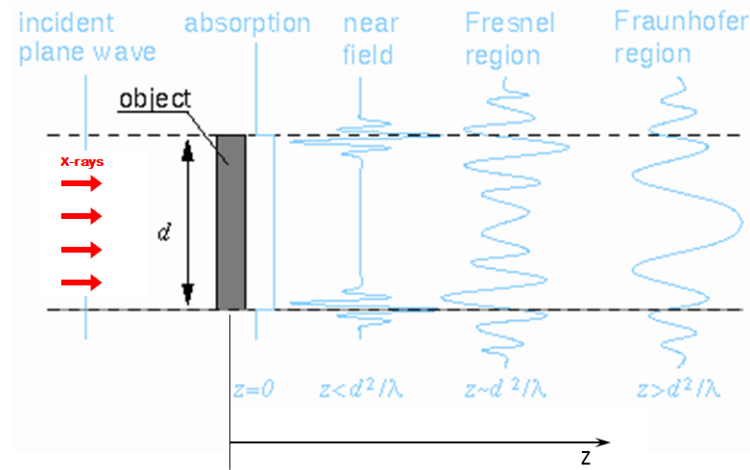


Figure 2.1: Illustration of the beam evolution along $-z$ - direction, after passing through the sample [25].

The calculation of total *absorption integral* of the incident beam $I(z)$ through the sample is:

$$p_{\varphi}(x_{\text{det}}, y_{\text{det}}) = \ln(I / I_0) = \int \mu \rho(x, z) dz \quad (2.1)$$

where $p_{\varphi}(x_{\text{det}}, y_{\text{det}})$ is the registered transmitted intensity corresponding to a particular pixel

$x_{\text{det}}, y_{\text{det}}$ coordinates on the detector at a specific rotation increment φ .

Principles of X-ray Tomography

Two principal experimental modes available to perform conventional (density) tomography are briefly described here. The acquisition strategy with the detector mounted at the closest possible distance to the sample is called *absorption tomography* and is able to discriminate between regions with different mass attenuation coefficient i.e. different materials or varying attenuation path length. The implementation of this technique is straightforward, and works particularly

well when objects contain regions with significant difference in atomic number (i.e. electron density) [19, 27].

The *phase contrast mode* [28, 29] is sensitive to small differences in the refractive index that may be present even for materials with similar attenuation coefficients, where, especially at submicron resolution, the absorption mode fails to produce satisfactory contrast [29, 30]. This effect is particularly prominent at lower X-ray energies, since the phase sensitivity varies proportionally to the fourth power of the wavelength. Increasing the sample - detector distance (propagation length) helps to emphasise the edges and interfaces of the object, and also allows the detection of defects that are smaller than the pixel size, as demonstrated by Cloetens *et al* [31]. At larger distances, scattering of coherent beams produces Fresnel fringes that feature prominently in the image, turning it into a hologram with less direct resemblance of the object [26].

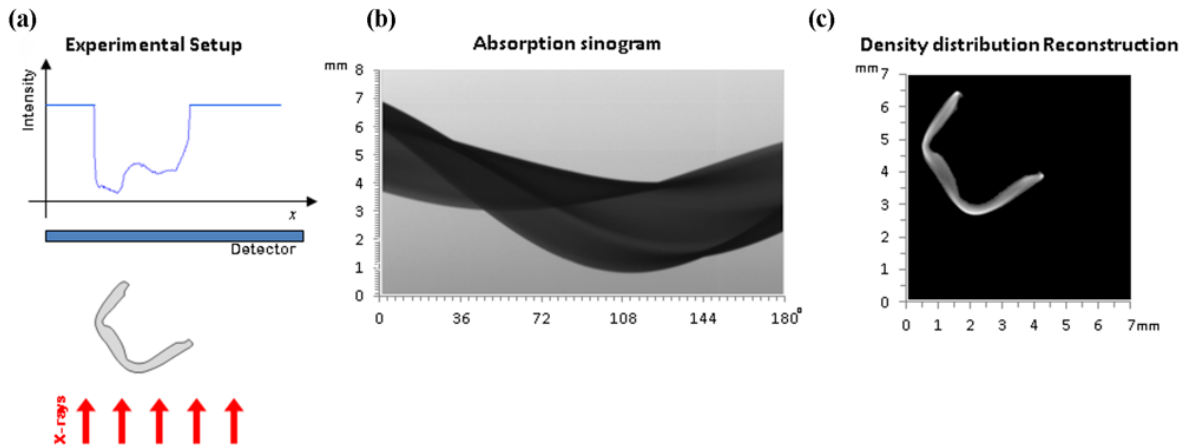


Figure 2.2: Summary of the absorption-contrast computed tomography procedure: (a) an example of the measured intensity profile, (b) the absorption sinogram produced on the basis of the data, and (c) the reconstruction of the density distribution (all data collected in the present project).

The experimental acquisition setup and procedure are shown in Figure 2.2. Starting at a fixed angle (0°), the projections of the 3-D sample onto the 2-D detector are obtained under consecutive rotations of the sample at constant rotation steps around the vertical axis that must be aligned to be parallel to the vertical row of pixels on the detector. Projections labelled $p_\phi(x_{\text{det}}, y_{\text{det}})$ are repeatedly collected over the range of angles from 0° to 180° . Under the conditions of perfect alignment, each horizontal row of pixels repeatedly captures different

angle projection images of one and the same sample slice perpendicular to the rotation axis. The sequence of line profiles obtained in this way can be summarised in a 2-D plot that is called the “sinogram” (Fig. 2.2 (b)). The sinogram provides the input to common inversion algorithms (e.g. the inverse radon transform, and Filtered Back Projection (FBP)) that can solve the inverse problem. The solution of the inverse problem produces the reconstruction shown on Figure 2.2 (c).

The key aspect of the tomographic back projection techniques is the *Fourier Slice Theorem* which relates the measured projection data to the 2-D Fourier transforms of the object cross section [19]. If $f(x, y)$ is a 2-D function, representing the mass attenuation coefficient variation on the sample, then at a given angle φ the projection of $f(x, y)$ onto the x axis is $p_\varphi(x_{\text{det}}, y_{\text{det}})$, where:

$$p_\varphi(x_{\text{det}}, y_{\text{det}}) = \int_{-\infty}^{\infty} f(x, y) dy \quad (2.2)$$

The Fourier transform of $f(x, y)$ is:

$$F(k_x, k_y) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(x, y) e^{-2\pi i(xk_x + yk_y)} dx dy \quad (2.3)$$

where (k_x, k_y) is the new local coordinate system at an angle φ around z axis. For a given “slice” $s(k_x)$, as shown in Figure 2.2 (b) the above simplifies to:

$$s(k_x) = F(k_x, 0) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(x, y) e^{-2\pi i x k_x} dx dy \quad (2.4)$$

But because the phase factor is no longer dependent on y , the integral can be written as:

$$s(k_x) = \int_{-\infty}^{\infty} \left[\int_{-\infty}^{\infty} f(x, y) dy \right] e^{-2\pi i x k_x} dx \quad (2.5)$$

But, because of Eq. (2.2) we get:

$$s(k_x) = \int_{-\infty}^{\infty} p(x, \varphi) e^{-2\pi i x k_x} dx \quad (2.6)$$

where $s(k_x)$ represents the 1-D Fourier Transform (FT) of the projection $p(x, \varphi)$. A MATLAB[®] programme written in this project uses the filtered back projection algorithm to perform the inverse Radon transform. The filter is designed directly in the frequency domain and then multiplied by the Fast Fourier Transform (FFT) of the projections. The projections are zero-padded to a power of 2 before filtering to prevent spatial domain aliasing.

2.2. X-ray and Neutron Diffraction Techniques

In the simplest case of a single-wavelength (monochromatic) incident beam, the irradiated volume sampled by the incident beam contains a large number of small crystallites of different orientations. Due to the high probability that a family of planes may be found in a powder sample consisting of thousands of randomly oriented grains in diffraction condition within the sampling volume, neutron or X-rays scatter into a cone of 2θ semi-angle around the beam direction as its axis. Different sets of $[hkl]$ planes give rise to different diffraction cones with the characteristic 2θ values [32], as shown in the schematic in Figure 2.3.

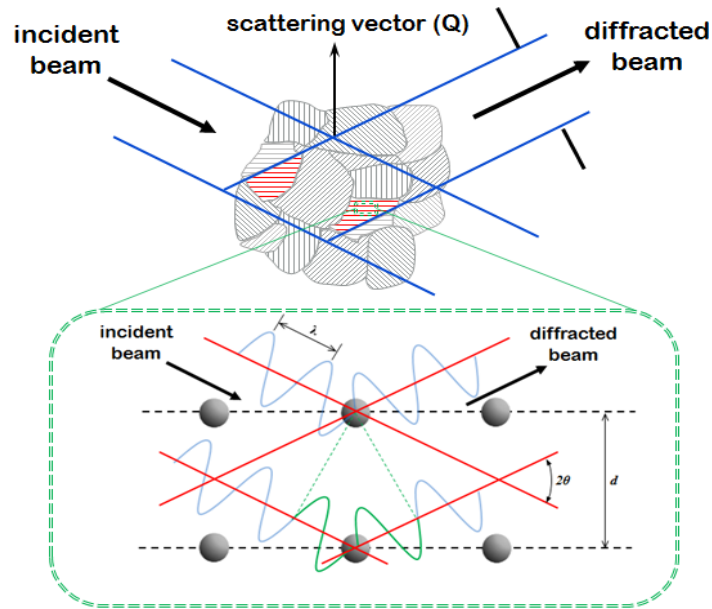


Figure 2.3: Schematic of X-ray Diffraction Condition

The elastic deformation of polycrystalline materials under loading affects the inter-planar spacing within its constituent grains. These grain-set-specific strains cause observable shifts of diffraction peaks. The deformation that occurs in the purely elastic regime is accommodated by

the lattice and therefore the peak shift registered is purely a measure of the elastic strain. It should be noted that plastic strain is accommodated by dislocation glide through crystals. This deformation causes relative shear of the atomic planes, but does not change the average lattice spacing. However, the presence of increasingly dense distributions of dislocations broadens the spread of strains around the average value determined by elastic deformation, and manifests itself in diffraction *peak broadening*.

Monochromatic X-ray powder diffraction

Also commonly known as the angle dispersive method, this setup uses a monochromatic beam (small wavelength bandwidth) to illuminate the sample. The energy definition ($\Delta E/E$) of the beam is typically in the range between 10^{-3} and 10^{-4} . An area detector can be used, mounted perpendicularly to the incoming beam direction, in order to collect the characteristic circular patterns (Debye-Scherrer rings) used for texture and strain analysis. Peak positions, intensities, and shapes can be accurately determined, allowing the evaluation of the lattice strain with the accuracy of at least 100 microstrain (1 microstrain = 10^{-6} strain).

The current trend at state-of-the-art synchrotron radiation facilities is to employ beam focusing arrangements in order to attain submicron spatial resolution. This kind of nanoscale powder diffraction experimental setup (Fig. 2.4 (a)) has been used in Test Beamline B16 at Diamond Light Source, Harwell Science and Innovation Campus, UK. The bending magnet delivers X-ray flux with smooth spectral distribution in the range between 8keV and 25keV. The beam can be passed through a double crystal monochromator, or above it at the fixed offset of 25mm in the white beam mode. Focusing in both modes can be achieved using a Kirkpatrick-Baez (KB) mirror pair. The mirrors are operated at a nominal 3mrad incidence angle and consist of a pair of elliptical pre-shaped silicon substrates coated with 40nm-thick Pt layer. Prior to collecting scattering data in the line scanning or mapping modes, the relative position of the focused beam is determined with respect to the main beam in the focal plane XY that is located 55mm downstream from the edge of the KB mirror chamber.

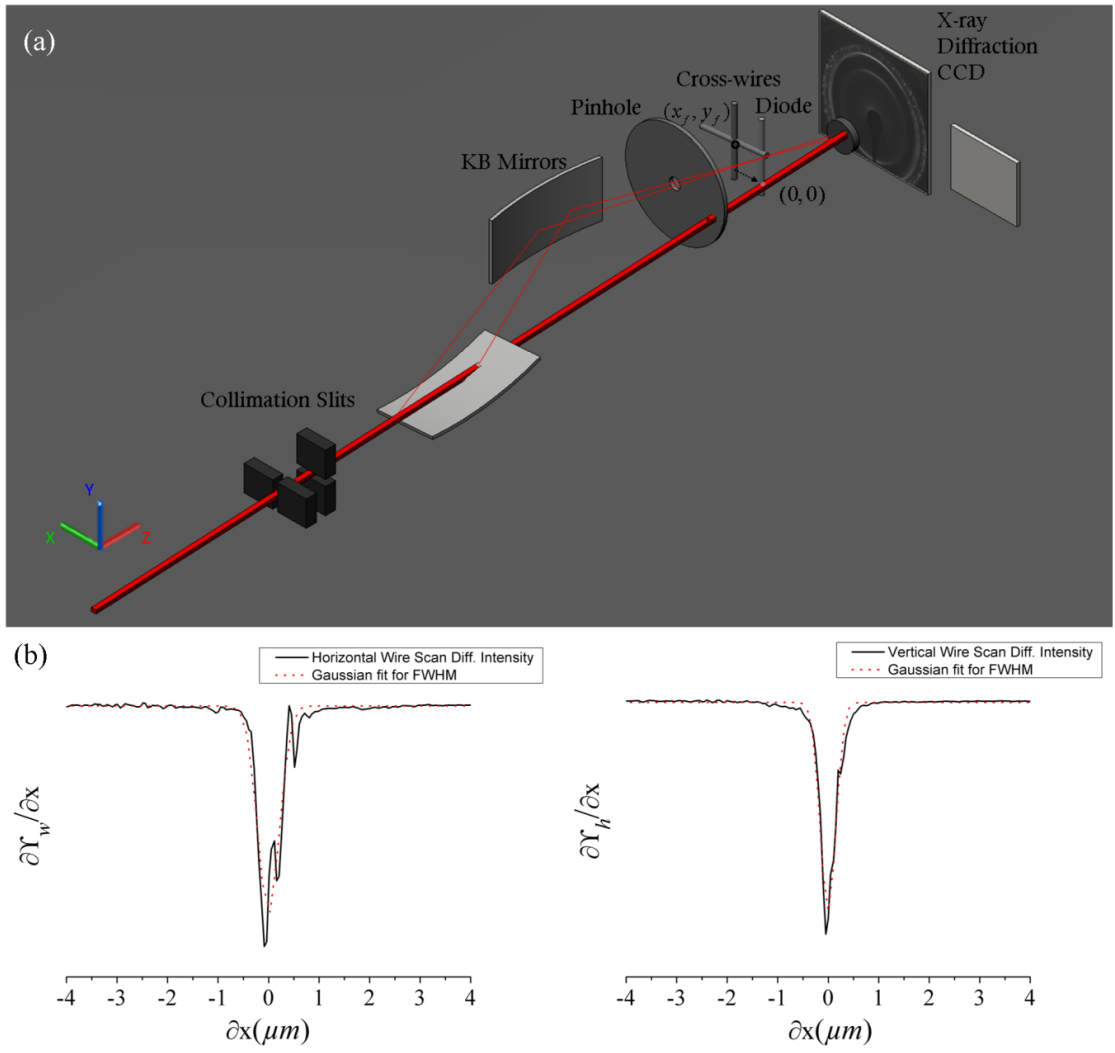


Figure 2.4:(a) Schematic of the nanoscale monochromatic X-ray Diffraction Setup and (b) focused beam wire scan across *width* and *height* to determine the accurate beam shape at the point of convergence.

A set of 200 μm diameter tungsten cross-wires is mounted on the XY translation stage. This arrangement allows precise determination of the beam position (x_{KB}, y_{KB}) with respect to the main beam, as well as its size (Fig. 2.4 (b)). In front of the cross-wires, there is a 150 μm diameter guard pinhole, made in tungsten plate, that lets through the focused beam, whilst blocking the weak remains of the main direct beam and single reflections of the mirrors. Both the cross-wires and the pinhole are placed on magnetic kinematic mounts that allow them to be repeatedly removed and repositioned to better than micron accuracy. The position of the cross-wires along the z axis collinear with the main beam is marked to micron precision by a theodolite. It is used to place the specimen in the correct z position (the focal plane).

Energy Dispersive X-ray Diffraction (EDXRD)

Commonly known as EDXRD (energy dispersive X-ray diffraction), the method works in *reflection* and *transmission* mode depending on the energy flux and the through-depth resolution required. EDXRD provides full X-ray spectra at a fixed diffraction angle. By following a multiple-peak Pawley-type refinement of the recorded diffraction signal, strain accuracies typically down to 10^{-5} can be achieved [33, 34]. In the reflection mode and at lower energies (up to 30keV), large diffraction angles ($\sim 45^\circ$) and high resolution (small “strain gauge volume” dimension) through depth can be achieved.

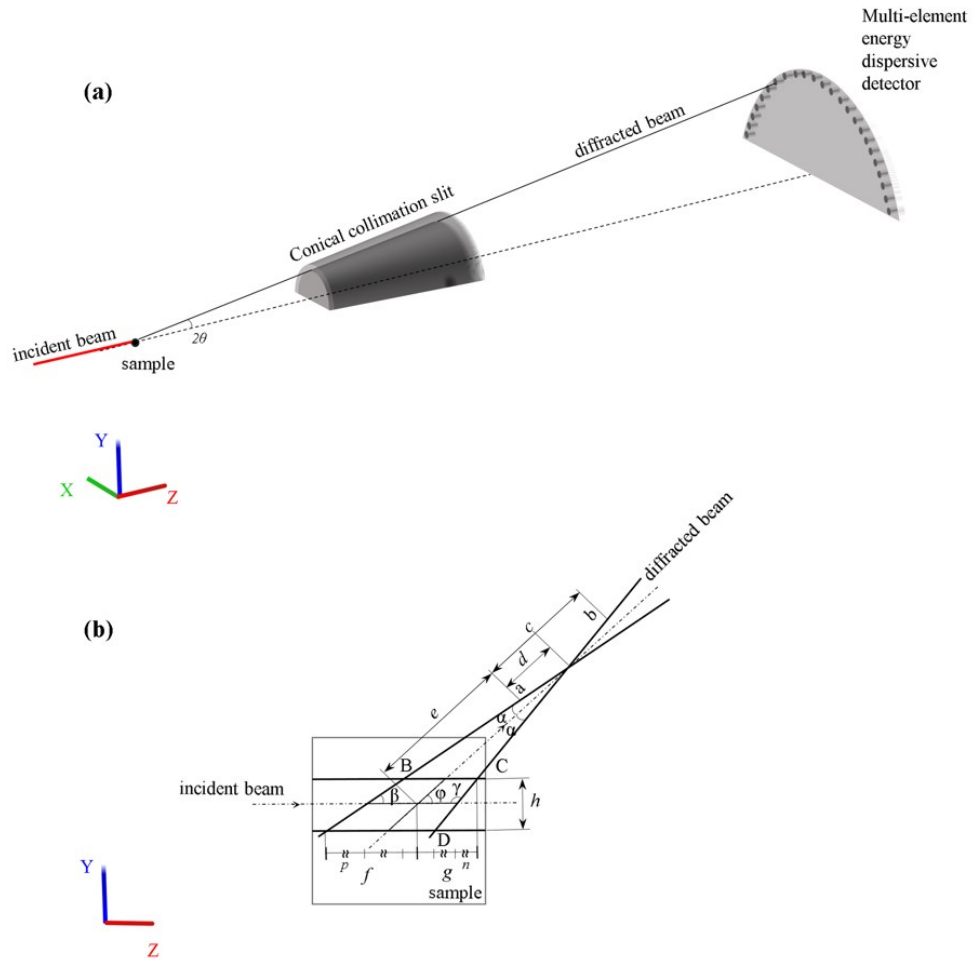


Figure 2.5: (a) White-beam X-ray Diffraction experimental setup in transmission and (b) collimation geometry for parallel beam setup [35].

For the analysis carried in the present thesis, high-energy transmission experimental setup (Fig. 2.5 (a)) has been used on beamline I12, DLS, UK, offering both high spatial resolution and strain sensitivity. For such an arrangement it is important to work at a higher energy spectrum, typically greater than 80keV and up to 300keV. The achieved penetration is sufficient

to probe a highly absorbing steel section up to 10mm in thickness. The experimental arrangement for such an experiment consists of a parallel incident X-ray beam, with a multi-element germanium energy-dispersive detector mounted at a set angle of $2\theta = 5^\circ$ to the incident beam. The incident beam is defined by a pair of perpendicular collimation slits to a rectangular shape normally down to a few tenths of microns in size. Due to the high-energy bandwidth it is typical to use a second pair of collimation slits set to the same centre and slightly smaller overall size, in order to eliminate scattering phenomena from the edges of the upstream collimation slits.

After the interaction of the incident beam with the sample, a secondary collimation procedure for the diffracted beam is followed. For that purpose, a high-precision set of conical slits is placed in between the sample and the detector, allowing only a narrow part of the beam coming from a specific position across the sample to be registered by the detector at the specified diffraction angle. Lastly, another set of collimation slits is placed in front of the detector pixels. The latter set of collimation slits is not shown on the schematic for clarity purposes. The collimation procedure in detail is presented in Figure 2.5 (b) and is what defines the "gauge" volume and therefore the in-depth spatial resolution across the bulk of the sample. The slits are aligned to the centre and perpendicular to the diffracted beam path at $\varphi=2\theta$. Let the acceptance angle of the diffracted beam be given by 2α , where:

$$\tan \alpha = \frac{a+b}{2c} \quad (2.7)$$

a, b: width of individual slits and c: the intermediate distance between them. Following, the angles $\beta = \varphi - \alpha$ and $\gamma = \pi - \varphi - \alpha$ can be calculated in relation to the known α and φ .

The distance, d , from the primary slits to the diffracted beam convergence point is:

$$d = \frac{ac}{a+b} \quad (2.8)$$

The individual lengths of f and g can then be determined as:

$$f = (e + d) \frac{\sin \alpha}{\sin \beta}, \quad (2.9)$$

$$g = (e + d) \frac{\sin \alpha}{\sin \gamma}, \quad (2.10)$$

respectively, where e is the geometrically determined known distance along the diffracted beam from the primary slits to centre of the "gauge" volume.

For a parallel incident beam case shown in Figure 2.5 (b), the lengths of the four sides of the "gauge" volume are given by:

$$\overline{AB} = \frac{h}{\sin \beta} \quad (2.11)$$

$$\overline{BC} = \sin \alpha \left(e + d - \frac{h}{2 \sin \varphi} \right) \left(\frac{1}{\sin \beta} + \frac{1}{\sin \gamma} \right) \quad (2.12)$$

$$\overline{CD} = \frac{h}{\sin \gamma} \quad (2.13)$$

$$\overline{DA} = \sin \alpha \left(e + d + \frac{h}{2 \sin \varphi} \right) \left(\frac{1}{\sin \beta} + \frac{1}{\sin \gamma} \right) \quad (2.14)$$

where h is the beam height determined by incident beam collimation. The distances p, n are:

$$n = \frac{h}{2 \tan \beta}, \quad (2.15)$$

$$p = -\frac{h}{2 \tan \gamma}. \quad (2.16)$$

Therefore, two characteristic magnitudes of the beam "gauge" length can be now calculated as a result of all the above (2.9-10, 2.15-16), which are L_{cent} and $L_{overall}$:

$$L_{cent} = f + n + g + p, \quad (2.17)$$

$$L_{overall} = f - n + g - p. \quad (2.18)$$

The L_{cent} is what is commonly used to determine the effective "gauge" volume and the resulting in-depth resolution of the instrument.

Ultimately, the combination of high flux, full-spectral acquisition and careful beam collimation leads to high in-depth and transverse spatial resolution.

X-ray Micro-Beam Laue Diffraction

When a very small (10-100 μ m dimension) white beam X-ray probe is used in combination with an area detector, the micro-beam Laue diffraction setup is obtained particularly useful for the investigation of crystal structure and orientation within individual grains and polycrystalline aggregates [36]. From the diffraction peak positions, the spatially resolved information about the lattice orientation and unit cell distortion of the crystal can be deduced. Once it is ensured that the beam size is smaller than the crystallites under investigation, Laue patterns are collected from sub-grain scattering volumes, and the variation of the intragranular orientation and elastic strain variation within the crystal can be evaluated.

A simple interpretation of the Laue diffraction condition from the irradiated crystal can be drawn. The crystal shall be considered to be a 3-D periodic arrangement of atoms into rows with adjacent atoms spaced by a along the x -axis, b along the y -axis and c along the z -axis. In the simplest 1-D case illustrated on Figure 2.6 (a) the condition for constructive interference of incident waves can be reduced to the consideration of the path differences (AB+CA) between waves scattered from adjacent atoms in the row along the x -axis:

$$(AB + CA) = a(\cos \alpha_0 + \cos \alpha_n) = n_x \lambda \quad (2.19)$$

where α_0 and α_n are the angles between the incident and diffracted beams to the x -axis, and n_x is an integer which determines the order of diffraction (Fig. 2.6 (b)). This expression is known as the first Laue equation and can be re-written in vector notation, in all directions as:

$$a(\cos \alpha_0 + \cos \alpha_n) = \mathbf{a} \cdot (\mathbf{S}_0 + \mathbf{S}_n) = n_x \lambda \quad (2.20)$$

$$b(\cos \beta_0 + \cos \beta_n) = \mathbf{b} \cdot (\mathbf{S}_0 + \mathbf{S}_n) = n_y \lambda \quad (2.21)$$

$$c(\cos \gamma_0 + \cos \gamma_n) = \mathbf{c} \cdot (\mathbf{S}_0 + \mathbf{S}_n) = n_z \lambda \quad (2.22)$$

where $\beta_0, \beta_n, \gamma_0, \gamma_n$ are the angles between the incident and diffracted beams to the y -axis and z -axis, and n_y, n_z are the respective integers prescribing the order of diffraction along different axes. For the constructive interference to occur in 3-D space, Eqs. (2.20-2.22) must be fulfilled simultaneously. This is a generalisation of what is presented for x -axis on Figure 2.6 (b) according to which the diffracted beam will only occur at the intersection of the diffracting cones along x, y and z axis respectively. Each diffracted beam, can be identified and represented by the three integers n_x, n_y and n_z which represent the order of diffraction. Alternatively, these integers can be simply labelled as h, k, l Miller indices that identify the reflecting planes in the crystal.

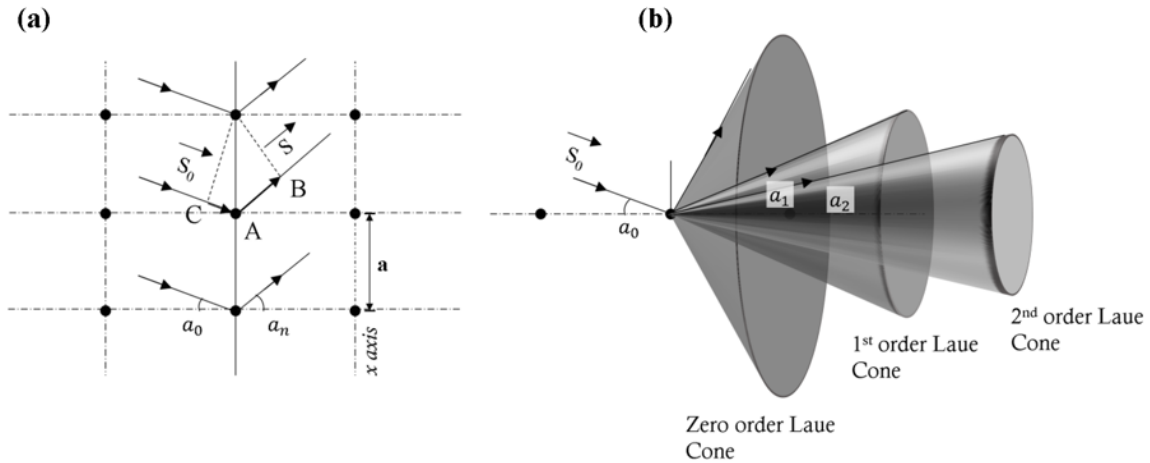


Figure 2.6: (a) Diffraction from a row of atoms arranged along the x axis. The diffraction condition can be determined by calculating the path difference between incident and diffracted beams in vector notation. (b) Schematic of the first three orders of Laue cones at the direction of the diffracted beam from a lattice row along the x -axis.

Micro-beam Laue diffraction has recently evolved into a powerful synchrotron-based diffraction technique. It allows the study of intra-granular variation of lattice orientation, elastic strains, stresses and dislocation microstructure within single crystals or individual crystallites of a polycrystalline material [36]. If a polychromatic X-ray beam is illuminating a sampling volume considerably smaller than the average grain size, then the resulting Laue diffraction pattern consisting of a number of diffraction spots can be recorded on an area detector. The spot indexation reveals the lattice orientation of each illuminated crystallite within the sampling volume. It is worth noting that the Laue diffraction pattern essentially contains information

about the relative angular disposition of the sets of atomic planes that diffract with respect to the incident beam and to each other. If the lattice undergoes purely hydrostatic expansion or contraction, these relationships do not change, and therefore this component of lattice strain is not detectable by pure Laue setup.

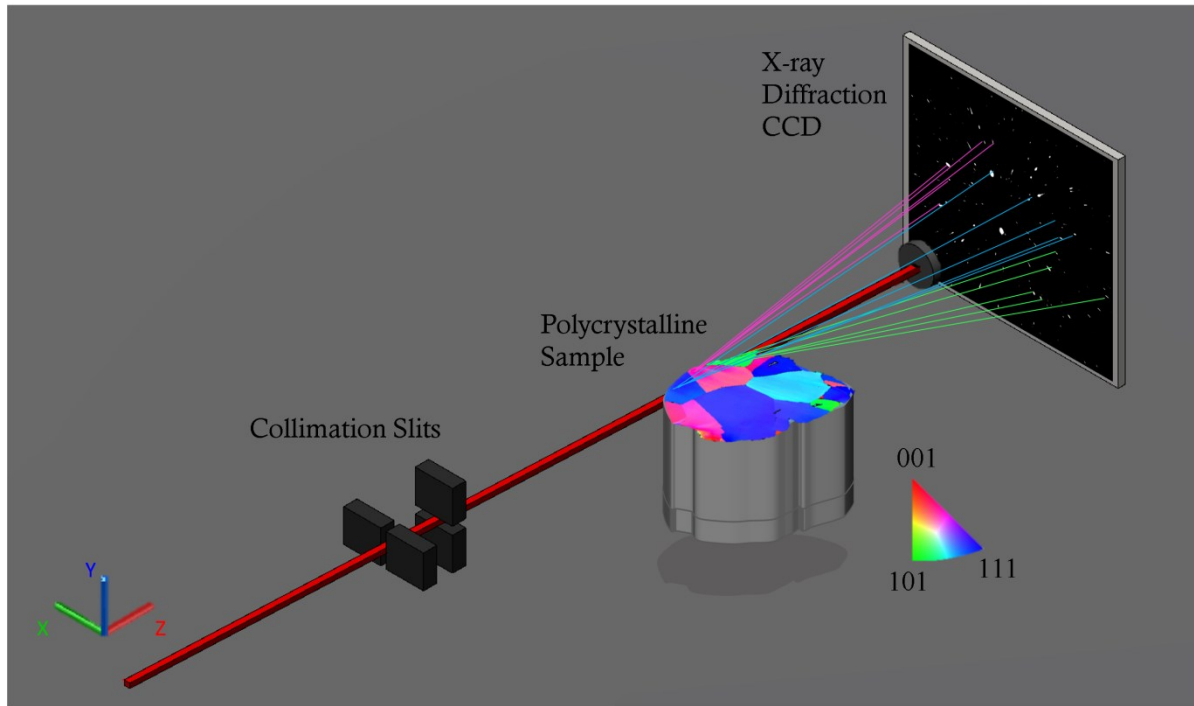


Figure 2.7: A micro-beam Laue transmission experimental schematic.

The X-ray micro-beam Laue setup is illustrated in Figure 2.7. A high-energy transmission micro-beam Laue diffraction setup has been implemented e.g. at beamline ID15A at ESRF, Grenoble [37]. In the mapping mode the sample is moved transversely to the collimated beam direction. Different Laue spots are recorded on the CCD area detector. By accurate calibration, individual grain orientations can be successfully indexed and distinguished. In Chapter 4 we are presenting Laue high energy transmission setup in a tomographic acquisition environment allowing us to reconstruct the shape of grains in a non-destructive 3-D approach.

Differential Aperture X-ray Microscopy (DAXM)

A competing method able to extract depth-resolved information within polycrystalline materials has been termed the Differential Aperture X-ray Microscopy (DAXM) [38-40]. In bulk samples where the incident beam illuminates several grains, the positions of individual

grains within the specimen cannot be found. In order to define the gauge volume, a tungsten wire is scanned between the detector and the sample. At each beam scanning position on the sample, a separate wire x, y axis scan is taking place. The exact depth from which a given reflection originates can be computed through triangulation of the Laue spot position on the detector, the grain and the wire-edge. The main deficiency of the method is mainly that it is too slow for scanning purposes, the difficulty of the experimental setup, and complexity of the data interpretation.

Diffraction Contrast Tomography–3DXRD

Another alternative method to micro-beam diffraction for the study of microstructure in three dimensions has been developed in the form of 3DXRD [41] and more recently DCT [42]. It uses full-field sample illumination and a tomographic acquisition strategy. A monochromatic “sheet” beam of the energy around 50-80keV is used to illuminate the specimen under investigation at different rotation increments (0° - 360°) around the axis perpendicular to the incident beam. A set of two area detectors is used for the acquisition. The transmission detector is mounted downstream of the sample in the field of view of the incident beam. The diffraction detector is mounted at a fixed offset angle with respect to the incident beam in the horizontal plane. As individual grains come into the Bragg scattering condition, a diffraction “projection” of the grain-specific diffracted intensity is harvested at the diffraction detector, whilst an extinction spot, i.e. inverted intensity “projection” appears in the transmission detector. The triangulation of the two projections gives the position of the grain within the bulk. In the course of a full 360° rotation due to the symmetry properties of the lattice there are several rotation increments at which the Bragg law is satisfied and therefore an equal number of projections will be collected. The DCT data acquisition rate is significantly greater than DAXM. On the other hand, very few projections normally occur, and those that do also often contain serious aberrations due to the lattice orientation gradients within the grains. Even though the position of individual grain centres can be determined relatively accurately, the tomographic

reconstruction has to be obtained using space-filling algorithms to locate the boundaries between adjacent grains.

Neutron Time-of-Flight Bragg Edge Diffraction

A technique for strain evaluation using neutron beams has recently been developed that has become known as transmission Bragg-edge neutron imaging [43-45]. The approach utilises the clearly observed step change in the transmitted intensity of the neutron beam at the wavelength corresponding to back-scattering, i.e. the limiting condition for the satisfaction of Bragg's law at the scattering angle of 180° . By registering the neutron beam spectrum using a Time-Of-Flight (TOF) detector, the average elastic strain component in the transmission direction is mapped. By using a pixelated wavelength-resolving area detector at full-field sample illumination [46], this technique allows the simultaneous collection of 2-D maps of the average elastic strain component associated with the incident beam direction, at the spatial resolution equal to the effective pixel size (Fig. 2.7 (a)), for the given sample geometry (Fig. 2.7 (b)).

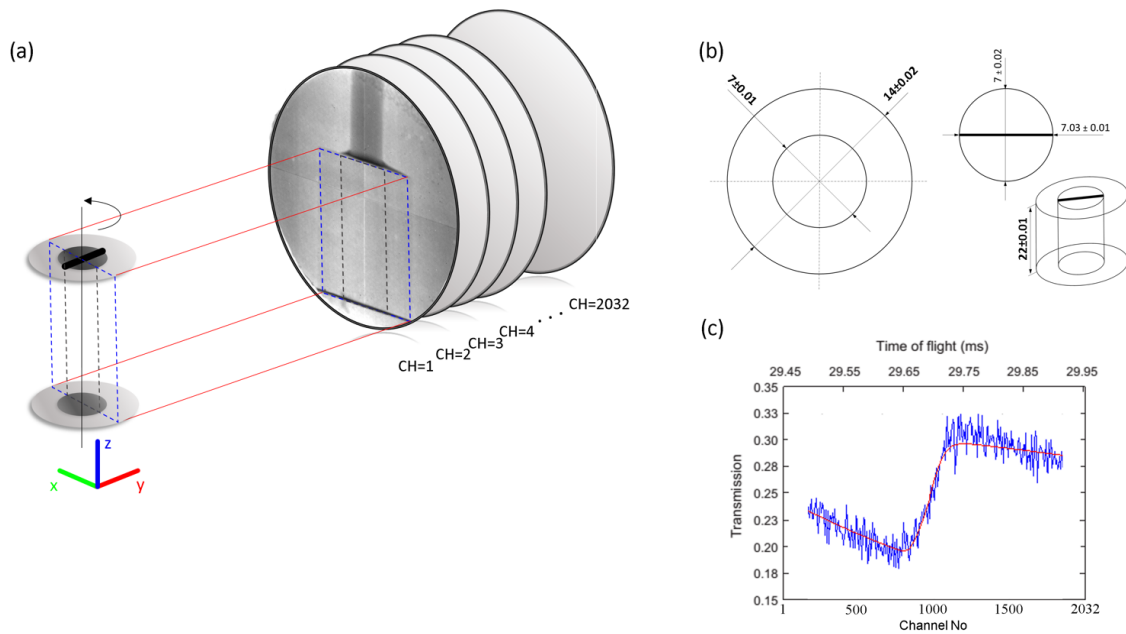


Figure 2.8: (a) Neutron Time-of-Flight transmission imaging setup. The sample outline appears together with the axis of rotation. The different projections are registered by the 2-D micro-channel plated energy "sensitive" detector; (b) schematic representation of the sample dimensions and geometry (c) Example of a typical neutron Bragg-edge referring to [111] peak of Al [45].

At the ISIS Neutron spallation source, Didcot, UK, energy-dispersive neutron beam diffraction experiments can be carried out on the dedicated engineering instrument ENGIN-X

using TOF neutron scattering technique, as illustrated in Figure 2.8 (a). Neutrons produced by spallation pulses that are produced when bunches of protons accelerated in a synchrotron hit a target. The neutrons produced have a speed distribution as they leave the source. Since the neutrons travel at different speeds, they require different TOF's to arrive at the detector. By registering the TOF taken for the neutrons to reach the detector times the h ($6.582 \times 10^{-16} \text{eV}\cdot\text{s}$) Planck's constant, and dividing this by the total flight length L and times the neutron rest mass ($939.565 \text{MeV}/c^2$), the de Broglie wavelength can be worked out as follows:

$$\lambda = \frac{h}{mv} = \frac{h}{mL} \text{TOF} \quad (2.23)$$

where v is the neutron speed (m/s). A characteristic Bragg-edge is shown on Figure 2.8 (c) with the horizontal scale at the bottom representing the channel number of the multi-element detector, and the horizontal scale at the top representing the TOF (ms). For a given value of interplanar lattice spacing d , Bragg equation can be satisfied by radiation with a suitable wavelength λ , provided $\sin\theta$ falls in the allowable range $0 < \sin\theta < 1$. In other words, the wavelengths at which the radiation undergoes reflection into angles ranging from small to the maximum back reflection angle of 180° is given by:

$$2d \leq \lambda \quad (2.24)$$

This is the condition that gives rise to the Bragg-edge phenomenon in the transmitted signal. The central wavelength value of the Bragg-edge $2d = \lambda$ corresponds to the back reflection condition, i.e. the scattering vector being collinear with the incident beam.

2.3. Micro-Mechanical Deformation Measuring Techniques

The accuracy of well-established non-destructive conventional X-ray diffraction techniques, such as 2-D strain mapping require no further validation and their results can safely be declared and produce the input for further modelling and engineering design purposes. In contrast, when a novel experimental technique is introduced, it is important to present to the reader the evidence of the validity of the proposed method and the accuracy of results obtained. Stress

evaluation techniques based on *semi-destructive* or *destructive* principles of various *stress relief methods* used for engineering components and assembly characterisation are briefly presented. The complementary techniques used in the course of the present project in order to verify and confirm the results are also introduced in this section.

Sectioning is a destructive method that involves the attachment of strain gauges and cutting out parts of the specimen (material removal). The strain relief measured as consecutive portions are progressively removed provides the basis for mathematical reconstruction of the prior residual stresses [47]. The *layer removal method* involves observing the deformation caused by the removal of a sequence of layers of material. The method is suited to flat plate and cylindrical specimens where the residual stresses are known to vary with depth from the surface, but to be uniform parallel to the surface.

The *Contour Method*, is a recently developed relaxation method that enables a 2-D residual stress map to be evaluated on a plane of interest. The contour method provides higher spatial resolution, while the sectioning technique is easier to apply since it does not require extensive numerical simulation. The theory of the contour method is based on a variation of Bueckner's elastic superposition principle [48]. The first application of the method was reported in detail in 2001 [49], where the contour method was numerically verified by 2-D FE modelling and experimentally validated on a bent steel beam with a known residual stress distribution. The potential of the contour method was later demonstrated on a 12-pass TIG BS4360 steel weld to measure a complex 2-D stress variation across the weld section [50]. The results were cross-correlated against non-destructive neutron diffraction. A high stress value in excess of the initial yield stress of the material was measured in that case. One of the latest developments of the method involved the determination of complete residual stress tensor in a VPPA weld using a multi-axial contour method implementation [51]. The analysis was performed in the FEM environment and involved the introduction of inelastic misfits, referred to as *eigenstrains*. The eigenstrain is derived from the measured displacements due to residual stress relaxation when the specimen is sectioned. By carrying out multiple cuts, the full residual stress tensor in a

continuously processed body can be determined. The simulation results were directly confirmed by non-destructive neutron diffraction measurements.

The application of the contour method can be separated into four steps:

- Specimen cutting is a critical step in implementing the contour method, as the subsequent procedures of profile measurement, data reduction and stress analysis are all dependent on the quality of the cutting. A wire Electric Discharge Machine (EDM) is used in this step because of its ability to produce a fully strain relieved surface on both sides of the specimen that can be examined together. The required fine surface finish and minimally disruptive cutting (in terms of the non-introduction of additional residual stresses) cannot be achieved by conventional hard cutting tools. Instead, electrical sparks between the workpiece and a conductive wire that is slowly advanced perpendicular to its own extend to create a planar cut. The correct setting of the EDM operating parameters for the cutting procedure is crucial for obtaining suitable section and subsequent data interpretation. Symmetric specimens are also preferred for those reasons, whereas non-symmetric irregular shaped specimens need to be inserted into a soft medium (typically complex shaped components are cast into tin-based moulds). This procedure allows the revolving bearings of the EDM, which circulate the wire, to be as close as possible to the edges of the specimen therefore ensuring a uniform charge application across the cutting section.
- The contour measurement involves measuring the relative height variations at the cut surface that arise as a consequence of the stress redistribution process. A Coordinate Measuring Machine (CMM) can achieve resolutions down to $\sim 4\mu\text{m}$ and offers an effective means of measuring the surface profile through direct contact of a solid hard sphere (sapphire) with the measured surface, or a laser probe performing a similar position measurement function. The CMM can output an array of x, y, z coordinate locations of the surface referring to the 3-D local topography across the specimen. By calibrating the probe to a local system of x, y coordinates defining the plane of cutting the z refers to the height of the tip positions across x and y respective 2-D grid positions on the specimen surface.

- An additional essential step for the contour method is data smoothing of the averaged displacements. This is vital as the averaged displacements consist of deformations resulting from both residual stress release and noise arising from cutting-induced surface roughness and measurement errors. As the noise is not the result of residual stress relaxation, it is important to remove it from the data before proceeding, otherwise it produces an amplified effect during the calculation of residual stresses. The noise is typically removed from the data by fitting the averaged displacements to a cubic spline function.
- FE modelling analysis is performed using the smoothed reduced displacement data from the previous step as input. The variation of local z heights at x and y positions are introduced to FE model resulting into a uniform surface which contains “hills” and “hollows” which represent strain relaxation. Displacements obtained from the cubic spline interpolation of the averaged data are applied to the model as displacement boundary conditions, and linear elastic stress analysis is performed to calculate the stress field normal to the plane of sectioning. The residual elastic strain within the object before cutting is thus extracted.

A further class of destructive techniques can be identified. The so-called semi-destructive methods involve the removal of very small, localised quantities of material to cause and measure the induced strain relief. The *hole-drilling* method [52] involves drilling a small hole at the surface of the specimen and measuring the deformations of the surrounding surface, traditionally using strain gauges, and more recently using full-field optical techniques such as *Moiré interferometry* [53] and *Electronic Speckle Pattern Interferometry* (ESPI) [54].

As the length scale is diminished to the range of a few microns, FIB can be used to make the microscopic cuts that may have the shape of annular rings [55], slots [56] etc. This approach enables the evaluation of the residual stress profiles within coatings. The introduction of traction-free surfaces using FIB milling, and the measurement of the induced strain changes using DIC is a relatively new technique that can be used to determine the residual stress at the micro/nanoscale [57]. The method used in the course of this thesis involves a micro-scale FIB-DIC approach that produces stress relief in a circular “island” of material surrounded by an

annular “trench” at the specimen surface. This technique, also known as the micro-ring-core method, is based on a repeated collection of SEM images of the central core during the milling process in order to quantify the surface strain variation as a function of the cut depth. A value for the average in-plane residual stress in the core region can then be determined by comparison with FE modelling results.

This technique offers a resolution sufficient to quantify the intragranular strains in microstructured materials of practical significance. It is also important to highlight the fact that the existing ring-core interpretation approaches are based on isotropic, continuum mechanics approximations. Thus, in the case of individual elastically anisotropic crystals, only the tensile or compressive nature of the pre-existing stress can still be determined. Further knowledge regarding crystal orientation can allow accurate crystal specific stress quantification. The analysis of the surface strain relief is nevertheless helpful in determining e.g. the influence of grinding and grain orientation on the residual stress state.

The quantitative approach to the FIB-DIC technique data analysis is significantly aided by the development of high precision dual column SEM with integrated FIB available today. This allows improving the insight into the micro-scale residual stresses, and push the boundaries of resolution to submicron levels. The simultaneous FIB-DIC data collection process and data implementation can be divided into several steps that are schematically presented in Figure 2.9.

- Initial testing is performed to refine the milling setup of the FIB/SEM procedure. Several parameters need to adjusted, including the stub diameter, the “trench” width, milling rate and the beam voltage and current. These parameters influence the resolution of the technique and require optimisation to minimise the effect of noise of measurement related to image drift, material re-deposition, and the requirement of focus adjustment during the milling procedure.
- The collected image stack (Fig. 2.9 (b)) is processed by DIC via the application of a local “seeding” process in the region of interest. This is a manual procedure, where the marker

seeding size is adjusted according length scale, and magnitude of deformation with respect to pixel size at consecutive increments (Fig. 2.9 (c)).

- An automated DIC suite implemented in MATLAB[®] is used to perform strain determination in the central “stub” region using an iterative process. Once this step is finished it is possible to deduce a first approximation of the relaxation process (Fig. 2.9 (d1)). There is clear evidence of the presence of outliers at specific pixels where the correlation has not been successful (Fig. 2.9 (d2)).
- The next step is to eliminate outlier points in a systematic manual approach. After the removal a much more reliable DIC tracing emerges (Fig. 2.9 (e1-2)). The results are plotted as a function of the marker displacement versus position. The strain calculation error is deduced by calculating the standard deviation of this fitting process.
- The resulting profile of strain relief as a function of milling depth is shown in Figure 2.9 (e3). It is used to determine the full strain relief $\Delta\epsilon_{\infty}$ at the ring-core surface. The strain relief profile is independent of material properties and can be approximated by a function $f(\Delta\epsilon_{\infty}, z)$ of the cut depth h and the ring-core diameter d such that $z = (h/0.42d)$:

$$f(\Delta\epsilon_{\infty}, z) = 1.12 \cdot \Delta\epsilon_{\infty} \cdot \frac{z}{1+z} \cdot \left(1 + \frac{z}{1+z^2}\right) \quad (2.25)$$

Since the cut depth at a given image number may not be known precisely, the parameter κ_{mill} is used to relate to the milling depth according to $h = I\kappa_{mill}$, where I refers to the image index number and δ_{start} is used as an offset for z to account for initial surface displacement due to its roughness. Therefore, z is now calculated according to:

$$z = \frac{I\kappa_{mill}}{0.42d} - \delta_{start} \quad (2.26)$$

Finally, a least squares fitting approach is used to optimise the parameters $\Delta\epsilon_{\infty}$, κ_{mill} and δ_{start} and to obtain the best fit value for the overall strain relaxation.

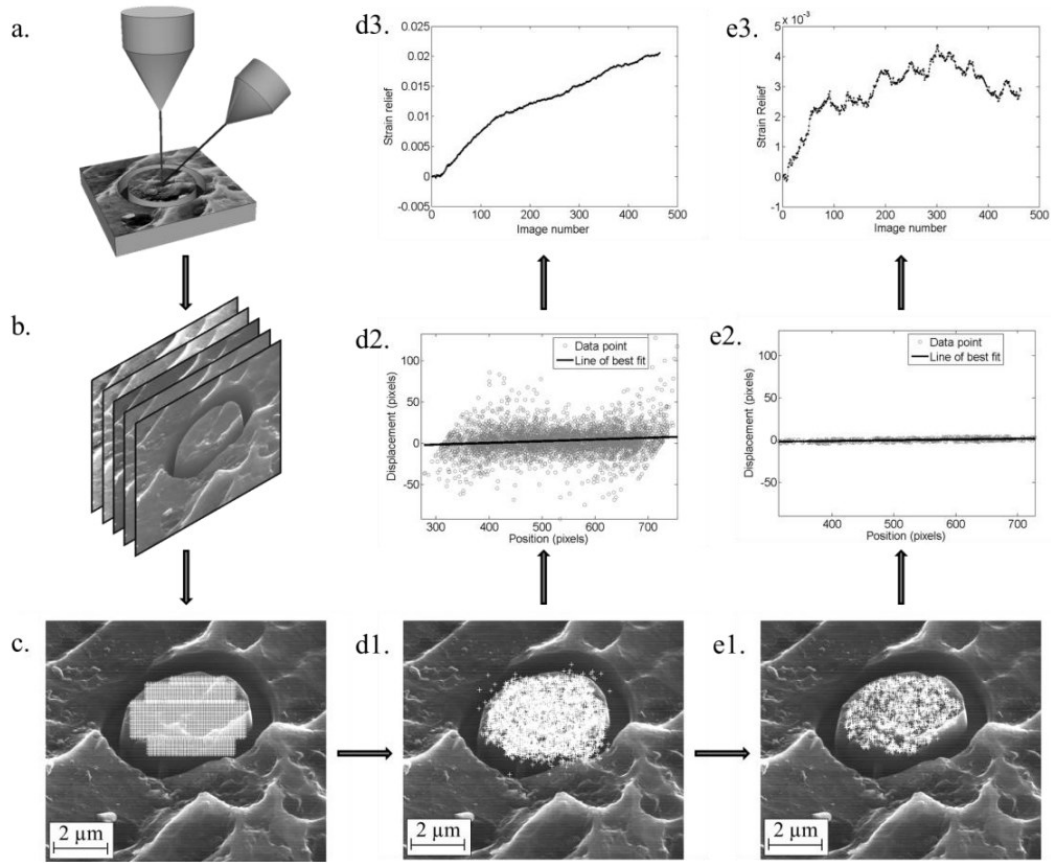


Figure 2.9: Schematic illustration of the FIB-DIC acquisition and implementation procedures. (a) Simultaneous FIB milling (normal to sample surface) and SEM imaging (tilted at 52°). (b) Image collection. (c) Segmentation of the strain relieved surface in appropriate window sizes (marker seeding). (d) Automated DIC showing a large number of aberrations. (e) Consequent manual removal of outliers and retention of well-tracked markers allows clear trends to be revealed in the displacement against pixel plot. The gradient of this plot is used to quantify strain relief as a function of image number [58].

At the scale of the crystal lattice (length scale of Å), observations can be made using *Transmission Electron Microscopy* (TEM). A white beam of electrons is transmitted through an ultra-thin specimen of the thickness in the order of 50-100nm. The interaction of the electrons travelling through the specimen with the lattice has been previously used to determine the inter-planar lattice distances. TEM setup can also be used to generate diffraction patterns similar to X-rays. This means that on a nanoscopic level, and taking into account the sample limitations posed by the TEM setup, structure and orientation of materials can also be investigated at the crystal lattice scale.

A useful review of the qualities and characteristics of all the relevant structural characterisation techniques is summarised in Table 2.1. The techniques mentioned above can be used to map regions of interest in the samples using different sample arrangements and at a variety of length scales. Nevertheless, in the present state-of-the-art this can be effectively done

for 2-D cases. Destructive serial sectioning can provide 3-D information, but the specimen is destroyed in the process. Engineering components are rarely flat, but rather are characterised by a complex shaped volume. For the purposes of characterisation, tensorial quantities such as stress/strain distributions need to be determined within this volume. Hence, alternative approaches need to be developed to achieve 3-D characterisation, either by combining limited information available from surfaces and sections (notably by using mechanically justified, eigenstrain-based rigorous theoretical framework described in the following section), or by achieving nearly isotropic spatial resolution (gauge volume shape) when non-destructive penetrating radiation methods are used.

Table 2.1: Comparison between methods used for evaluating mechanical state of components and their main characteristic features.

	General purpose	Quick test	Quantitative	Low damage	Stress profile	Near surface	Deep interior
Sectioning	X		X		XX	X	XX
Hole-drilling	XX	X	XX		X	XX	
DIC	X		XX		X	X	
Layer removal	X		XX		XX	X	XX
Contour Method	X		XX		XX		XX
FIB	X		X		X	XX	
EDXRD	XX	X	XX	XX	XX	X	X
TEM			XX	XX	X	X	

The motivation for this thesis is to introduce and apply novel concepts for the non-destructive, quantitative and precise determination of structural characteristics within engineering materials and components. This is accomplished by combining principles from conventional imaging X-ray experiments with the structural information contained within X-ray diffraction patterns.

2.4. Eigenstrain Theory

The general residual stress state in a solid is represented by the six components of the stress tensor (σ) at each point. Due to this inherent complexity it is practically impossible to recover each strain component by direct experimental onslaught, particularly given the spatial resolution required to probe a quantity that varies as a function of three spatial coordinates

within the sample. Furthermore, it is important to note that the residual stress distribution must by definition be self-equilibrating. This requirement establishes a relationship between the gradients of different components of the stress tensor ($\nabla \boldsymbol{\sigma} = 0$). The stresses must also satisfy the traction-free boundary conditions ($\boldsymbol{\sigma} \cdot \mathbf{n} = 0$), where $\mathbf{n}$ denotes the surface normal. Enforcing these requirements on an experimentally acquired stress state is extremely challenging, unless a rational analytical approach is adopted that enforces these field and boundary conditions by the appropriate choice of the basis of description.

One such approach was proposed using the concept of eigenstrain and the notation ε^* was first introduced by Mura in 1982 [59], who suggested the word based on the title of the German paper "Eigenspannungen und Eigenspannungsquellen" (Reissner, 1931). "Eigenstrain" is a generic name given to such non-elastic strains as thermal expansion, phase transformation, initial strains and misfit strains. "Eigenstress" is a generic name given to self-equilibrated internal stresses caused by one or several of those eigenstrains in bodies which are free from any external force and surface constrain [59]. The routes of generation of those inherent transformations can be traced during the consecutive industrial manufacturing steps, from the raw material to the final product layout. Raw materials can initially be formed through roller bearings to sheets containing non-uniform structure and contain plastic deformation due to the severe shape change. They routinely undergo thermo-mechanical treatments which affect their internal structure, surface chemical treatments for corrosion protection or through shot peening processes to impose near surface compressive stress distributions. Further, mechanical bonding operations (cf. welding) or cutting pasting operations (cf. the procedure employed in shrink-fitting manufacturing processes) continuously modify the internal structure, and thus the residual stress distributions. It is therefore convenient to introduce a generic term eigenstrain to refer to all such sources collectively. From the viewpoint of residual stress analysis the precise origins of different eigenstrain terms are somewhat unimportant, since only the overall misfit strains govern the residual stress state [60, 61] The concept accounts for all permanent strain

origins which exhibit inelastic behaviour and give rise to residual stresses, but is not merely a sum of the various non-linear strains.

The Concept of Eigenstrain

Before the introduction of the term eigenstrain, the term *nuclei of strain* had been used by Mindlin and Cheng (1950) to refer to a source of residual elastic stress. With the introduction of the *eigenstrain* concept, Mura in 1982 also established a mathematical framework for the determination of residual stress corresponding to a given eigenstrain distribution, for the case of an infinite 3-D body. The process of introducing an eigenstrain distribution into a solid object is illustrated Figure 2.10 (a). The example considers a volume change introduction in the particular area of the FE model which is numerically approximated by the application of a thermal coefficient variation on a specific model node under a uniform temperature change [62]. For clarity, the response of the system is given only for the horizontal elastic component. At the region where eigenstrain is applied the residual stress state shown on the right is uniform and opposite sign with respect to the imposed eigenstrain profile on the left (Eshelby, 1957). The surrounding area represents the influence of eigenstrain to the local surroundings and decays to zero at large distances outside the inclusion.

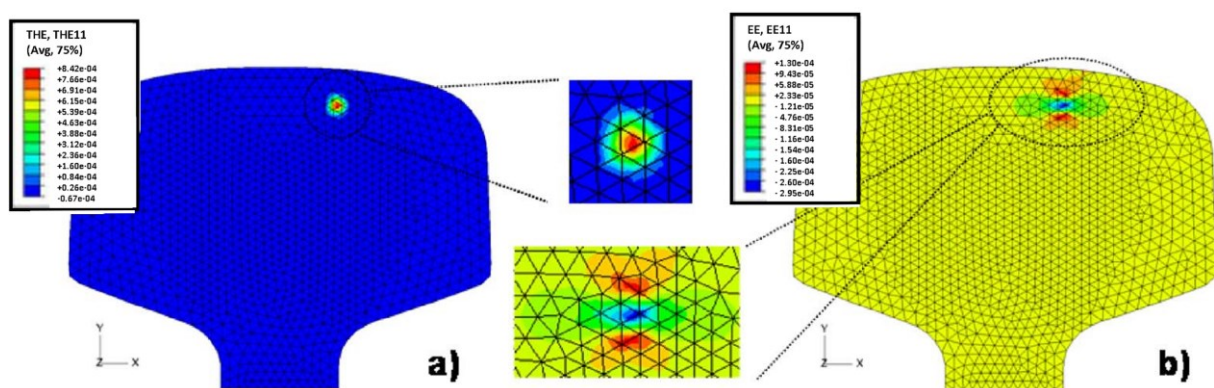


Figure 2.10: (a) Demonstration of an imposed eigenstrain field in the form of a “tent” base function in an FE model. (b) The elastic strain response of the system to a single eigenstrain “tent” base function [62].

In the absence of inelastic (permanent) strain of some origin, or when such inelastic strain is uniform throughout the sample, the body remains stress-free without external loading. For a

known non-uniform eigenstrain distribution $\boldsymbol{\varepsilon}_{mn}^*(x')$, the elastic residual strain $\boldsymbol{e}_{ij}(x)$ (and hence stress) in the body can be found using the following formula [59]:

$$\boldsymbol{e}_{ij} = -\boldsymbol{\varepsilon}_{ij}^*(x) - \int_{-\infty}^{\infty} C_{pqmn} \boldsymbol{\varepsilon}_{mn}^*(x') G_{ipqj}(x - x') dx' \quad (2.27)$$

where C_{pqmn} is the known tensor of elastic constants, and $G_{ipqj}(x - x')$ is the Green's function. The Green's function is specific to the particular geometry, representing the displacement component in the i direction at x when a body force is applied at x' in the p direction in an infinitely extended material. However, there is a practical limitation in that the Green's function is known explicitly for the special geometries such as infinite space, semi-infinite space bounded by a plane, and the corresponding 2-D solution. Consequently, it is practically impossible to seek analytical expressions for the geometries typical of engineering components of complex shape.

A second limitation arises from the fact that the eigenstrain theory needs to be applied inversely for the case of residual stress analysis. That is, given some knowledge of the residual deformation state, the underlying eigenstrain distribution needs to be found. Therefore, the framework for the analytical approach to the *inverse problem* of eigenstrain theory is the quantitative comparison between point-wise experimental data and the predictions of the reconstructed elastic fields at the same locations. The details of the framework and its development in a tomographic acquisition environment in the present investigation are demonstrated in Chapter 6. This forms the basis of the Tomographic Eigenstrain Reconstruction Method (TERM):

The purpose of TERM is to reconstruct the residual strain and stress distributions in a variety of engineering components from limited tomographic experimental data. For residual strain measurements in a tomographic acquisition strategy, the crucial issue that emerges is not the type of measurements being used, but the coupled character of measured strain components in the different projections at different rotation increments. The inverse eigenstrain problem has

been successfully implemented by Abbey et al. [46] for an axisymmetric strain variation on a shrink-fit "ring-and-plug" type of sample. The determination of a uniform single component of eigenstrain requires the acquisition of single projection and the respective inverse ERM. The introduction of non-axisymmetric strain variations requires more angular increments to be taken into account and therefore requires a combined inverse eigenstrain analysis in order to deduce the elastic strain solution.

The ill-posed nature of TERM is also an important aspect of the methodology. The inverse problem of tomographic eigenstrain reconstruction theory makes use of the knowledge of residual elastic strains (or stresses) at a number of measurement points and at different rotation increments to retrieve the underlying source of eigenstrain field. The data quality in terms of the resolution, fitting accuracy, and number of projections has a significant effect on the solution. The acquisition strategy and the inverse problem formulation significantly affect the result and the robustness of the reconstruction. Therefore, a number of constraints need to be imposed to regularise the reconstruction problem that is based on the limited projection data, the reduced fitting accuracy and the nature or lengthscale of the investigated eigenstrain variation.

2.5. Conclusions and remarks

Due to the sheer variety of different experimental techniques it is impossible to cover the full aspects of each individual method in detail in the course of Chapter 2. This would be counterintuitive even to the experienced reader, and therefore would not allow to have an overview of the proposed experimental framework. Therefore, the approach was selected in this section that identifies the principles, background and extent of the existing experimental techniques. The aim of this Chapter was to outline the limitations of the current methodology into the examination of structural and mechanical characteristics of engineering components through an up-to-date literature review of contemporary non-destructive and destructive techniques.

The proposed “hybrid” X-ray imaging and diffraction techniques are introduced and thoroughly investigated in terms of implementation and results in the following Chapters according to their fundamental experimental acquisition routes. The inverse eigenstrain modelling technique is used for looking into the thermo-mechanical processing history of a unidirectional SiC carbon-core fibre (Chapter 5) and in order to reconstruct elastic strain distribution from a tomographic set of neutron ToF angular projections (Chapter 6). The contour method is used to confirm the Diffraction Strain Tomography data acquired on a Ni superalloy Direct Laser Deposition (DLD) fabricated aerospace jet engine blade (Chapter 4). The combined FIB-DIC approach is used in order to confirm the experimental diffraction data at nanoscale spatial distribution within a carbon fibre in Chapter 5. This formulation ensures deeper understanding of the principles and allows the reader an easy overview of the different topics covered in the thesis at a gradually increasing level of complexity.

Chapter 3

Tomography-Digital Volume Correlation

“Hybrid” X-ray tomography is a term often used to indicate the ability to obtain, non-destructively, important information about the internal structure characteristics, local features within materials or components, as well as probing mechanical deformation quantities.

Digital Volume Correlation (DVC) is a state-of-the-art technique that is capable of full field 3-D displacement evaluation. Its ability to extract data pertaining to 3-D internal deformation is particularly useful in understanding the mechanical behaviour of various engineering component [63, 64]. High-resolution X-ray micro-tomography data, acquired at consecutive loading increments during an *in situ* loading experiment, provide typical input for the analysis. In analogy to 2-D DIC, the basic principle of DVC lies in the matching (or tracking) of patterns or features from the reference volume image to the deformed volume image, with the best match determining the relative displacement vector. To achieve 3-D mapping of the displacement field, sub-volumes (typically cubic) on a grid located in the reference volume image are selected [65]. The corresponding volumes in the deformed volume image are identified and a 3-D grid of 3-D displacement vectors is calculated. Although fundamentally simple in principle, the practical implementation of DVC involves significant challenges, including implementation complexity, measurement accuracy and computational efficiency.

The key advantage of the DVC method lies in its suitability to probe displacement fields under severe plastic deformation. A progressive incremental load application in parallel with *in*

situ tomographic acquisition, and subsequent reconstruction of data of consistent quality is sufficient for the DVC correlation of sub-volumes to converge to a solution. The optical correlation strategy is therefore not susceptible to the lattice related challenges for the determination of strain, in the way that X-ray diffraction measurements are dependent on the diffraction peak shape change in order to deduce strain variation. The ability of individual material grains to deform elastically under external load is finite and bounded by the material elastic limit and strength. Once the inelastic deformation mechanisms are activated upon material yielding, diffraction peak broadening phenomena are observed, whilst the peak position remains unchanged. Digital image correlation examines X-ray tomographic imaging datasets relying on good contrast between distinct features within a material cross-section in order to retrieve deformation information. The contrast variation is related to inhomogeneities of absorption coefficient amongst different materials, inclusions, voids, cracks etc., or generated from differences in the refraction index of similarly absorbing material phases.

The flow chart representation of DVC analysis is shown in Figure 3.1. The process starts with the *in situ* acquisition of tomographic data sets under incremental loading and is followed by tomographic data reconstruction as described in Chapter 2. Typically, full-size 16-bit 3-D reconstruction data sets allocate several gigabytes of memory, making the DVC calculation non-feasible. Therefore a data reduction process on the resulting 3-D volume is routinely followed. The reconstructed volume, free of noise and artefacts, can be segmented into sub-volumes in order to reduce data size and investigate local phenomena (i.e. cracks, materials inclusions, etc.). Data reduction can alternatively be achieved by pixel binning which significantly improves calculation speed and statistics. The output can be then used to inform numerical FE models providing a representative volume and end-plane boundary conditions at consecutive loading increments ($N=1...n$). Alternatively, the bulk can be directly meshed with the use of commercial software packages, such as SIMPLEWARE[®]. The response of the model in the form of the full displacement fields can then be compared with the DVC results. The

imposition of representative displacement boundary conditions ensures correct evaluation of the internal deformation field via numerical simulation.

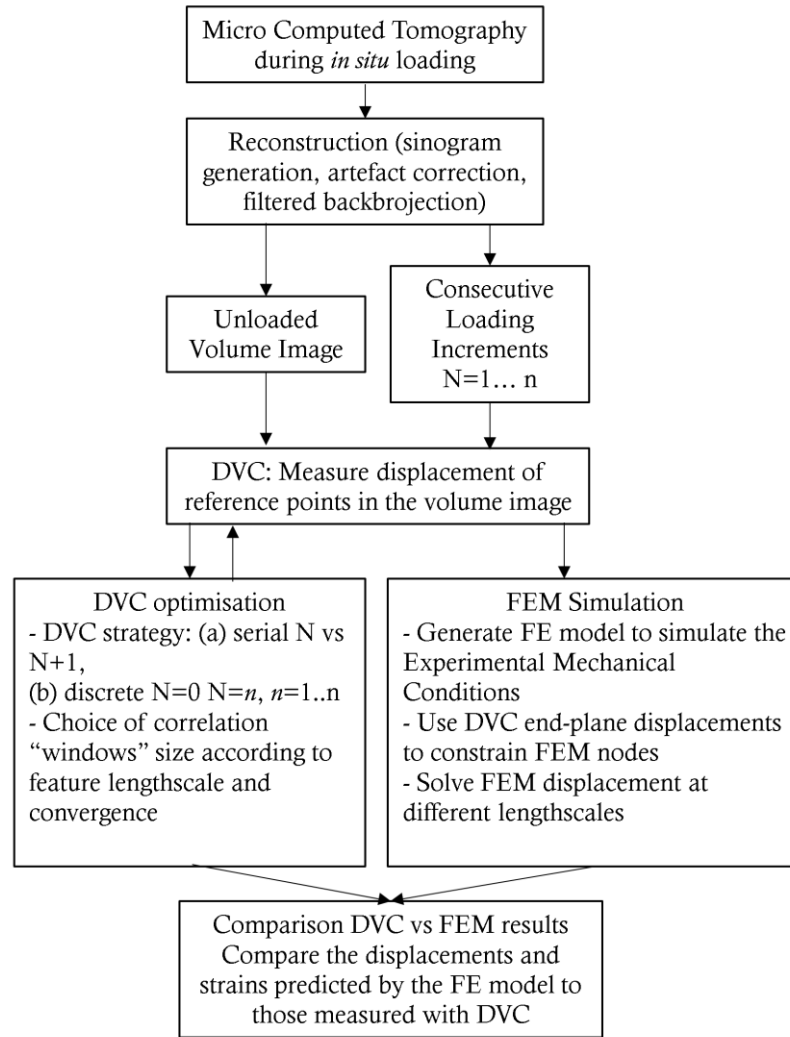


Figure 3.1: Flow chart of DVC strategy from the experimental acquisition to data implementation steps.

3.1. Synchrotron-based Tomographic Data Reconstruction Algorithm

Dedicated imaging setups can be found in 3rd generation synchrotron based experimental endstations such as TOMCAT, Swiss Light Source (SLS, CH), ID13, European Synchrotron Radiation Facility (ESRF, FR) and I12 JEEP, Diamond Light Source (DLS, UK), allowing for 3-D tomographic data set acquisition during *in situ* deformation experiments at submicron resolution [29]. The beamline setup mainly involves a monochromatic parallel beam (z incident beam direction) and a full-field sample illumination setup (x - y sample projection plane). The sample environment allows mainly high-resolution sample rotation perpendicular to the

incident beam and several translations and tilting required for sample positioning and alignment of the centre of rotation with respect to the incident beam (Fig. 2.2).

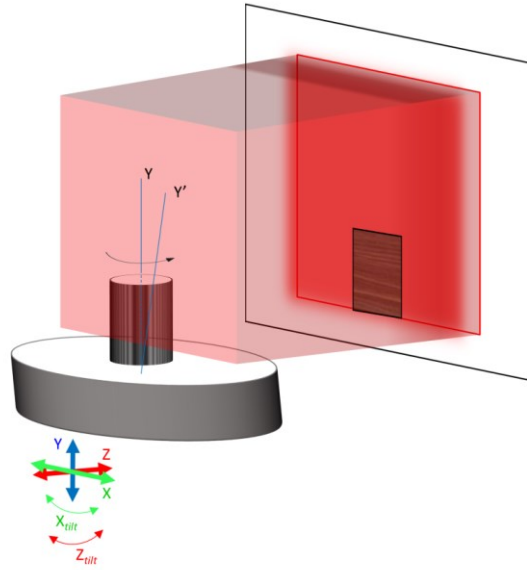


Figure 3.2: Tomographic acquisition setup indicated misalignment of the axis of rotation with respect to the imaging detector.

Sample alignment to the axis of rotation is a procedure that is extremely important for successful tomography acquisition. Dedicated imaging beamlines often use a fixed setup that is periodically re-aligned for consistency. Ambiguities related to the misalignment of the centre of rotation have detrimental consequences in terms of the produced artefacts during post-processing of the tomographic datasets. Therefore, most precise high spatial resolution tilting motors are available for the purpose. A sharp pin target is positioned off centre with respect to the sample mount and the x - y - z sample positions are adjusted so that it remains within the field of view at all rotation increments. Projections are taken at 0° and 180° and the X_{tilt} is adjusted (Fig 3.2), so that the pin-end remains at the same height on the detector. The same procedure is followed for 90° and 270° increments, and followed by adjustment of Z_{tilt} axis (Fig. 3.2). Further sample alignment and local tilting arrangements above the rotation stage, which is aligned to the “global” centre of rotation will not have an effect on the projection centre of rotation. In the aim to achieve submicron resolution on macroscopic specimens, Regions Of Interest (ROIs) are typically positioned into the field of view at different angular positions at a pre-acquisition stage.

Computed X-ray tomography requires subtraction of a “flat field” (I_0) image (i.e. beam illumination of the detector with no sample in the field of view) from each projection image in order to remove the effects that arise due to the variations in detector sensitivity. It is common practice to use a single flat field image to correct all projections, particularly when the scan time is short. Nevertheless, in the case when there is a variation of the specimen background at different rotation increments a different “flat field” acquisition strategy needs to be employed. Sample surroundings, such as a sample container, that present a non-uniform absorption throughout the projection spectrum, shall be taken into account by collecting a full set of tomographic projections ($I_{0,\theta}$) at the same sample position and rotation increments after removing the sample. Furthermore, several exposures of the detector at same exposure rates and with no beam (I_{dark}) are averaged and subtracted from both the “flat field” exposures and actual projections. Therefore the experimental measured value harvested by the detector is purely the transmitted intensity $I_\theta(x)$ through the object at the specific angular increment θ . The expression for the total absorption *integral* of the incident beam through the sample Eq. (2.3) is written as:

$$p(x, \theta) = \ln \left[(I_\theta - I_{dark}) / (I_{0,\theta} - I_{dark}) \right] \quad (3.1)$$

For the purposes of parallel beam tomographic reconstruction, an in-house MATLAB[®] implementation of the algorithm for sinogram generation and filtered back-projection reconstruction has been used. Even though the filtered back-projection reconstruction strategy only requires a sinogram generation routine and the following use of inverse Radon transformation, several filtering procedures are crucial for exploiting the full capacity of the data according to the set-up (Lab-based Micro CT vs synchrotron based CT), and the incurring acquisition parameters.

Sinogram generation is a CPU “expensive” procedure. At dedicated facilities this task is usually undertaken by processing clusters able to import simultaneously individual lines across

the projection stack in order to form sinograms. For each sinogram it is required that a single row of pixels is withdrawn from each projection across every rotation increment for 0° to 180° . The sinogram contains the absorption line profile of the examined cross-section at a specific height on the projection of the specimen at each rotation step. The optimisation of this process requires that all projection images are imported into the memory and, following a serial or a parallel process, sinograms are constructed by extracting the same row of data across all projections. The information in the form of a 2-D matrix image is then saved. The limited RAM availability in current desktop machines required the introduction of a pseudo-memory allocation procedure. A convenient strategy for experimental data memory allocation can be found by HDF Group [66] specifically on MATLAB[®] implementation.

The sinogram generation procedure is followed by a series of correction filtering procedures before final reconstruction. These tools are specific to the experimental setup:

- Background equilibration (applied to data collected on long exposure times at I12 JEEP, DLS): The storage ring of DLS operates top-up every 15min approximately under normal conditions. During this procedure, a batch of accelerated electrons is injected to the main storage ring to maintain the current ($\sim 300\text{mA}$) of the beam at the endstations (i.e. the beamlines) which are distributed across the periphery of the ring. The purpose of this procedure is to maintain a constant level of flux. However, the result of subtle variations of flux are registered by highly-sensitive modern measuring equipment (i.e. 16-bit pixelated CCD detectors coupled to scintillators) causing a respective change of the registered photon counts at each exposure. The effect can be corrected by normalising the projections according to the maximum integrated detector intensity registered at all rotation increments.

- Stripe artefact correction (applied to data collected at I12 JEEP, DLS): A combined *wavelet-Fourier filtering* ring artefact removal technique was used introduced by Munch and co-workers [67] and implemented in MATLAB[®]. The best results were obtained with a global DB10 wavelet filter, with parameters $L=8$ and $\sigma=6$, where L is the highest decomposition level

of the multi-scale wavelet transform and σ is the width of the filter in the FFT for vertical strip removal. A finer artefact removal approach with a DB10 wavelet filter, $L=6$ and $\sigma=6$ was performed at the central region of the sinograms (120 pixels wide). The results and comparison of the artefact correction procedure are shown on Figure 3.3 at different spatial resolution data sets. The dashed region on Figures 3.3 (c) and (d) shows an additional artefact produced by the ring artefact correction procedure. The outcome artefact is located at the central region of the reconstruction (axis of rotation) and did not affect the DVC process as it was away from the crack propagation ROI.

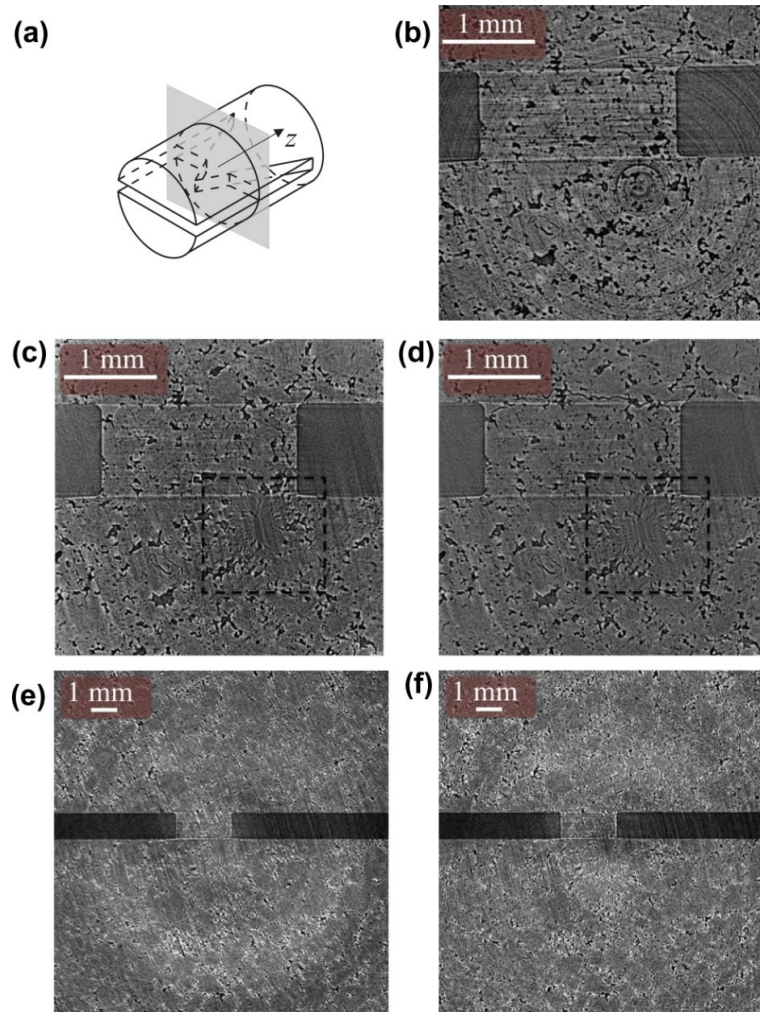


Figure 3.3: Example of a reconstructed slice cross-section: (a) slice orientation schematic; (b) reconstructed cross-section example without ring artefact correction; (c) artefact correction in cropped field of view 2672×2672 pixels – uncracked region; (d) artefact correction in cropped field of view identical to (c); (e) artefact correction of lower spatial resolution data in cropped field of view 3122×3122 pixels – uncracked; (f) artefact correction of lower spatial resolution data in cropped field of view identical to (e) – cracked. The dashed box shows the artefact produced by combined wavelet–Fourier ring artefact removal algorithm [68].

- Reconstruction (applied to data collected at I12 JEEP, DLS): The corrected sinograms after appropriate flat field correction, according to Eq. (3.1), are reconstructed using the $I =$

`iradon` ($R, \theta, \text{'filter'}$) *inverse Radon transform* function available in MATLAB® for filtered back projection reconstruction. The output of the `iradon` function is the image I from projection data, at rotation increments θ in the 2-D array R in the shape of a sinogram, using various filtering methods (“filter”). The filter value that was used in the particular reconstruction was “Hann” filter which multiplies the default “Ram-Lak” filter by a Hann window offering better reconstruction quality. Lastly, an anti-speckle filter was applied to the sinograms and median filtering was performed on the reconstructed images $[4 \times 4]$ window size following the reconstruction.

3.2. Digital Volume Correlation (DVC) of synchrotron X-ray tomography datasets

For the purposes DVC analysis, commercial software DaVis 8.1 (LaVision, Goettingen, Germany) was used. The software employs a proprietary FFT approach [69], similar to the principle used for solving Particle Image Velocimetry (PIV) problems.

The correlation procedure begins with the 3-D volume being subdivided into cube-shaped sub-volumes of voxels, referred to as 'windows'. The sub-volumes can be represented as discrete functions $f(x, y, z)$ in the unloaded step and $g(x+i, y+j, z+k)$ for the offset position (i, j, k) at subsequent deformed increments. The functions f and g represent the sub-volumes in the x, y and z coordinates, and i, j, k are the offset values in the x, y and z directions respectively. The selection of 'window' size determines the length scale at which the correlation is performed [63, 70]. If a smaller window size is selected, the mechanical behaviour at the microstructural level can be captured, provided that the tomographic reconstruction possesses sufficient resolution to reveal the microstructural features at that length scale. The use of larger window size, on the other hand, naturally delivers information about macroscopic, “coarse-grained” behaviour. These non-overlapping 'windows' are shown in Figure 3.4 where each region in the reference state is correlated with its counterpart after loading, and its relative displacement is

calculated. The average displacement is computed for each $f(x, y, z)$ by computing the offset that maximises the cross-correlation function.

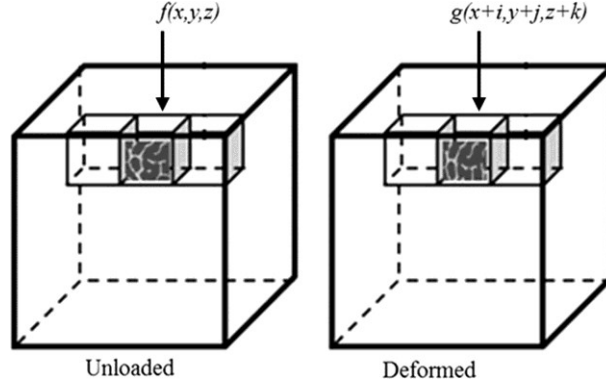


Figure 3.4: For the DVC procedure, the unloaded and deformed image volumes are divided into, non-overlapping sub-volumes. An average displacement is computed for each $f(x, y, z)$ by finding the offset (i, j, k) that maximises the cross-correlation function [71].

In order to correlate between the reference and the deformed tomographic volumes, a shape function is introduced which defines the number of DOF to be determined at each point. LaVision DaVis software uses a linear shape function with twelve DOFs, containing displacement in each direction and their first derivatives [72]:

$$\mathbf{u} = \begin{bmatrix} i & j & k & \frac{\partial i}{\partial x} & \frac{\partial i}{\partial y} & \frac{\partial i}{\partial z} & \frac{\partial j}{\partial x} & \frac{\partial j}{\partial y} & \frac{\partial j}{\partial z} & \frac{\partial k}{\partial x} & \frac{\partial k}{\partial y} & \frac{\partial k}{\partial z} \end{bmatrix} \quad (3.2)$$

$$\hat{\mathbf{x}}(\mathbf{u}) = \begin{bmatrix} \hat{\mathbf{x}}(i) \\ \hat{\mathbf{x}}(j) \\ \hat{\mathbf{x}}(k) \end{bmatrix} = \begin{bmatrix} x + i + \frac{\partial i}{\partial x} \Delta x + \frac{\partial i}{\partial y} \Delta y + \frac{\partial i}{\partial z} \Delta z \\ y + j + \frac{\partial j}{\partial x} \Delta x + \frac{\partial j}{\partial y} \Delta y + \frac{\partial j}{\partial z} \Delta z \\ z + k + \frac{\partial k}{\partial x} \Delta x + \frac{\partial k}{\partial y} \Delta y + \frac{\partial k}{\partial z} \Delta z \end{bmatrix} \quad (3.3)$$

where $\mathbf{u}$ is the 12-DOF linear shape function and $\hat{\mathbf{x}}(\mathbf{u})$ represents a point in the sub-volume, with $\Delta x = x - i$, $\Delta y = y - j$, $\Delta z = z - k$. In its simplest form, the local approach computes the pattern shift within each sub-volume, without any reference to the neighbouring sub-volumes. The routine involves the following steps: (a) Both sub-volumes f and g are processed by 3-D FFT. (b) The cross product of the FFT of the reference sub-volume $f^*(x, y, z)$ and the FFT of

the deformed sub-volume $\overline{g^*}(x+i, y+j, z+k)$ conjugate is computed. In the last step (c), the inverse FFT of the result of (b) is determined [73] and the output 3-D deformation as a function of position within the bulk is extracted for the examined loading increments.

An alternative global approach may be adopted which links neighbouring sub-volumes enabling the solution to be minimised simultaneously for the entire volume [74]. LaVision's software uses the normalised cross-correlation function C to correlate between the paired sub-volumes $f(x, y, z)$ and $g(x+i, y+j, z+k)$ for the offset position i, j and k . The cross-correlation function is described by the following equation [73]:

$$C = \frac{\sum_{x,y,z} f(x, y, z) \cdot g(x+i, y+j, z+k)}{\sqrt{\sum_{x,y,z} f(x, y, z)^2 \cdot \sum_{x,y,z} g(x+i, y+j, z+k)^2}} \quad (3.4)$$

where “.” represents the dot product of the matrices for the discrete X, Y, Z positions. The displacements are deduced at the position of the maximum in the correlation table. A Gaussian interpolation technique can be used locally, near the peak position, for the position determination with sub-pixel accuracy [73]. Correlation is an iterative process; a large window size is first selected and its average displacement is calculated; then it is broken down into smaller sub-windows, and the average displacement for each sub-window is calculated. This process may be continued until it is found that the sub-window does not contain enough features for the correlation function to achieve a match. Larger windows achieve higher precision, of up to a few percent of a voxel [75]. The uncertainty in the displacement vector determination increases rapidly (approximately exponentially) with decreasing window size. Window overlapping may be employed, increasing the number of calculated displacement vectors. This is helpful for sufficiently shallow displacement gradients, but may not provide useful information about more heterogeneous deformation at the microstructural length scales.

3.3. Three-dimensional crack observation, quantification and simulation in quasi-brittle materials

Structural integrity of quasi-brittle materials, such as graphite [76-78], ceramic composites [79] and bone replacements [80], is commonly assessed using the assumption of brittle fracture (e.g. Ref. [81, 82]). Although the nonlinear cracking behaviour of some quasi-brittle materials such as concrete has been investigated (e.g. Ref. [83-85]), the inelastic behaviour of many quasi-brittle materials is generally treated as negligible [81, 86, 87]. Nonetheless, tension-softening behaviour during crack propagation can play an important role in the integrity of components that are made of quasi-brittle materials. Understanding and quantifying this non-linear behaviour of quasi-brittle materials can effectively improve confidence in the safety margins of their structural integrity assessment.

A partially-cracked component, fabricated from a quasi-brittle material, develops a fracture process zone due to distributed microcracking ahead of the crack tip [77, 88] during indentation loading experiments. The extent of the fracture process zone dictates the magnitude of the deviation of the component's deformation and fracture behaviour from that predicted by linear elastic analysis [68]. An analogy can be established between the fracture process zone in quasi-brittle materials and the plastic zone in ductile materials [89]. A larger fracture process zone contributes more redundant inelastic energy to the measured critical energy release rate, resulting in higher fracture resistance. Moreover, the fracture process zone increases in size during crack propagation [90], leading to the classic R-curve behaviour [91].

The fundamental difference between the crack opening displacement (COD) obtained by models that consider the strip yield model, and the predictions of linear elastic or elastic-plastic fracture mechanics [92, 93] is schematically illustrated in Figure 3.5 (a). A recent investigation in polygranular graphite showed that the measured COD was close to that suggested by strip yield models [94]. Crack propagation in materials that demonstrate strip yield model behaviour [95] may be simulated by cohesive zone modelling. Proposed by Hillerborg, Modeer and Petersson [83] cohesive zone models have been applied widely, through a traction-separation

law (Fig. 3.5 (b)), simulating fracture in concrete [84, 96-99], adhesively bonded joints [100, 101] and ceramic-metal composites [102].

Gilsocarbon graphite is a near-isotropic, heterogeneous material placed in load-bearing applications in moderators and reflectors within nuclear fission reactors [103]. It is used in the form of blocks that are keyed in a structure that forms the reactor core. As a result of neutron irradiation and thermal strains, dimensional changes develop multi-axial stresses within the graphite structure which can act as fracture initiation regions in the moderator blocks [104]. By taking into account elastic-plastic behaviour of quasi-brittle materials the in service life of components can be safely extended, by reducing over-conservatism in their fracture assessments.

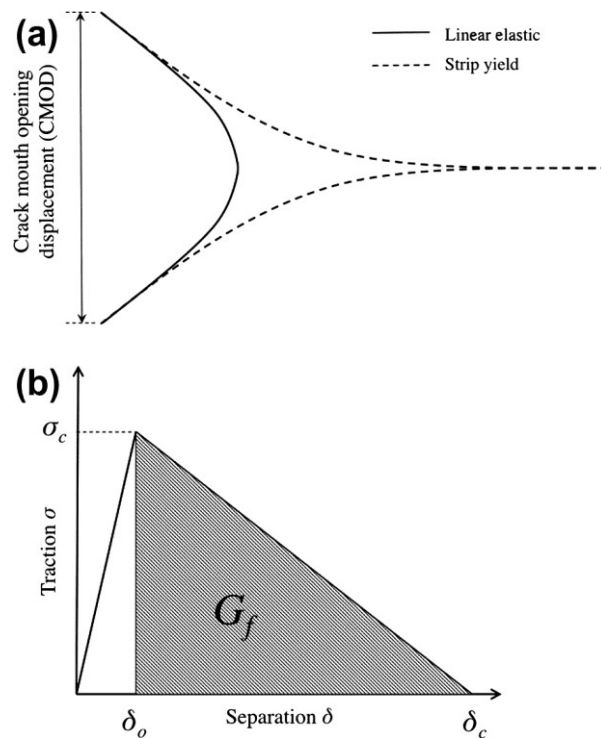


Figure 3.5: Cohesive zone model: (a) crack opening profile; (b) traction-separation law [68].

In the context of the chapter, crack initiation and propagation were examined using a chevron notch specimen fabricated from “virgin” (un-irradiated) Gilsocarbon graphite. 3-D X-ray tomographic datasets were collected with the specimen in its un-damaged condition at beamline I12 JEEP in DLS. A sharp wedge was driven progressively into the specimen's notch to initiate and propagate a crack; tomographic images were taken at each loading stage and

subsequently analysed using commercially available digital volume correlation software [105] to obtain full-field 3-D displacements, thereby measuring the crack opening profile.

3.3.1. Experimental Setup - Data Processing

Specimen geometry and loading

The short bar chevron notch specimen was designed following the recommendations of the International Society of Rock Mechanics [106]. The geometry of the specimen is shown in Figure 3.6 (a), and its dimensions are reported in Table 3.1. In Table 3.1 the values denoted as nominal are those used in the technical drawings, and the ones denoted as tomography were measured using the XCT images. It can be seen that the measured values are within 6% of the specified values. The crack is initiated and propagated by carefully driving a wedge into the notch, causing the two sides of the specimen to open. The coordinate system used throughout this paper, for a crack of length a , is depicted in Figure 3.6 (b).

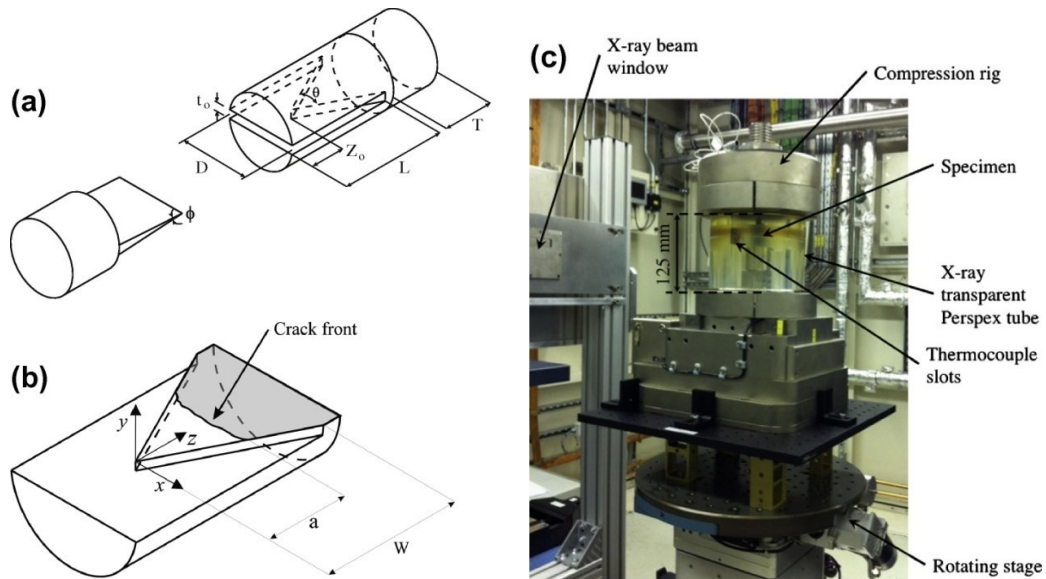


Figure 3.6: Experiment overview: (a) chevron notch specimen; (b) half chevron notch specimen; (c) experiment setup in the beamline [68].

The specimen was scanned in the as-received condition (loading step 0) and then loaded until a maximum load of 310N was achieved (loading step A). The load on the wedge was reduced to zero by progressively withdrawing the wedge (loading step C). The specimen was then loaded progressively to 360N (loading step G). Intermediate scans were performed (step

D to F), but are not reported here in detail. The loading step is summarised in Table 3.2. Inspection of the tomographic reconstructions identified a macroscopic crack that was approximately 3.4mm in loading step A, which propagated to a length of 3.7mm by loading step G. The macroscopic crack wake (i.e. the material behind the crack tip) was removed using electro-discharge machining up to a position within $\pm 0.2\text{mm}$ of the identified crack tip, and the final scan was recorded (loading step H).

Table 3.1: Component dimensions

Parameter	Symbol	Measure		Unit
		Nominal	Tomography	
Notch thickness	t_o	1	1.06 ± 0.02	mm
Specimen diameter	D	24	-	mm
Notch height	Z_o	12	-	mm
Notch angle	θ	55	55.4 ± 0.4	Degrees ($^\circ$)
Specimen length	L	35	-	mm
Wedge angle	φ	10	10.4 ± 0.5	Degrees ($^\circ$)
Ligament	W	23	-	mm

Table 3.2: Loading increments steps

Loading step	Resolution ^a	Load (N)	Description
0	HR, SR	0	As-received
A	HR, SR	310	Step 1 – loaded
B	HR	250	Step 1 – partially unloaded
C	HR, SR	0	Step 1 – unloaded
D	SR	330	Step 2 – loaded
E	SR	280	Step 2 – load drop
F	SR	330	Step 2 – repeat
G	SR	340	Step 3
H	SR	0	Cut specimen

^aSR: standard resolution (5 μm voxel size); HR: high resolution (1.8 μm voxel size).

Tomography procedure - Beamline Setup

In situ loading was carried out with a compression rig equipped with a PMMA tube, transparent to visible and X-ray light. The loading rig (manufactured by Deben UK) is shown in Figure 3.6 (c) on the I12 stage. The beam was monochromated at 80keV using a double crystal Si monochromator; 4500 projections over 180° were recorded (i.e. at 0.04° rotation increments) with the specimen rotating around an axis parallel to the z direction (Fig. 3.6 (b)). A high resolution imaging detector (PCO.4000) was used for data acquisition. The detector field of view is 4000×2672 pixels ($x \times y$), with two adjustable scintillator-objective sets at different

magnifications used to record the projections: standard resolution (denoted by SR) with voxel size $\sim 5\mu\text{m}$ and high resolution (denoted by HR) with voxel size $\sim 1.8\mu\text{m}$. The SR scans allowed an area of $20 \times 13.4 \text{ mm}^2$ to be radiographed. The HR scans, although observing a smaller area ($7.2 \times 4.8 \text{ mm}^2$) provided better microstructural observations.

Table 3.3: Crack characteristic summary

Loading step	Observed crack length ^a (mm)	Estimated linear elastic crack length ^b (mm)	Crack mouth opening displacement (CMOD) ^c (mm)	Crack opening compliance (CMOD/F) ($10^{-6} \times \text{mm N}^{-1}$)	Estimated fracture toughness ^d ($\text{MPa m}^{0.5}$)
A					
SR	3.40 ± 0.02	5.14 ± 0.01	0.0291 ± 0.0048	93.87 ± 15.48	1.19 ± 0.19
HR	3.45 ± 0.01	5.25 ± 0.02	0.0315 ± 0.0011	101.61 ± 3.55	1.21 ± 0.04
B					
SR	–	–	–	–	–
HR	3.31 ± 0.00	5.21 ± 0.02	0.0311 ± 0.0011	124.40 ± 4.40	NA
C					
SR	3.40 ± 0.05	5.12 ± 0.01	0.0283 ± 0.0050	NA	NA
HR	3.41 ± 0.02	5.15 ± 0.04	0.0294 ± 0.0017	NA	NA
D					
SR	3.43 ± 0.01	5.29 ± 0.01	0.0283 ± 0.0049	85.76 ± 14.85	1.17 ± 0.20
HR	–	–	–	–	–
E					
SR	3.43 ± 0.02	5.19 ± 0.01	0.0291 ± 0.0049	103.93 ± 17.5	1.17 ± 0.20
HR	–	–	–	–	–
F					
SR	3.60 ± 0.02	5.08 ± 0.01	0.0295 ± 0.0051	89.39 ± 15.45	1.19 ± 0.21
HR	–	–	–	–	–
G					
SR	3.68 ± 0.02	5.16 ± 0.00	0.0312 ± 0.00	91.76 ± 16.18	1.24 ± 0.22
HR	–	–	–	–	–

^aThe error is the visual variation in the crack lengths.

^bThe error is calculated from the variation in the normalized crack opening gradients.

^cThe error is calculated from the variation of the average displacement.

^dThe error is calculated from the variation in the crack length and normalized opening gradients.

Table 3.2 summarises the magnification used at each loading sequence. The exposure times were 2s and 3s for the SR and HR scans respectively, with an extra second required for the data transfer. The time per tomography scan was therefore four hours for SR and five hours for HR observations. Visual observations of the crack tip position show no significant difference between SR and HR modes, as shown in Table 3.3.

Digital volume correlation analysis

Each tomography acquisition scan provided 2672 virtual slices in the form of 4008×2672 (horizontal resolution of the detector) pixel 16-bit images. Reconstructions were afterwards converted to 8-bit raw stacks for input into the DVC calculation. DVC analysis was performed on the tomography datasets, using DaVis StrainMaster software (Version 8.1) as mentioned in the previous section [105]. Each was correlated against the as-received condition (loading step 0) with a $256 \times 256 \times 256$ interrogation window, 50% overlap and 2 passes, followed by $64 \times 64 \times 64$ interrogation window, 50% overlap, 2 passes and finally followed by $32 \times 32 \times 32$ interrogation window, 50% overlap and 4 passes. Examples of the displacement fields (V_y) are presented in Figure 3.7 (a) and (b), at the approximate position of the virtual slices shown in Figure 3.3 (previous section) for the HR and SR scans respectively.

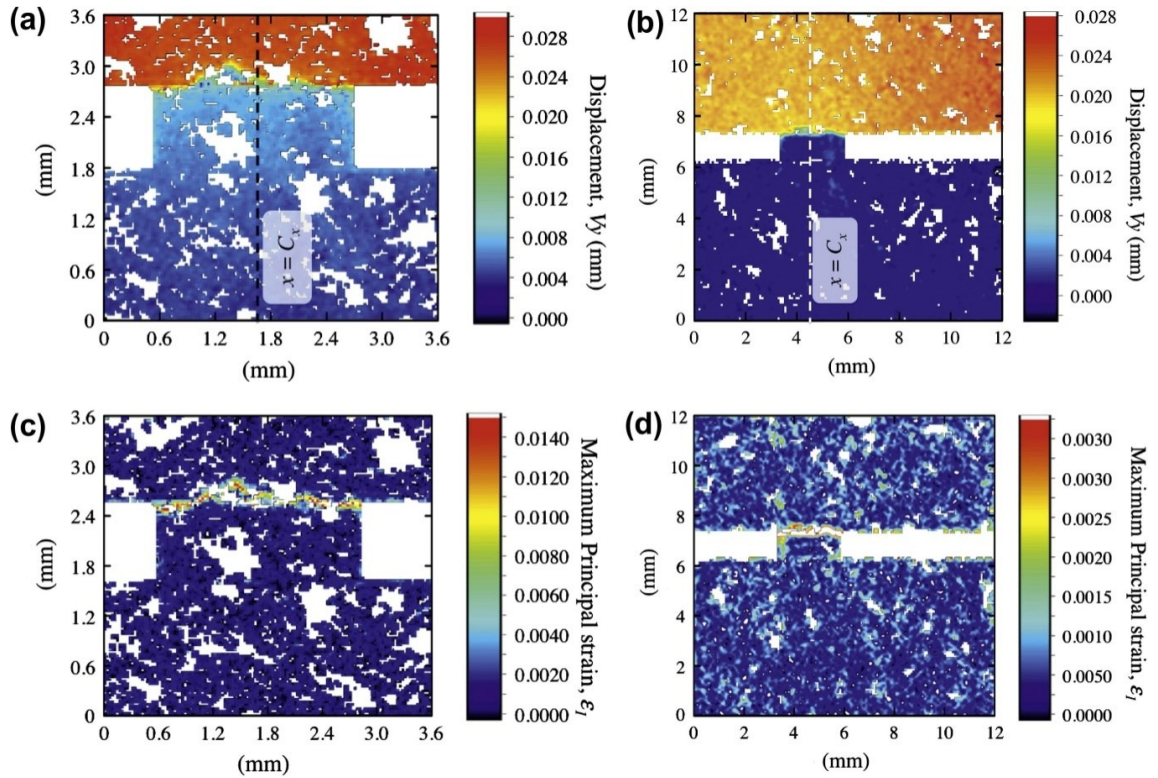


Figure 3.7: Examples of opening displacement and maximum principal strain distribution of the virtual slice shown in Figure 3.3: (a) HR opening displacement (V_y); (b) SR opening displacement (V_y); (c) HR maximum principal strain (ϵ_I); (d) SR maximum principal strain (ϵ_I) [68].

Due to the size of the interrogation window with 50% overlap in the analysis, the displacement field slice represents material within ± 16 voxels of the absorption tomography slice, which is equivalent to ± 0.03 mm for HR and ± 0.08 mm for SR. White patches in Figure

3.7 indicate the areas where the displacement vectors were deleted due to a low coefficient of correlation (<0.5), attributed to effects of image noise and artefacts. The figures show a clear step in the displacement field at the crack location. The shown displacements are absolute values that incorporate the rigid body movement of the specimen. The crack opening is obtained from the relative displacement, described below. Strain maps, calculated from the displacement field, are an effective way to visualise the crack opening [107] demonstrating an artificially high strain at its location, as shown in Figure 3.7 (c) and (d) for HR and SR data respectively.

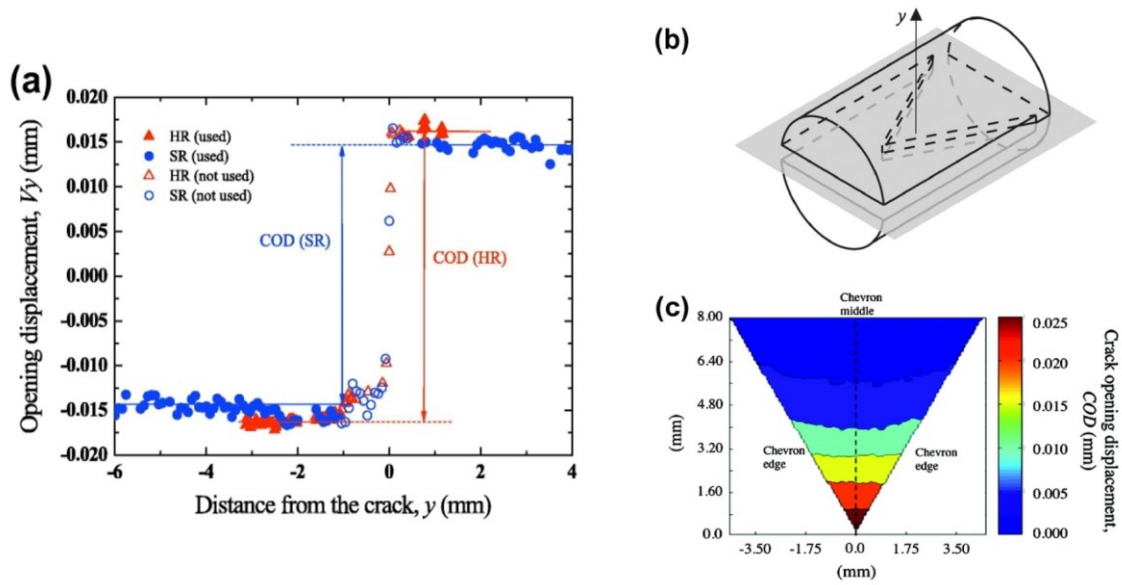


Figure 3.8: Measured COD: (a) example of the opening displacement profile perpendicular to the crack along $x = c_x$ (see Fig.3.7 (a) and (b)) – solid line shows the length on which the average has been calculated; (b) virtual slice; (c) COD distribution on the virtual slice at the first loading step (A) calculated from the SR data [68].

To calculate the COD, the difference between the displacements (displacement along the y direction V_y) on either side the crack was used. Examples of the distribution of V_y along an arbitrary profile perpendicular to the nominal crack plane ($x = C_x$) (shown in Fig. 3.7 (a) and (b)) are plotted in Figure 3.8 (a) for HR and SR data, showing that the displacements either side of the crack are approximately constant (as observed previously in the less precise laboratory experiment [94]), i.e. there is no measureable strain in the material adjacent to the crack. The average V_y on either side of the crack was calculated over the projected surface of the crack (see Fig. 3.8 (b)) using data only from image correlation windows that did not overlap the crack (for more detail on the method see Ref. [108]). An example of the obtained map of COD is

shown in Figure 3.8 (c) (SR scan at loading sequence A); the crack is observed to open more near the chevron edge compared with the middle.

The COD profile along the crack from its mouth to its tip was obtained using the averaged values along a series of lines perpendicular to a sectioning plane that is defined in Figure 3.9 (a). Figure 3.9 (b) shows an example of the reconstructed image on the sectioning plane at the middle of the chevron. The visible crack length in this slice is 3.31 mm. Figure 3.9 (b) shows two artefacts in the form of high contrast horizontal lines, which form along the edges of the notch. These artefacts are due to the strong difference in X-ray attenuation between air and graphite, and are generally observed at the boundaries of reconstructed images of specimens. Here they arise from notch edges. They do not interfere with the DVC analysis since they move with sample deformation.

The SR data for loading steps A, C, F and G that correspond to the progressive increase in the crack length are shown in Figure 3.9 (c). The measurements near the crack mouth are magnified in Figure 3.10 (d). The equivalent measurements obtained from HR data, which have less noise, are shown in Figure 3.9 (e) for comparison. Data were unfortunately not available from the crack tip. The average crack mouth opening displacement (CMOD) increased from 0.0298 to 0.0312mm (SR scans - see Table 3.3 for full data, the values calculated from by analysis of SR and HR data at the same loading state differ by less than 5%). Crack mouth opening displacement is dependent on the geometry, loading arrangement and therefore can serve as a useful overall descriptor of the problem. In contrast, the crack tip opening displacement (CTOD), also often referred to simply as the crack opening displacement (COD) characterises solely the crack tip geometry and is most directly related to the stress intensity factor and the near-tip stress fields that determine the conditions of crack propagation. At other points on the crack face, the difference between HR and SR measurements of the COD is between 3% and 13%. Using the RMS variation of the average V_y either side of the crack to obtain an approximate measurement error, the uncertainty in the SR data CMOD measurement is estimated at 0.0055mm, which is significant compared to the average CMOD change with

crack growth. However, comparison of loading sequence D and F, which were imaged at the same applied load, shows a difference of less than 0.001mm in the average CMOD. The progressive increase in mean CMOD could therefore be due to crack extension or damage development.

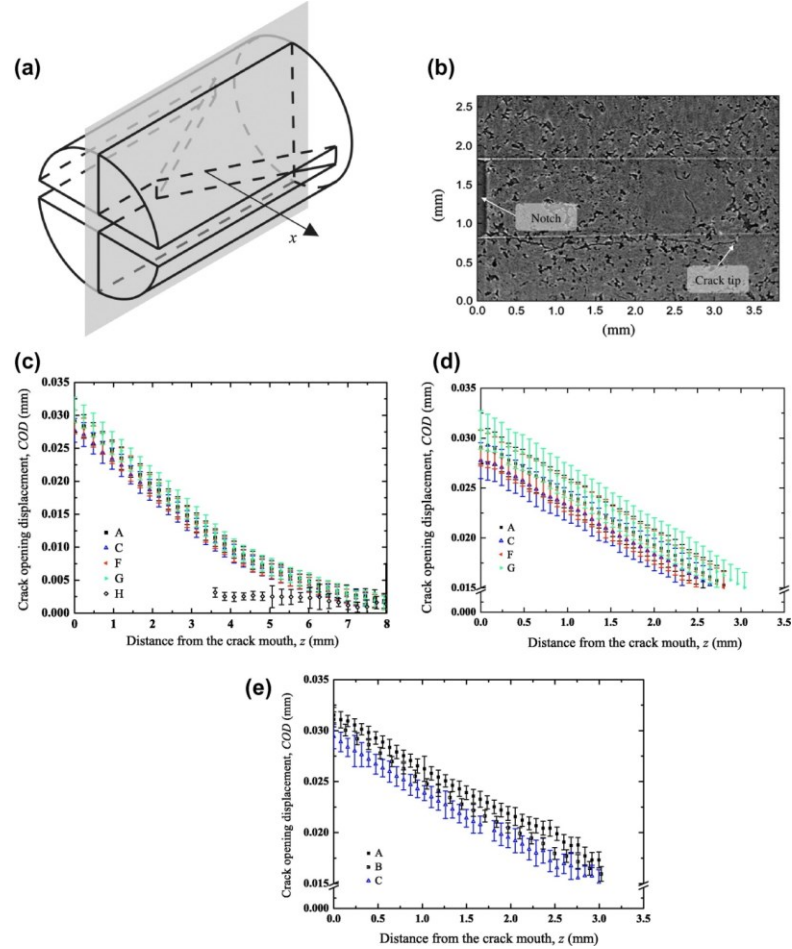


Figure 3.9: Crack opening profiles: (a) virtual slice on which the profile is shown; (b) example of reconstructed virtual slice loading sequence A; (c) averaged COD distribution at different loading steps (SR data); (d) focus on the crack mouth opening displacement (SR data); (e) averaged COD distribution at different loading step (HR data – only every fifth data point is shown) [68].

To discern the effects of possible in-plane shear (mode II) and out-of-plane shear (mode III) loading that might have been applied to the specimen due to misalignment, crack displacements in the x and z directions were also calculated. Two volumes of $70 \times 47 \times 58 \mu\text{m}^3$ ($x \times y \times z$) either side of the chevron were selected and their average displacements in x (V_x) and y (V_y) directions were extracted. The Mode II and mode III displacements were measured, with less than 0.5% variation, to be 0.0004 and 0.0016 mm, which are 1% and 5% of the mode I crack mouth opening displacement respectively. The components of mode II and mode III loading are thus negligible.

3.3.2. Finite element simulation

For the purposes of accurately characterising the crack opening behaviour, both a linear elastic and a cohesive zone model were implemented by Mostafavi et al. [68] to analyse, match and map the characteristic behaviour of Gilsocarbon graphite samples that were used. A linear elastic model was employed to determine the crack related mechanical parameters, such as COD and fracture toughness (K_I) and relate them to the experimental observations. Furthermore, a strip-yield model of the crack opening was simulated, employing a cohesive zone modelling technique [109] for analysing the observed crack-propagation behaviour based on the experimental data observation that cohesive energy (i.e. the fracture resistance) of quasi-brittle materials is constraint-dependent, similar to elastic-plastic materials [110]. A bilinear traction-separation law (Fig. 3.5 (b)) was used to describe the cohesive properties, taking its parameters from extant uniaxial strength and high constraint fracture toughness data [107, 111], as suggested by Cornec, Scheider and Schwalbe [112]. Both models were implemented using Abaqus/standard V.6.10 [113].

Linear elastic model

3-D linear elastic models with different crack lengths ($a = 1.0$ to 5.8mm , at 0.2mm steps) were created. Other dimensions are the nominal values reported in Table 3.1, which are within 6% of the real values measured through XCT images. Linear 8-node hexahedral elements (in total, 57984) were used to simulate the specimen. Figure 3.10 (a) shows the overview of the FE mesh. Nominal Gilsocarbon material properties [114] $E=10.9\text{GPa}$ (its elastic modulus is more than ten times that of graphite) and frictionless contact between the rigid wedge and graphite specimen was defined. Friction does not affect the relation between load and COD for this geometry [93]. Since the shape of the crack profile near the crack tip and the stress distributions around it were not the focus of this study, it was not necessary [115] to use specialised crack tip meshes such as the crack-type concentrated [116] and degenerate mesh [117] with singular elements [118].

A previous linear elastic FE analysis has shown [93] that the gradient of the linear part of the COD profile from the crack mouth depends on the crack length. For each model, the normalised (COD/CMOD) crack openings in the xz plane were extracted from the FE results. Each distribution corresponding to a certain crack length was then used to calculate the average normalised crack opening displacement, as was done for the experimental data. The normalised COD was then used to obtain the linear variation from the crack opening mouth ($z = 0$) to $z = 3\text{mm}$ (R-values better than 0.98). By extracting the displacement vectors along a line orthogonal to the notch plane and fitting a fifth-order polynomial (with R-value better than 0.99), the difference between the maximum and then minimum value was taken as the COD. The crack length obtained was approximately 5.2mm for all loading conditions (Table 3.3), which is about 1.6mm longer than visually observed in the tomography absorption images (see Fig. 3.3).

In the short rod chevron sample ($a/W < 0.5$) a stable propagating crack in an elastic material has limited effect on the fracture toughness [106], where a is the crack length and W is the testpiece thickness. Consequently the crack lengths and the crack mouth opening displacements under load may be used with the linear elastic FE analysis to estimate the fracture toughness. To this end, the mode I stress intensity factor K_I was calculated using ABAQUS® via the interaction integral method [119] for the crack length $a=5.2\text{ mm}$ (the average effective linear elastic crack length) and the crack mouth opening displacement of 0.0291mm. The stress intensity factor calculation by this method is largely independent of the mesh when dealing with mode I only [120], so there is no need for crack-type mesh. The measured crack mouth opening and effective linear elastic crack lengths for all tests are very similar (within 3%), so the linear FE-simulated stress intensity factor, K_I , was normalised by the modelled nominal opening stress σ ($\sigma = F/A$; F is the applied force and A is the area of the ligament ahead of the crack front) and crack length a . The relationship below was used to calculate the normalised fracture toughness at each of the crack mouth opening displacements and crack lengths reported in Table 3.3 [94].

$$\bar{K} = \frac{K_I}{\sigma\sqrt{a}} \quad (3.5)$$

The average fracture toughness ($1.20 \pm 0.18 \text{ MPa}\cdot\text{mm}^{0.5}$) compares very well with the fracture toughness of Gilsocarbon graphite measured standard high constraint geometries (e.g. compact tension specimen [121]).

Cohesive zone model

The experimental data show the shape of the crack opening profile. The crack lengths obtained from its linear elastic analysis differ from the experimentally observed behaviour (see Fig. 3.5 (a)). To address this, a cohesive zone FE simulation was carried out. Two half models (cut in the x - z plane shown in Fig. 3.8 (b)) were created; the mesh on either side was identical. A tie equation was defined to attach the two halves of the specimen remote from where fracture occurs: this was at the end of the chevron notch at $z = 23 \text{ mm}$ (i.e. where the threads were cut into the specimen, denoted by T in Figure 3.6 (a)). From the tip of the chevron (i.e. the crack mouth at $z=0$) to $z = 3.6 \text{ mm}$ (observed in the experiment as the macroscopic crack tip) the two faces were not constrained and could open freely. A cohesive contact was defined from $z = 3.6 \text{ mm}$ to $z = 23 \text{ mm}$. The model therefore allows the faces of the crack (from $z = 0$ to $z = 3.6 \text{ mm}$) to open freely and for damage to occur from $z = 3.6 \text{ mm}$ to the end of the chevron. The sample is solid beyond the end of the chevron notch.

The elastic material properties were defined as in the linear elastic model, with a bilinear traction-separation law (Fig. 3.5 (b)) applied to the cohesive contact area. It has been argued that the defining parameters for a bilinear traction law can be obtained experimentally [112]: the cohesive energy G_f may be assumed to be equivalent to the critical energy release rate in a highly constrained geometry (e.g. compact tension test) and the critical stress σ_c may be assumed to be equivalent to the ultimate stress in a tensile test. These assumptions leave enough flexibility in terms of the length parameters in the cohesive zone definition to obtain satisfactory model calibration [122]. For Gilsocarbon graphite, the typical uniaxial flexural strength has

been measured to be 28MPa [108] and the critical energy release rate to be 260J/m² [123]. These studies used the same Gilsocarbon grade of nuclear graphite, with a nominal void volume fraction of about 20% (the flexural strength and critical energy release rate are microstructure dependent, being affected by the porosity of graphite [124]).

An example of the simulation of a deformed specimen (when CMOD = 0.0291 mm, i.e. loading step A) is shown in Figure 3.10 (b) and (c). The distribution of damage in the x - z plane coincident with the crack is shown in Figure 3.10 (d). There is full separation (damage parameter = 1) in front of the 3.6mm crack over a distance of 1.6mm, giving a total crack length of 5.2mm. This is in excellent agreement (within 1%) with the effective crack length obtained from the linear elastic model using the measured COD profile (see Table 3). The comparison between the experimentally measured COD distribution (HR and SR) and linear elastic and cohesive contact models is shown in Figure 3.10 (e).

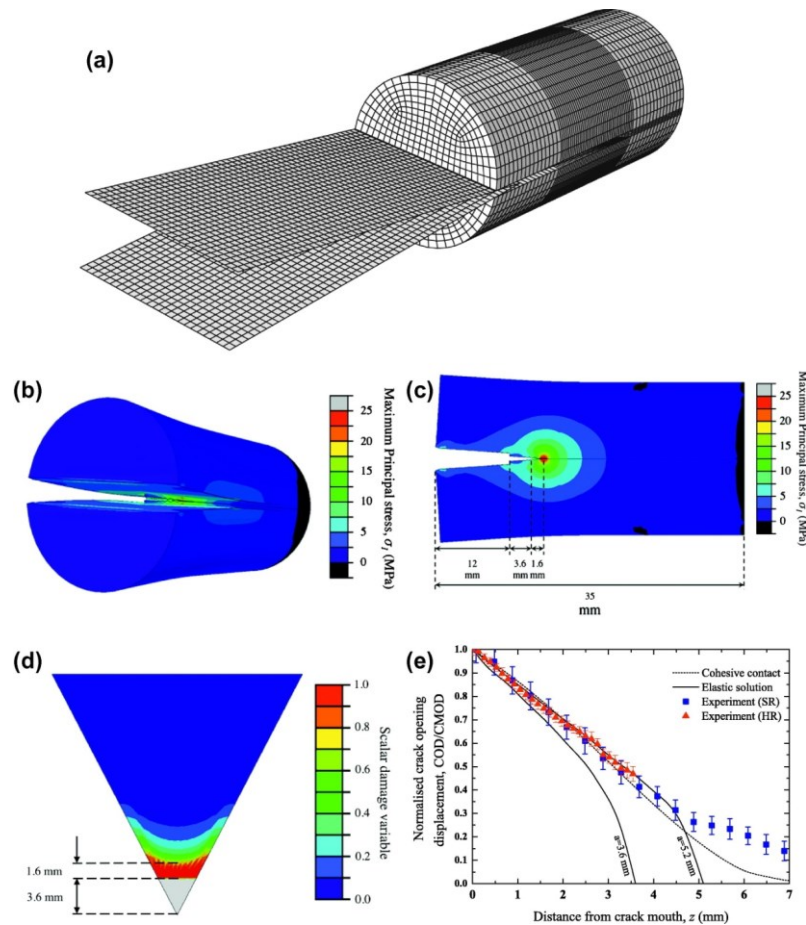


Figure 3.10: FE simulation and experiment comparison: (a) FE mesh; (b) example of full deformation; (c) example of deformation in yz plane; (d) damage distribution in xz plane (orientations as in Figure 3.6 (b)); (e) experimentally measured COD in comparison with elastic and cohesive contact FE models. The deformations in (b and c) are magnified 25 times [68].

To study the sensitivity of the simulation to the cohesive zone parameters, the effects of changing G_f and σ_c over reasonable experimental ranges were examined ($\sim \pm 30\%$ variation). In the first set of simulations the cohesive energy remained constant ($G_f = 260 \text{ J/m}^2$) and the critical stress was varied from 23 to 35 MPa. In each simulation the load was increased until the crack mouth opening displacement reached the experimentally measured value for loading step A (i.e. 0.0291 mm). The second series of simulations used a constant critical stress ($\sigma_c = 28 \text{ MPa}$) and varied the cohesive energy G_f from 180 to 340 J/m^2 . Profiles of the scalar damage variable d ($d = 0$ before damage initiation, $d = 1$ corresponding to complete local failure of the interface) in the middle of the chevron are shown in Figure 3.11 (a) and (b). The effects on the average COD/CMOD profiles are shown in Figure 3.11 (c) and (d), compared with the linear elastic model for crack lengths $a = 3.6 \text{ mm}$ and $a = 5.2 \text{ mm}$.

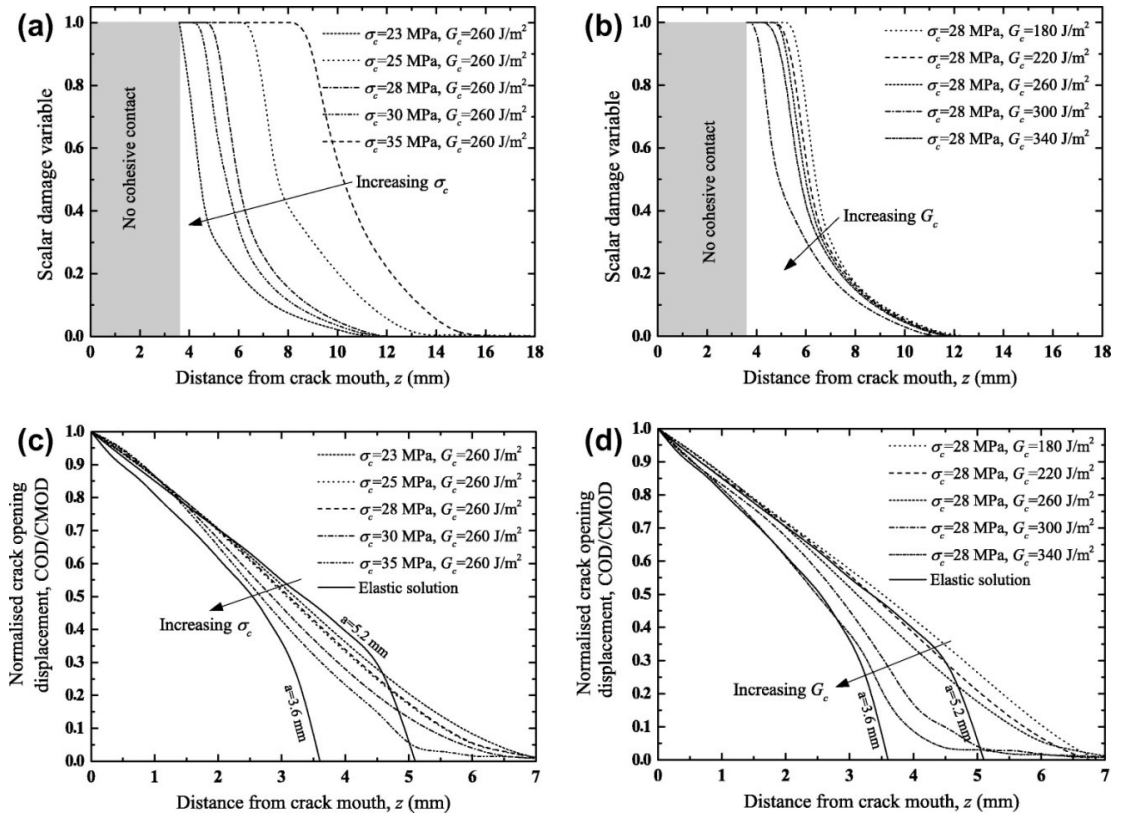


Figure 3.11: Sensitivity study of the bilinear traction-separation law: (a) damage profile with respect to varying critical stress; (b) damage profile with respect to varying cohesive energy; (c) crack opening profile with respect to varying critical stress; (d) crack opening profile with respect to varying cohesive energy [68].

The crack opening profiles of experimental data, pure elastic model (with crack length $a = 5.2 \text{ mm}$) and cohesive zone model with $G_f = 260 \text{ J/m}^2$ and $\sigma_c = 28 \text{ MPa}$ are compared in Figure

3.12 (a) and (b) for the SR and HR data respectively, using the 3-D data to examine the opening at the middle and edge of the chevron (Fig. 3.8 (c) for the definition of chevron edge and chevron middle). The data are normalised by the measured crack mouth opening displacement ($\delta = 0.0291\text{mm}$). The cohesive zone model predicts greater opening in the damaged zone at the chevron edge, compared to the middle, and this is also seen in the experimental data.

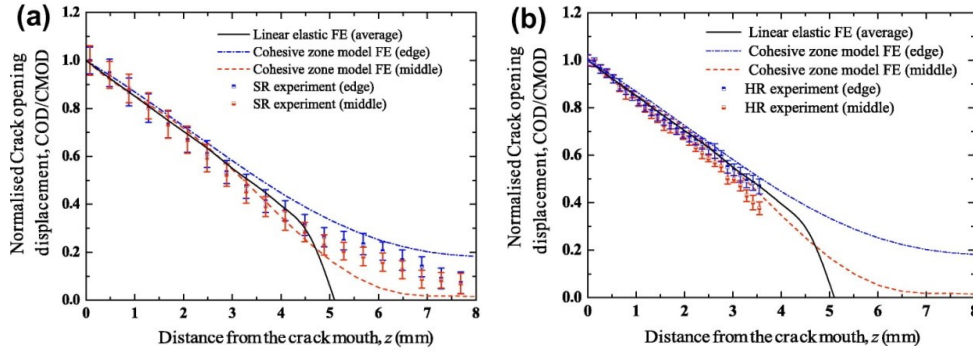


Figure 3.12: FE simulation and experimental measurement comparison of normalized COD: (a) SR data; (b) HR data (every fifth data point is shown) [68].

3.3.3. Conclusions

Combined DVC and XCT was used in this study to measure the full-field 3-D displacements in a short bar chevron notch specimen fabricated from Gilsocarbon graphite. The artificial high strains arising from the crack opening displacements were exploited to segment the crack, which was otherwise difficult to achieve in the absorption contrast images of this porous and complicated microstructure. The 3-D interaction of the propagating crack with the microstructure was observed by combining the strain visualisation and the CT data. Such crack behaviour cannot be readily observed in 2-D studies. More importantly, the measured displacement field was used to quantify the crack opening displacement (e.g. Fig. 3.8 (c)). The average profile of crack opening displacement (Fig. 3.9) confirmed that the response of that the response of graphite is far from linear elastic and close to the characteristics of the strip yield or cohesive zone model that is commonly applied to concrete (a quasi-brittle material).

Linear elastic modelling was employed to estimate the effective crack length from the gradient of the measured crack opening profile, obtaining crack lengths that were approximately 1.5mm longer than the macroscopic cracks observed in the absorption contrast tomographic

images. Cohesive zone modelling, using a simple bilinear traction-separation law with experimentally measureable parameters (cohesive energy and critical stress), correctly predicted both the general characteristics of the measured crack opening profile and the same effective crack length as that obtained using a simple elastic model, fitted to the crack mouth opening profile.

The sensitivity study of the bilinear traction-separation law in the cohesive zone model (Fig. 3.6) showed that the cohesive energy and critical stress have a combined effect on the distribution of the damage index; this arises from the interaction between damage and the stress field in the sample. The fracture process zone is therefore not independent of the sample geometry. The main effect of the critical stress is on the extent of the fully damaged area, whereas the cohesive energy mainly changes the gradient of the damage distribution.

Comparison of the measured and modelled profiles at the middle and edge of the crack (Fig. 3.12 (a) and (b)) showed good agreement (better than 10%) between the cohesive zone model and the experiment over the length of the macroscopic crack. No significant difference over this region between the opening at the middle and edge. However, within the fracture process zone (i.e. from $z = 3.6$ to $z = 5.2\text{mm}$), the difference between the model and experiment increased. In the middle the model under-predicted the COD, whilst over-predicting it at the edge near the free surfaces.

The middle and edge of the fracture process zone experience different degrees of constraint from the surrounding material. The through-thickness variation of constraint (plane stress and plane strain in extreme conditions) is usually referred to as out-of-plane constraint. Using the behaviour shown in Figure 3.11 the cohesive energy should be decreased to increase the matching with the experimental results, assuming that critical stress is independent of constraint level. However, to fit the edge measurements the cohesive energy should be increased. Therefore it is not possible to use a single pair of cohesive energy and critical stress parameters to simulate the observed crack openings in the fracture process zone. This parameter

dependence on the out-of-plane constraint is in agreement with the expectation that the quasi-brittle behaviour of materials, such as graphite, is similar to that observed in elastic-plastic materials which exhibit a prominent constraint effect (see for example [125]). This effect may become significant in some sample geometries (such as notch specimens with $a/W < 0.5$ under flexure [126]), and is in agreement, for instance, with the observed dependence of the fracture resistance curve in polygranular graphite on the stress state [108].

A cohesive contact technique was implemented by Mostafavi et al. [68] for the purpose of simulating the experimental data. The approach requires a predefined crack propagation direction and assumes a smeared 2-D fracture process zone in front of the crack tip. The cohesive contact method is suitable for controlled crack propagation in a test specimen such as reported here, and from the point of view of engineering design it is always preferred for cracks with known propagation paths. A more general modelling route is brittle cracking technique that does not require a pre-determined path and could be used to simulate the same macro-mechanical behaviour, including the development of a diffuse 3-D fracture process zone [127-129]. The loading in this experiment was mode I, but the cohesive contact used in this study is capable of simulating damage in shear (mode II and III), as the contribution of opening versus sliding can be defined in the critical stress and separation energy. A mixed mode experiment would be required to validate further simulations performed using the cohesive contact method that has been presented here.

3.4. Three-dimensional multi-scale *in situ* deformation analysis of (Al-12%Si)-12%Ti metal-metal composite

Metal-metal composites are different from conventional composites in that both the matrix and the reinforcing phase are ductile metals [2, 130], for example, a face centred cubic (fcc) metal reinforced by a body centred cubic (bcc) metal, e.g. Cu-Nb or Al-Nb, or a fcc metal with a hexagonal close-packed (hcp) metal, e.g. Al-Ti. Typically, such metal-metal composites are manufactured using either power mixing [130] or co-spray forming [131] and then extruded or

rolled into a bar or sheet form. Ultimate tensile strengths of 2200, 1030 and 890MPa have been reported for Cu–18 vol (%) Nb, Al–20 vol (%) Nb, and Al–20 vol (%) Ti [130]. These strengths are greater than those predicted by the rule of mixtures because the minority metallic phase is drawn into micro- or nano-scale fibres with a high interfacial area with the crystallographically different majority matrix phase, across which dislocation motion is impeded. The precise strengthening mechanism involved forms an active research area [2].

The principal focus of the present study was to evaluate the mechanical response of a hierarchical composite material to *in situ* mechanical loading [132]. Samples of Al-12 wt (%) Si alloy with co-injected 12 wt (%) Ti powder (grain size $<150\text{ }\mu\text{m}$, $D_{50} = 80\text{ }\mu\text{m}$) [2] were studied by absorption tomography under *in situ* compression. Collecting microstructural information, during multiple loading increments, allows the extraction of quantitative information about the mechanical response by combining high resolution micro-tomography with DVC. The key advantage of the section concerns the multi-scale deformation analysis of *in situ* loading sequences of a composite material [133], interpreted using the DVC technique [71, 130, 134-137]. The validation of these DVC results is obtained by comparison with FE simulation and MATLAB[®] deformation analysis within intensity saturated Ti inclusions. The range of potential applications of this approach is discussed below. In particular, the potential is assessed of DVC interpretation of tomographic datasets to provide a means of evaluating sample strain over multiple length scales, i.e. at the macro- and microscopic levels. The results demonstrate the power of tomographic techniques combined with advanced post-processing methodologies, and pave the way to wider application of these approaches for better multi-dimensional, multi-scale characterisation of deformation in complex-shaped components under various loading conditions.

3.4.1. Experimental Setup - Data Processing

Sample preparation

The sample was manufactured by the co-spray forming process. Ti powders are injected and mixed with the Al–12 wt (%) Si alloy droplets during the atomisation of the Al–Si melt stream at Peak Werkstoff GmbH, Germany to form $\varnothing 300\text{mm}$ composite billets which are then machined down to $\varnothing 297\text{mm}$ and extruded using an eightfold area reduction die to $\varnothing 30\text{mm}$ bars at the deformation ratio of ~ 9 . Following that, 50mm bar sections were cut and extruded further to 1 mm(*thickness*) $\times$ 20 mm(*width*) strips at the deformation ratio of 408 [131]. The deformation parameters and the mechanical properties are given in Table 3.4.

Table 3.4: Extrusion parameters and properties of the sample before and after extrusion [131].

Sample dimensions	Extrusion parameters			
	Ratio	True strain	Temperature ($^{\circ}\text{C}$)	Speed (mm/min)
$\varnothing 30\text{ mm bar } 12.2.5.350$ 600	9	2.5	350	600
1.0 mm $\times$ 20 mm strip	408	6.1	415	1.27
	Mechanical properties			
	Yield stress (Mpa)	Elongation (%)		
$\varnothing 30\text{ mm bar } 12.2.5.350$ 600	94 ± 5	17 ± 0.6		
1.0 mm $\times$ 20 mm strip	129 ± 1.5	17 ± 1		

A small rectangular sample with the approximate size of 1mm(*l*) $\times$ 1mm(*w*) $\times$ 1.5mm(*h*) was cut out of the extruded strip. A typical sample microstructure is shown in cross section in Figure 3.13 comprising angular Ti particles in the Al-(Si) matrix. Larger particles are only slightly deformed whereas smaller particles are progressively deformed into long, continuous fibres (Fig. 3.14).

Tomography procedure - Beamline Setup

The hard X-ray micro-tomography setup available at the TOMCAT beamline at the Swiss Light Source (SLS) was used. The beam was monochromated to 20keV, and a 20 μm thick scintillator (type: LuAG:Ce) was used as a trade-off between detection efficiency and spatial resolution.

An optical objective lens (UPLAPO10x) was employed offering a $1.5 \times 1.5 \text{ mm}$ field of view and $0.74 \times 0.74 \mu\text{m}$ nominal pixel size. The exposure time was adjusted to 700ms for optimum contrast, and 1200 projections were collected at an angular step of 0.15° from 0° to 180° . Tomographic reconstruction was carried out using the "Gridrec" Fourier based tomographic reconstruction software package available to the users of beamline TOMCAT at SLS [138].

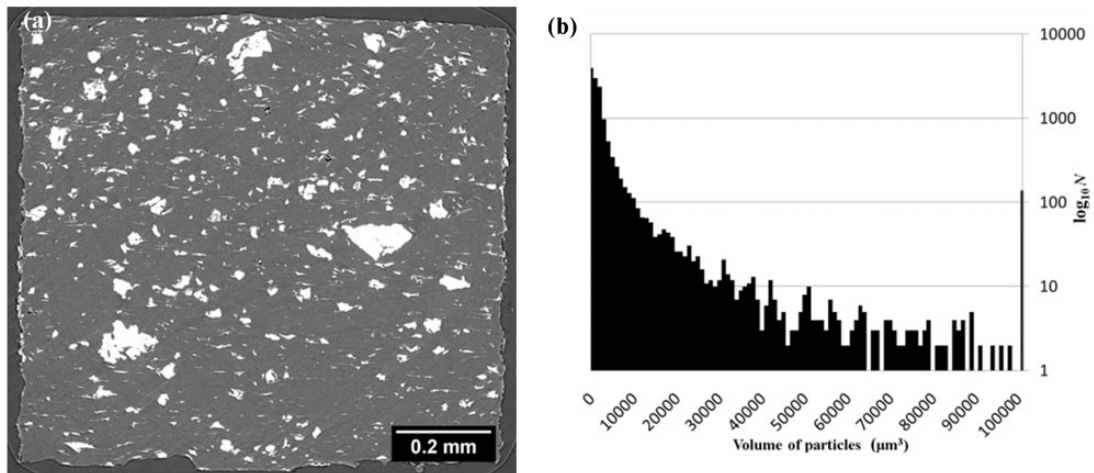


Figure 3.13: (a) Tomographically reconstructed cross-section. Ti reinforcement phase is shown as white patches in the grey Al matrix, (b) Histogram of the volume size (μm^3) of Ti particles (N) within the specimen.

For *in situ* loading, the sample was mounted within a custom-made manual-loading rig, and tomography data sets were collected at 10 compression increments (denoted $N=0$ to 9). The rig did not allow measurement of the sample load, but the displacement of the compression platens could be readily determined from radiographic data. Sections of the tomographic acquisition at different increments are shown on Figure 3.15. The total nominal applied compressive strain was 2.6%, calculated from the bulk change in sample dimension. A 3-D rendering of the specimen is shown in Figure 3.14. Image rendering of the 3-D tomographic reconstruction reveals that Ti inclusions are elongated in the direction of extrusion (Table 3.4) that also corresponded to the principal loading direction. The overall dimensions are shown, together with several cross-sections at various cutting angles across the specimen. The lower half of the image contains a colour rendering of the Ti inclusions that was done in order to provide a better illustration of the size distribution (for quantitative information see Fig. 3.13) and their random dispersion within the bulk.

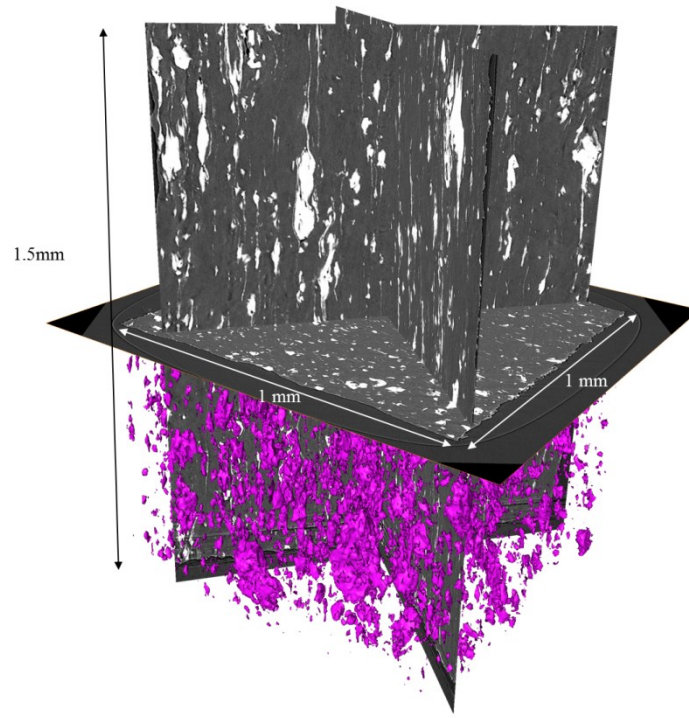


Figure 3.14: 3-D image rendering of the Al-Ti metal composite. Ti reinforcement phase is shown as white patches in the grey Al matrix above the horizontal section, and as coloured 3-D “inclusions” below.

Digital Volume Correlation

The DVC evaluation performed in the present study was carried out using DaVis StrainMaster 8.0 [105] software package. Each dataset comprised $1789 \times 1609 \times 2048$ voxels. An initial correlation window of size $512 \times 512 \times 512$ was used, followed by windows of $256 \times 256 \times 256$, $128 \times 128 \times 128$, $64 \times 64 \times 64$ voxels, and finally $32 \times 32 \times 32$ voxels. 50% overlap was used at all levels, meaning that the minimum spacing of the underlying grid for displacement evaluation was less than $12\mu\text{m}$.

Two strategies could be adopted to analyse the displacement data from the multi-step *in situ* loading experiment. In the first approach, each pair of consecutive increments would be correlated (referred to as the “N/(N+1)” correlation), and the final displacement calculated by accumulating the displacement at each increment. Alternatively, each increment can be compared with the initial or reference (unloaded) dataset (the “1/N” correlation). The former can be done with smaller window size, since the deformation between two consecutive increments is small. However, this approach may also accumulate the numerical errors at each stage. The “1/N” correlation needs to be performed with a larger window size, since the

deformation between the two correlated images can be very substantial, and better statistics are required to reduce the numerical error. However, in the case of plastic deformation, the strain calculated from the latter strategy may be incorrect, as the strain from each correlation is calculated simply from the gradients of the relative displacements, which does not lead to correct values of true strain in the case of large deformation analysis.

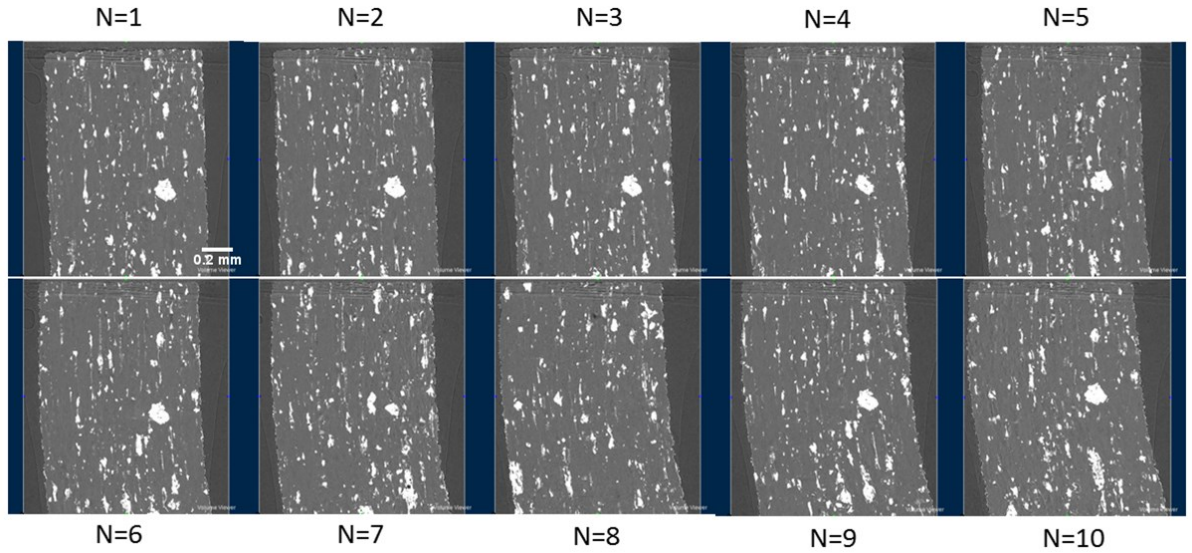


Figure 3.15: Tomographic reconstruction of the central vertical cross-section for consecutive loading increments: N=0 to 9, the width of the specimen is approximately 1 mm.

Both strategies were followed to analyse the N=9 loading stage with respect to stage N=0. It was observed that the calculated displacement fields are similar for both cases (less than 10% difference). An analysis using $256 \times 256 \times 256$ window size was followed by the analysis using $64 \times 64 \times 64$ window size for both strategies. Unlike displacements, the strain fields were found to deviate from the second loading stage (N=2) onwards, so that the final difference of up to 70% between the two strategies could be found. Therefore, the former strategy (the incremental “N/(N+1)” approach) was used hereafter. The results of the DVC method for all the loading increments are illustrated in Figure 3.16 at the same cross section as the tomographic data (Fig. 3.15).

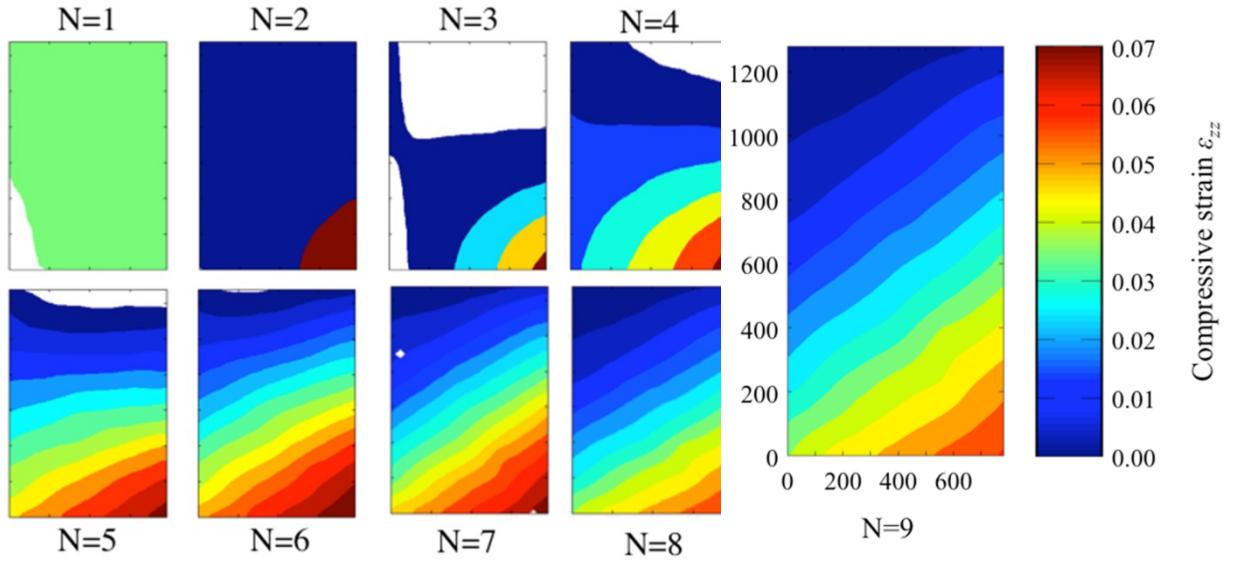


Figure 3.16: Consecutive loading stage displacement field obtained from DVC. Dimensions are in given in μm (overall sample width is approximately 1mm).

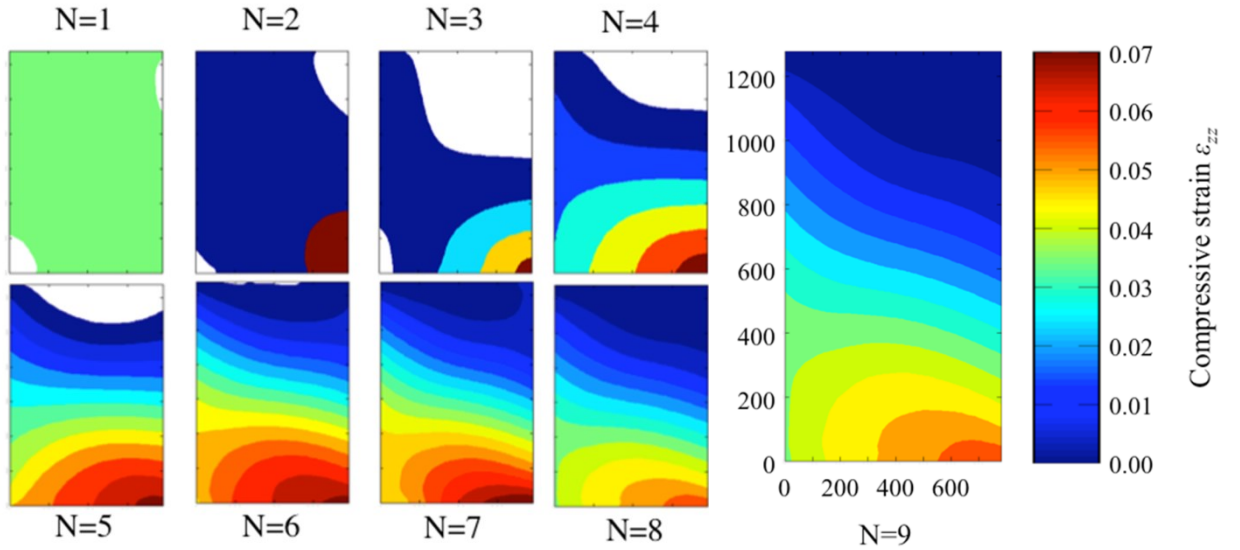


Figure 3.17: Consecutive loading stages from ABAQUS FE model. Sample dimensions are given in μm (sample width approximately corresponds to 1mm).

3.4.2. Finite element modelling

A 3-D FE model was used to simulate the behaviour of the specimen under consecutive loading increments. The multi-axial nature of loading allows to examine the relationships between the boundary conditions of the deformation and the distribution of strain (including the plastic strain part). The model used consisted of a total of 58800 8-node brick elements. The Al alloy matrix was treated as isotropic and homogeneous, with Young's modulus and Poisson's ratio of 67 GPa and 0.33, respectively, i.e. within the typical range for aluminium-silicon alloys

[139]. The Ramberg-Osgood [140] material model was used to describe the strain hardening properties of the Al-12Si alloy + 12Ti particulate composite. The Ramberg-Osgood equation [133] can effectively describe the non-linear relationship between stress and strain in materials that deform plastically beyond the yield limit. Knowledge of the detail of the hardening mechanism was not required for the continuum level approach, but is likely to be primarily due to strain hardening and morphological changes in the Al [131]. The hardening behaviour of specific materials can be recovered by alternating the Ramberg-Osgood exponent ν matching experimental observations to the FE model simulation.

The “true” original state of the specimen was not available, due to some inevitable deformation of the specimen that may have occurred during sample machining and mounting in the rig to ensure stable positioning with respect to the beam during subsequent compression. To reflect the fact that plastic flow onset was effectively immediate upon loading, the yield offset in the FE model was set to a very low value (1MPa), together with strong hardening. The specimen was meshed so that the nodes were collocated with the displacement vectors calculated from DVC. In order to obtain satisfactory agreement with observations, a single model parameter was varied, namely, the Ramberg-Osgood exponent ν . The loading conditions on the sample were reproduced within the FE model by applying the displacement boundary conditions obtained from DVC analysis of the top and bottom boundaries of the sample. This allowed direct comparison to be made between DVC and FEM. The results of the FEM method for all the loading increments are illustrated at the same cross section as the tomographic data (Fig. 3.15) on Figure 3.17.

3.4.3. Results - Discussion

Macroscopic examination

Examples of central vertical slices through the tomographic reconstruction of the sample at consecutive observations $N=0$ to $N=9$ are shown in Figure 3.15. The Ti reinforcement phase exists in the form of thin fibrils elongated along the extrusion direction, as well as some more

bulky Ti particles that have not been elongated during extrusion, to which we will refer to as “blobs”. These observations of the microstructure are in good agreement with scanning electron microscopy (SEM) observations by Kelly *et al.* [131] of the same material. The tomography data show clearly that the specimen experiences a combination of bending and compression during loading. The deformation is not uniform within the sample. This is due to minor misalignment of the loading anvils, and the presence of friction between sample surface and the anvils that leads to sideways buckling and plastic bending of the sample. This effect becomes particularly visually apparent from N=4 onwards, and continues to develop further up until the last loading increment (N=9). Additionally, some rigid body displacement was observed, causing the specimen to shift across the field of view, particularly at the higher loading increments. This is likely to have occurred due to compression platens being not entirely parallel, causing the sample to be loaded in a wedge-like fashion, with it being gradually squeezed out to one side. Detailed analysis of the orientation of the top and bottom platens during loading revealed that the angles between them remained in the range 1° to 1.5° , with slight changes during loading. The sample plastic deformation was quantitatively assessed on the basis of the DVC results (3.17), where examples of the measured displacement fields are shown for the same vertical slice within the sample. The displacements are measured relative to one point at the top surface of the specimen. Rigid body movements and sample rotation were accounted for in the DVC analysis.

The displacements calculated from the FE model for the same vertical slice, using the same reference point, are shown in Figure 3.17. Visual comparison with Figure 3.16 shows that satisfactory agreement is achieved between the patterns of displacement fields from the DVC and the FE model, when the Ramberg-Osgood exponent of $\nu = 9$ was used. The histograms of strains from the two analyses are compared in Figure 3.18, for compressive strains developed at different loading increments.

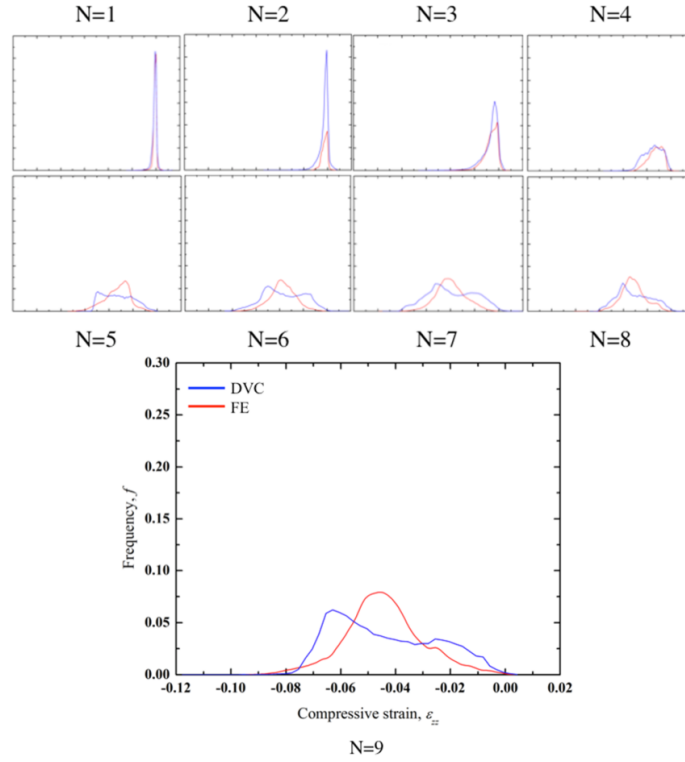


Figure 3.18: FE and DVC histograms of compressive strain (ϵ_{zz}) distribution at different loading stages.

The mean value and the range of maximum principal strains across the entire sample at each loading stage are shown in Figure 3.18 that incorporates the results calculated by DVC and from the FE analysis. At the continuum level, the results are very similar for DVC and FE. However, the distribution of DVC compressive strains is wider. It is interesting to observe that the data are most consistent with a homogenous continuum representation of the material (as assumed in the FE model) in the earlier loading stages (N=1 to 4) in Figure 3.18. For the higher loading stages, the strain histogram obtained by DVC has two peaks, in contrast to the single-peak FE result. This behaviour is consistent with the heterogeneous deformation of the aluminium matrix and harder titanium particles, which is not captured by the continuum FE model.

As shown in Figure 3.19, at any load increment there is a range of strains within the sample. Figure 3.20 (a) presents the data from all load increments for the strain ratio, i.e. the ratio of the local principal strains measured by DVC (i.e. the local values of “Poisson’s ratio under plastic conditions”). The values of the strain ratio mostly lie between 0.4 and 0.5. The negative ratio of the total transverse strains to the total axial strain under the conditions of plastic deformation

is expected to approach 0.5 progressively closer as the plastic strain dominates the elastic part. Data for the median strain ratio as a function of loading increment is shown in Figure 3.20 (b). The median strain ratio rises towards a constant value of around 0.5, which is consistent with the progressive spread of plastic deformation as the deforming sample accommodates the initially misaligned loading.

As shown in Figure 3.19, at any load increment there is a range of strains within the sample. Figure 3.20 (a) presents the data from all load increments for the strain ratio, i.e. the ratio of the local principal strains measured by DVC (i.e. the local values of “Poisson’s ratio under plastic conditions”). The values of the strain ratio mostly lie between 0.4 and 0.5. This is as expected, since the value of this parameter for an incompressible plastically deforming solid is 0.5. Data for the median strain ratio as a function of loading increment is shown in Figure 3.20 (b). The median strain ratio rises towards a constant value of around 0.5, which is consistent with the progressive spread of plastic deformation as the deforming sample accommodates the initially misaligned loading.

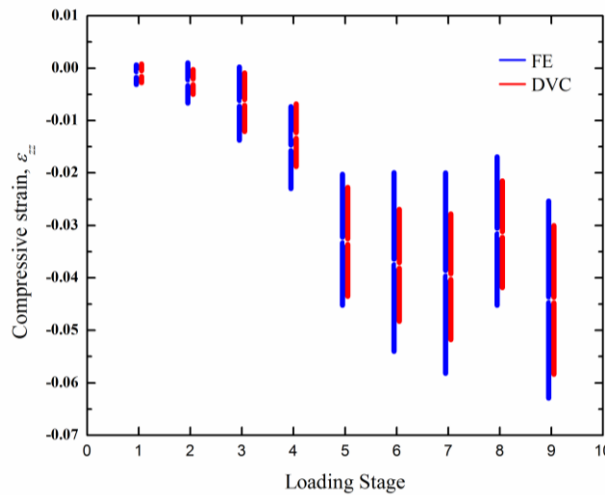


Figure 3.19: The ranges of compressive strain across the entire sample at each loading stage calculated by DVC and compared to FE modelling results. The average value corresponds to the gap in the bar.

The observations reported here provide confidence that DVC applied to the tomographic images is capable of measuring specimen deformation with sufficient reliability and accuracy to capture the evolution of sample plastic flow. However, DVC is also able to reveal the heterogeneous nature of the deformation within the two-phase microstructure considered. This

is discussed further in the following section. Presently we note that, with appropriate measurement of applied loads, the combined DVC-FE approach may allow the extraction of unknown bulk mechanical properties (i.e. elastic moduli and strain-hardening behaviour) from the observation of specimen response under complex deformation. This may be particularly useful for small specimen testing of heterogeneous materials or structures with graded microstructures.

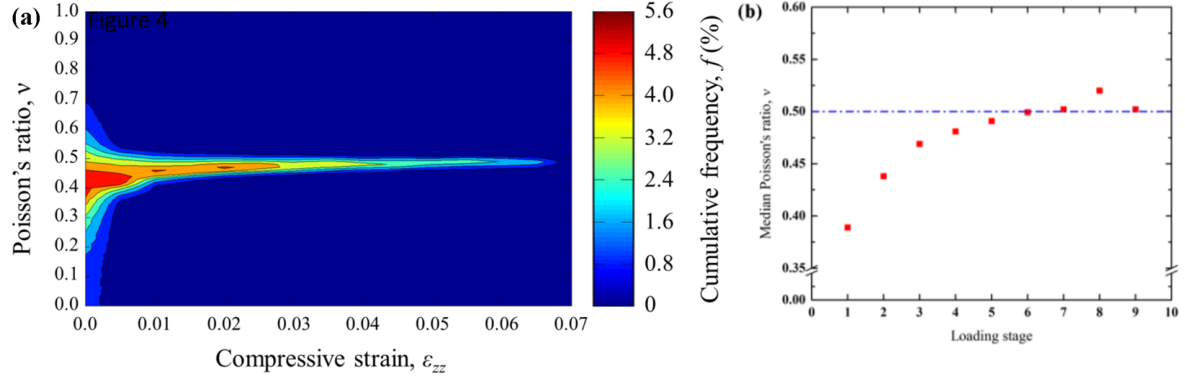


Figure 3.20: Data for Poisson's ratio calculated directly from DVC results; (a) the evolution of the histogram of local Poisson's ratio as a function of the average axial compressive strain, (b) the evolution of the median Poisson's ratio as a function of the loading increment number

Multi-scale analysis of deformation fields

In the analysis presented so far we concentrated on the evaluation of deformation at the length scale of fractions of a millimetre, whereby a Representative Volume Element (RVE) chosen would contain multiple larger and medium-sized phases that serve as helpful markers for displacement determination. This mode of analysis corresponds naturally to the homogenisation of the composite into a uniform continuum that is modelled appropriately as a single medium using FE. However, often it is precisely the local scale inhomogeneities of deformation that are of great interest, e.g. local strain fields induced by the flow of softer matrix around more resistant phases in the reinforced composites. This mode of analysis reveals microscopic strain oscillations at the short length scales on the order of $10\mu\text{m}$.

In the material considered in the present study, the regions of the Ti phase ranged in size from the larger ones of $\sim 100\mu\text{m}$ diameter down to the smallest ones that could be registered by the present technique of only $\sim 1\mu\text{m}$ in size (Fig. 3.13). The smaller sized Ti particles, along

with density fluctuations in the matrix from non-resolved inclusions, served to provide contrast for micro-scale DVC analysis.

Figure 3.21 illustrates the level of detail that could be attained using this approach. It shows a region of interest within the sample that contains a few larger Ti particles (“blobs”) surrounded by a cloud of smaller “dots”. DVC evaluation of spatially resolved displacements could not be accomplished inside the larger inclusions, since these appeared fully saturated in absorption contrast radiography images. However, the overall average strain in each direction could be computed by tracking the displacement of the phase boundary, determining the change in size along each coordinate axis, and then working out the average strain values. Let the phase occupy the domain $\Omega(x,y,z)$. Domain identification is readily achieved by thresholding, i.e. placing in the 3-D matrix of voxels a unity if the voxel finds itself inside the inclusion, and zero otherwise. Additionally, smaller particles were eliminated from the vicinity of the main inclusion by means of mapping the particle size distribution.

Assuming the inclusion has uniform density, the position $r_c = (x_c, y_c, z_c)$ of its centre of gravity can be computed as follows:

$$x_c = \frac{\int \Omega(x, y, z) x dV}{\int \Omega(x, y, z) dV}, \quad y_c = \frac{\int \Omega(x, y, z) y dV}{\int \Omega(x, y, z) dV}, \quad z_c = \frac{\int \Omega(x, y, z) z dV}{\int \Omega(x, y, z) dV}, \quad (3.6)$$

where all integrals are taken over the entire volume of the inclusion.

If the same operations are performed on the deformed dataset, the new position of the centre of gravity r'_c is then found. Applying the uniform displacement equal to $r'_c - r_c$ would bring the inclusion image in the reference data set into “registration” (in terms of the centre of gravity) with its new position after deformation. However, the overall fit will be deficient, due to the inclusion shape change. In order to account for this effect, the following transformation from the point coordinate in the reference data set r to its position in the deformed data set r' needs to be applied:

$$r' - r'_c = (I + \varepsilon)(r - r_c), \quad (3.7)$$

where I and ε are the unit tensor and the strain tensor, respectively. A simplification that we apply in the present case is to assume that the strain tensor ε is diagonal, i.e. contains only three principal direct strains; and that the strain remains constant within the inclusion.

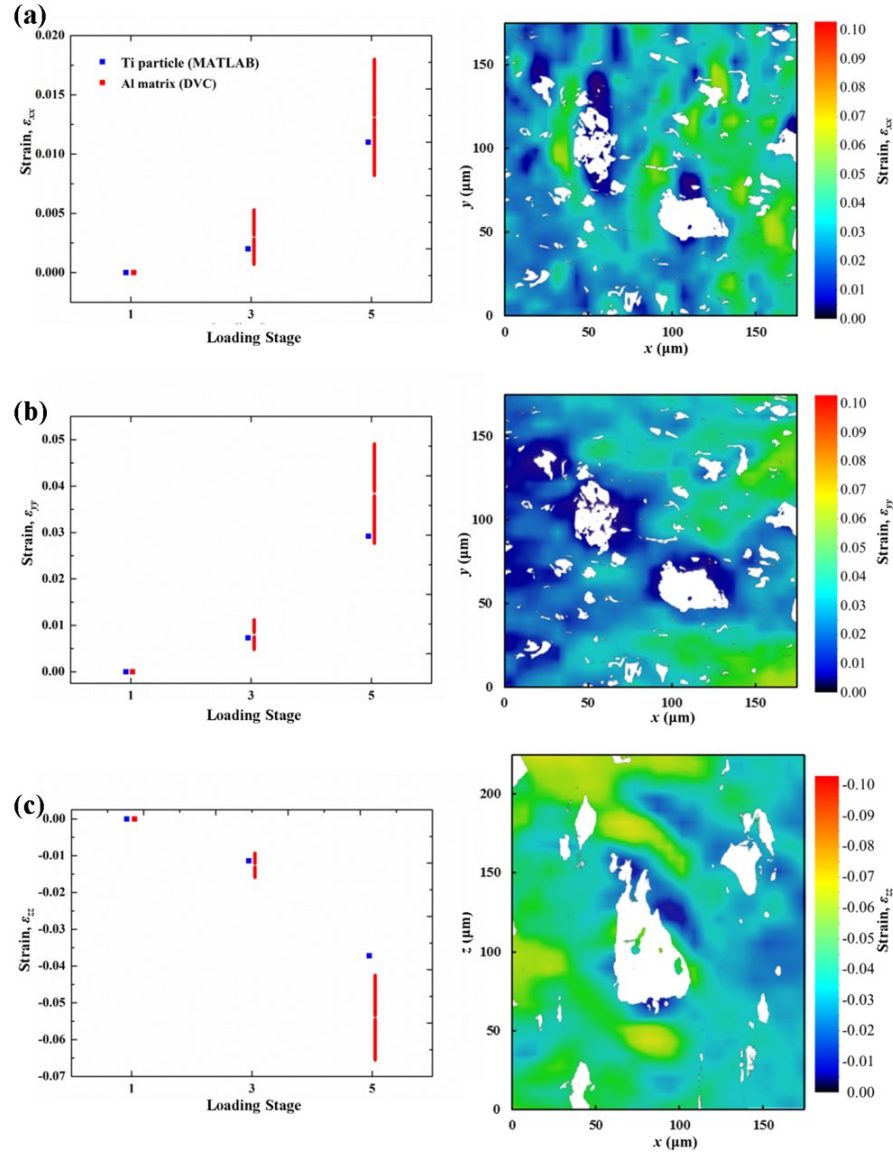


Figure 3.21: Principal strain representation of local deformation results computed with DVC software on the composite's matrix and MATLAB[®] implemented algorithm on the Ti “blobs” (left hand side). Equivalent contour plots of local strain field computed by DVC superimposed on absorption contrast (white indicates dense inclusions) tomographic cross-sections showing contour plots of strain components (a) ε_{xx} (b) ε_{yy} and (c) ε_{zz} .

A procedure employed to determine the best-fitting values of the three strain components for the inclusion can be called “strain correlation”. It consists of applying the transformation given above to the initial dataset, and computing the correlation between the result and the deformed dataset. The values of strain components that maximise the correlation function are taken as the average inclusion strains [141]. Alternatively, the average inclusion strain can be

evaluated by simply computing the weighted average over the inclusion of the elongation for each sequence of pixels extended, e.g. in the z direction (for the z -component of direct strain).

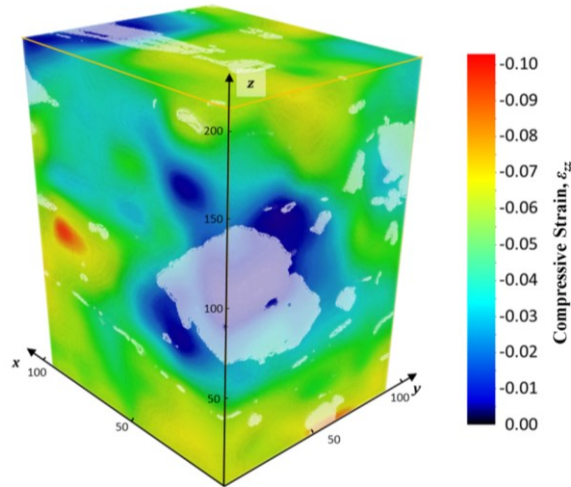


Figure 3.22: Principal strain representation (ϵ_{zz}) using 3-D contour rendering superimposed on absorption tomography reconstruction.

DVC analysis of the matrix strain fields led to the results shown in Figure 3.22. The strain localisation that takes place in the vicinity of the larger particle is apparent. We took care to ensure that this was not a boundary effect, i.e. that the displacement calculation was not affected by the overlap between the inclusion and the control window used by the DVC algorithm. The length scale of the strain heterogeneity is large compared to the control window.

3.5. Conclusions and remarks

This chapter presented an account of the application of advanced synchrotron X-ray tomographic imaging and DVC procedures to the analysis of *in situ* deformation. The method applications reported were as follows.

- The 3-D multiscale deformation behaviour of a co-sprayed metal-metal Al-Ti composite was studied. In order to achieve the best match with the experiment, realistic boundary conditions were applied to a continuum FE model, and results were compared with the observation. At the homogenised, continuum level, satisfactory agreement was achieved between the DVC results and FE model predictions. It was also shown that the analysis of microscopic displacement fields around individual inclusions was possible on the basis of multi-scale DVC analysis. The demonstration of determining local strain concentrations by CT-

DVC opens up the prospects of systematic application of this approach to non-destructive analysis of inhomogeneous internal deformation within the bulk of samples of microstructured materials.

- The 3-D crack opening displacements were mapped on polygranular Gilsocarbon graphite during quasi-static fracture, identifying the damage and fracture process zone with a size of approximately 1.5mm. This is consistent with the expected quasi-brittle fracture behaviour of this material. An effect of out-of-plane constraint on the fracture resistance is deduced that is a general characteristic of quasi-brittle materials. A more accurate model would require a cohesive law with parameters that can be made dependent on constraint. 3-D cohesive zone modelling was used to simulate successfully the experimentally observed crack opening and fracture process zone.

Further advancement of tomographic reconstruction modalities and algorithms is necessary based on the growing scientific and engineering need to interrogate real 3-D objects in a non-destructive way, in order to access a variety of important physical and mechanical parameters, such as crystal orientation and quality, lattice distortion and mis-orientation, elastic and plastic components of the strain tensor, etc.

Progress towards these goals is likely to involve X-ray techniques, as these beams of (high energy) photons provide a unique non-destructive probe that is sensitive to all aspects of material's electronic, crystallographic and micro-structure. On the other hand, we expect the development of both experimental techniques and algorithms for measuring and reconstructing ever more intricate and complex quantities that describe material behaviour, both mechanical and functional. The data obtained in this way lends itself naturally to direct validation of modern 3-D simulations of deformation and other related parameters, e.g. grain rotation, fracture, voiding, polarization, etc.

Chapter 4

X-ray diffraction ‘rich’ tomography techniques

The fundamental, crucial advantage of penetrating radiation is that it allows tomographic images to be obtained without sectioning the sample, i.e. in a non-destructive fashion. Experimental data collection is achieved by collecting projections of the same sample repeatedly after sample is rotated incrementally around a fixed rotation axis. Multiple “projections” which contain integrals *along different lines* of e.g. transmitted intensity. Following the collection of such redundant data sets, different reconstruction strategies can be employed to construct a 3-D representation of the examined internal quantity variation i.e. spectral content.

The ability of X-ray penetrative radiation to undergo diffraction by the crystal lattice planes provides an “atomic strain gauge” able to measure the average elastic strains within the scattering volume. It can also provide information regarding the orientation distribution of differently oriented material grains and even probe the deformation and mechanical state of individual grains within the bulk. Many techniques are available for the task of characterisation by diffraction, provided the region of interest concerns the sample surface, or a thin layer that is either supported by a substrate (as in the case of coatings) or is free-standing (as in the case of transmission electron microscopy foils). However, many situations arise, particularly in the context of biomedical, materials science or mechanical engineering studies, when the non-destructive determination of *internal* (bulk) strain distributions becomes desirable, and penetrating radiation (or electromagnetic waves) must be employed [22].

Over the last two decades the use of diffraction of penetrating radiation for strain determination has undergone rapid development. This is associated with a broad range of factors, including the availability of sources and instruments, focusing optics and better detectors [22]. Once the detection systems became available that allowed large portions of polycrystalline diffraction patterns containing multiple diffraction peaks to be routinely collected, procedures were advanced for the determination of average elastic strains using Rietveld refinement [142]. With the advent of high-brightness third-generation synchrotron sources offering higher X-ray beam energies, the measurement of lattice parameters and hence strains became possible in the bulk of engineering components and samples (e.g. Ref. [143]), not only at the surface or in small powder samples [144]. Furthermore, the development and use of energy-dispersive polycrystalline diffraction led to the application of Rietveld and Pawley refinement approaches for the interpretation of multiple peak diffraction patterns in terms of average elastic strains within the polycrystal [145]. The method was also shown to confer significant advantages in terms of flux utilization and the speed of data collection.

The bulk strain data collected at synchrotron instruments provides important insight across the scales into the deformation behaviour of e.g. novel composite materials [143, 146] and polycrystalline aggregates [147, 148]. Alongside the determination of macroscopic average elastic strain, it is also possible to evaluate mesoscale intergranular strains [148]. The peak shape and broadening, on the other hand, are known to be related to the intragranular deformation, and can provide information about the plastic strain and dislocation substructure [149]. The methodology of synchrotron diffraction strain measurement has been successfully applied to spatially resolved mapping of strains around important features, such as fatigue crack tips [150].

In a combined mode of operation, X-ray diffraction can be used in conjunction with conventional tomographic imaging to achieve diffraction contrast and to perform 3-D mapping of the internal microstructure of polycrystalline samples [151]. Alternatively, transmission

energy-dispersive diffraction imaging (TEDDI) can be used to construct 3-D volume data sets, particularly relating to the distributions of crystallographic phases [152].

With the development of higher flux sources and improved focusing capabilities, the usable beam dimensions became smaller, down to sub-micron [153]. The paradigm of powder diffraction then runs into limitations, and needs to be replaced by grain-to-grain interrogation, e.g. by grain scanning and indexing in the monochromatic mode, or by using Laue diffraction in the white beam mode [154]. The application of this approach to the material of interest for the present study has been reported [155].

High-resolution mapping of powder-average internal strains within bulk components remains elusive. For example, point-wise mapping of strain distributions is readily possible with neutron beams, using the approximately equiaxed shape of the scattering volume. However, the best spatial resolution remains of the order of 1mm, due to the limitations of flux and focusing.

Much smaller beam spot sizes are possible at synchrotron sources. However, one of the consequences of the use of high-energy is that the diffraction scattering volume becomes elongated into the form of a lozenge, so that the spatial resolution is degraded along the beam (typically by a factor of 10 or more).

4.1. Tomographic Acquisition Principles

The limitations of the strongly elongated shape of the scattering volume in high-energy X-ray diffraction are described in more detail in Korsunsky et al. [21], where the principle of "*diffraction strain tomography*" (DST) was proposed. The approach makes use of tomographic reconstruction methods applied in the context of imaging in order to obtain 2-D strain maps from redundant, multiple-angle projections of the same object. The original demonstration described in Ref. [21] was carried out for the simplified case of axially symmetric residual strain distribution.

Further reduction of the incident beam size can allow for the individual inquiry of separate grains within polycrystalline aggregates. The process, as described in more detail by Hoffman et al. [36] is based on the ability to capture and eliminate Laue patterns for an individual grain from a collected pattern of a multi-grained material cross-section. A multiple-angle set of projections using transmission Laue diffraction setup is collected. The method is named “Laue orientation tomography” [36], which best summarises the tomographic strategy and the grain-specific intensity that is reconstructed. Preliminary grain reconstruction results, within the bulk, demonstrated the viability and potential value of the proposed principle for the analysis of granular structure in polycrystalline materials.

“Pencil” beam translate-rotate acquisition setup

The fundamental link between the implemented “rich tomography” techniques in the present section lies in the *translate-rotate* technique used during acquisition. It allows the collection of data leading to the familiar visual representation of the raw data in the form of *sinograms* (see Chapter 2). The resolution of the reconstruction obtained afterwards is related to the beam spot size. This is roughly equivalent to the resolution (pixel size) of the CCD used in absorption imaging. For each rotation angle φ in the range of 0° to 180° , the sample is translated across x with a step size equal to the horizontal beam size, over the total width that covers the whole sample (Fig. 4.1 (b)). The same scanning acquisition procedure is followed at all rotation increments. It is important to stress that the use of this acquisition technique is primarily feasible at high “brilliance” 3rd generation synchrotron light sources, which allows for sub-second exposure times.

Figure 4.1 (a) is a 3-D representation of the diffraction geometry, using a collimated “pencil” shape beam and the translate-rotate acquisition approach. The incident beam of $\sim 20 \times 20 \mu\text{m}^2$ (depending on the collimation arrangements and the size of the specimen) illuminates part of the sample and undergoes Bragg diffraction, whereupon the diffracted beam is registered on a 2-D area detector. No collimation of the diffracted beam is used so that the

diffraction pattern arises from scattering by the entire length $L(x, \varphi)$ of the gauge volume. When high-energy synchrotron X-ray beams are used in diffraction experiments, Bragg scattering angles for representative lattice spacing values encountered in structural materials typically become very small ($<15^\circ$). A consequence of this is the appearance of a highly elongated gauge volume that is given by the intersection between the incident and diffracted beams. Typically, the aspect ratio of the gauge volume is of the order of 10:1–20:1 [22].

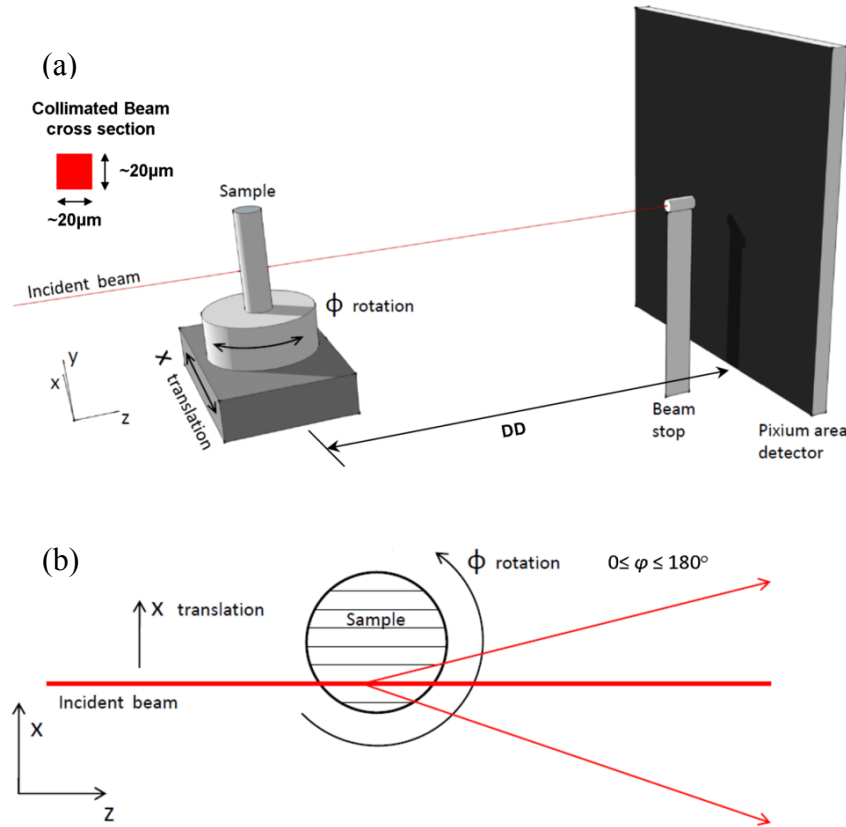


Figure 4.1: (a) Illustration of the scattering geometry used in the strain tomography and Laue orientation tomography diffraction experiments (3-D view of the experimental setup). The incident beam (from the left) illuminates part of the sample highlighted and undergoes Bragg diffraction and is registered on the detector on the right. (b) A schematic representation of the diffraction experiment (plan view of sample cross-section from the top). Data collection is performed by incrementing the rotation angle φ through the range between 0° and 180° in equidistant increments, and scanning the sample across the beam in steps along the x axis at each angle φ [36].

4.2. Powder Diffraction Strain Tomography (DST)

The Diffraction Strain tomography (DST) approach makes use of tomographic reconstruction methods that are conventionally applied in the context of imaging in order to obtain two-dimensional strain maps of unknown strain distributions within cross-sections of arbitrary shaped samples under various loading conditions or residual strain variation. A multiple-angle set of projections of the specimen under interrogation is collected using a monochromatic

powder diffraction setup probing information regarding the interplanar crystal lattice spacing distributions (average strain distribution) across the sample at angles varying from 0° to 180° as described above.

The characteristic of high-energy X-ray scattering configuration, causing an elongated gauge volume presents a problem for spatially resolved diffraction mapping, due to the severe imbalance of spatial resolution in the directions across the beam (high resolution) and along the beam (low resolution). Rotating the sample around the axis perpendicular to the beam could help, but the problem arises when extracting the data at uniformly high spatial resolution from the series of measurements that represent convolutions over elongated sampling volumes.

The situation is similar to that encountered in parallel beam absorption tomography, where the intensity registered by each detector pixel allows the calculation of the total integral absorption of the incident beam by the sample as described by Eq. (2.1). Figure 4.1 (b) shows a schematic representation of the diffraction experiment in-plane view (i.e. in the x - y plane). The translate-rotate experimental acquisition procedure involves the following data collection approach. The outer data collection loop consists of incrementing the rotation angle φ in the range between 0° and 180° in steps typically of $1 - 4^\circ$. The precise choice of rotation step is determined by the anticipated level of detail required in the reconstructed strain distribution. The inner data collection loop consists of translating the sample along the x axis across the beam in steps related to the chosen horizontal size of the collimated beam at each angle φ . The data set collected for each angle φ is thus similar to that contained in a horizontal row of pixels on the imaging detector used in absorption tomography experiments.

However, some important distinctions are present between the method of strain reconstruction tomography used here, and conventional absorption coefficient tomography. Firstly, the set-up ensures that the quantity being “imaged” is lattice spacing (or strain), rather than sample density. Secondly, within standard tomographic data set prepared for reconstruction by filtered back-projection (inverse Radon transform), each individual pixel

value corresponds to the path integral of the unknown quantity along the beam path length, $L(x, \varphi)$. This is indicated by the expression for the data element corresponding to each particular pixel given in Eq. (2.1). In contrast, each individual pixel value in the experimental data set collected in our diffraction experiments corresponds to the path average $\langle \alpha(x, \varphi) \rangle$ of the unknown parameter (strain) along the path length $L(x, \varphi)$, i.e.

$$\langle \alpha(x, \varphi) \rangle = \frac{\int_{L(x, \varphi)} \alpha(x, z) dz}{\int_{L(x, \varphi)} dz} = \frac{\int_{L(x, \varphi)} \alpha(x, z) dz}{L(x, \varphi)} \quad (4.1)$$

Therefore, in order to prepare the data in the form suitable for filtered back-projection, the experimentally measured data must be converted from the average to integral values, i.e. treated as follows:

$$p(x, \varphi) = \int_{L(x, \varphi)} \alpha(x, z) dz = \langle \alpha(x, \varphi) \rangle L(x, \varphi) \quad (4.2)$$

In other words, even though the resolution in the direction along the beam is infinitely poor, the collection and interpretation of redundant diffraction data sets at a sufficient number of sample rotations allows the 2-D distribution of density to be reconstructed. This can be accomplished by proving the key relationship between the *overall lattice parameter* obtained from experimental measurements and the *average lattice parameter* within the gauge volume.

Average lattice parameter numerical representation

The key hypothesis that lies in the above mentioned relationship between overall lattice parameter, deduced as a result of scattering from an elongated volume, and the "real" average lattice parameter, contained in the bulk of the gauge volume needs to be evaluated and confirmed. In this formulation Eq. (4.2) can be used in a good approximation to reconstruct lattice parameter in a cross-section using the same filtered back-projection strategy as in conventional tomographic imaging.

Overall precise determination of the sample detector distance (SDD) using tomography and knowing the distance from the peak centre to the straight-through beam (AB) allows the precise application and correction of the average strain value that is initially calculated by the fitting routine (Fig. 4.2). Imprecise determination of these distances (micron scale) has a deleterious effect on the correct determination of microstrain. As an example in the case of sample detector distance $SDD = 500\text{mm}$ and over a $2\theta \sim 10.2^\circ$ scattering angle a $500\mu\text{m}$ SDD miscalculation will result into ~ 1000 microstrain ($= 1000 \times 10^{-6}$ strain) error.

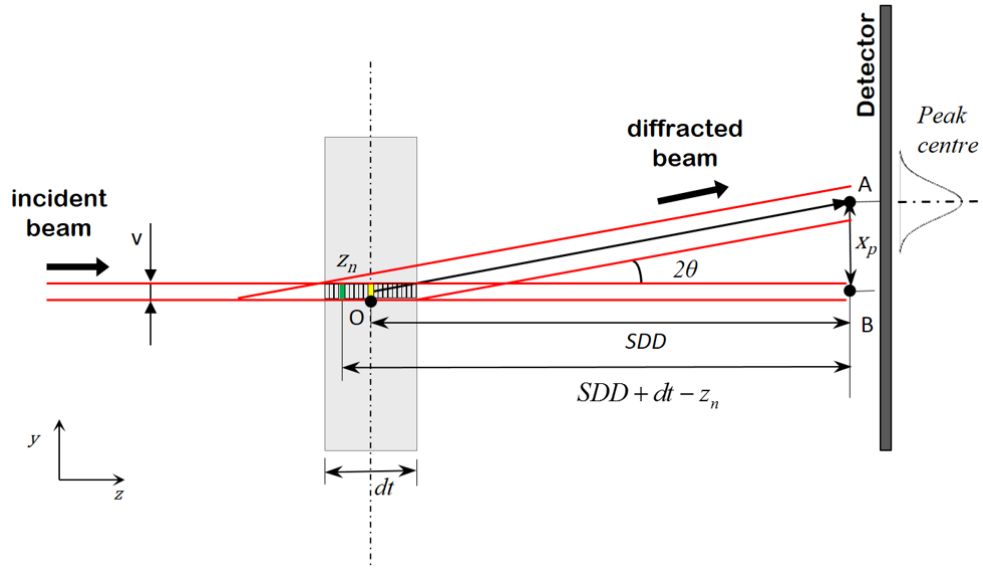


Figure 4.2: Schematic of the average strain calculation simulation.

Simulations for different strain distributions across the sample have been carried out to monitor the evolution of fitting error. In this context a Gaussian peak is representing the diffraction pattern (i.e. Debye-Scherrer ring) registered on the detector as a result of the beam interaction with a cuboidal ($0.1 \times 0.1 \times 0.1\text{mm}$) gauge volume in the middle of the beam path (indicated with yellow in Fig. 4.2). This result can be directly compared to a peak generated from accumulating (n) Gaussian peaks from individual points $n = 1 \dots N$ along the path of the beam $-dt/2 \leq z_n \leq dt/2$. The lattice parameter distribution $f(z_n)$ of these points can be considered to be an arbitrary function.

The distance from a random scattering point located within the gauge (i.e. illuminated) volume to the detector and the distance (x_p) from the diffraction peak centre (A) to the straight-

through beam (B) can be related through a simple geometric relationship via diffraction scattering angle 2θ :

$$x_p = \left(SDD + \frac{dt}{2} - z_n \right) \tan 2\theta \quad (4.3)$$

The corresponding peak $g(z_n)$ as registered on the detector, as a result of scattering from a random point within the gauge volume, can be represented by a Gaussian peak distribution based on the lattice parameter and the experimental setup:

$$g(z_n) = y_0 + h e^{-\frac{(x-x_p)^2}{2c^2}}, \quad n=1 \dots N \quad (4.4)$$

where y_0 is the background noise (intensity counts), h indicates the peak height and c indicates the Gaussian width.

Taking into account the contributions of individual $g(z_n)$ diffraction peaks across the illuminated volume, the overall peak registered by the detector (G) can be reproduced using the formulation $G = \sum_{n=1, \dots, n} g(z_n)$, where n is the amount of discretisation steps within the bulk.

A least squares fitting method can be then used to fit the simulated peak and calculate the *overall lattice parameter* (a_G) corresponding to the simulated experimental data. The calculated *average lattice parameter* as a result of the lattice distribution function $f(z_n)$ is set as a_0 . The relative difference between a_G with a_0 can be defined as $s_1 = (a_G - a_0)/a_0 \times 10^6$ and describes the corresponding error in *microstrain* units. Table 4.1 lists the resulting error (s_1) for various simulated input strain distributions. The strain ranges for each variation are given values typically encountered in diffraction experiments within the elastic deformation limit.

The maximum error (s_1) value was determined for a step function at 10 microstrain that can be considered as negligible. The results demonstrate the validity of the hypothesis and confirm that the overall lattice parameter represents the average strain value within the sampling volume $\langle \alpha(x, \varphi) \rangle$ appearing in Eq. (4.2).

Table 4.1: Parameters used for the average strain distribution simulation, together with their strain range and the calculated error.

Distribution	Lattice Distribution function ($f(z)$)	Strain range	s_1
Constant	$a = 4.01$	-	-3.481×10^{-6}
Linear	$a = -0.001z + 4.01$	$[-436.4, 436.4]$	-0.4130
Quadratic	$a = 7.85 \times 10^{-4} z^2 + 4.01$	$[-210.89, 388.48]$	-7.7255
Step	$a = 2 \cdot 7.85 \times 10^{-4} \text{sign}(z - dt / 4) + 4.01$	$[-195.95, 587.86]$	-10.7429

The following postulate can be formulated on the basis of the above consideration:

The lattice spacing integral over the entire cross-section remains constant for all values of φ .

For each gauge volume, the average lattice spacing referred to in the postulate is given by Eq. (4.1), i.e. involves averaging along the gauge volume in the y direction. Now consider the lattice spacing integral over the entire cross-sectional area Ω . Using Eq. (4.2), this can be written as:

$$\int_{\Omega} a(x, z) dz dx = \int_W \left(\int_{L(x, \varphi)} a(x, z) dz \right) = \int_W \langle a(x, \varphi) \rangle L(x, \varphi) dx \quad (4.5)$$

where W denotes the scan width in the x direction. Replacing the last integral by a sum over the experimental data, we get:

$$\int_{\Omega} a(x, y) dy dx \approx \sum_{i=1 \dots N} \langle a(x_i, \varphi) \rangle L(x_i, \varphi) = A(\varphi) \quad (4.6)$$

where $i=1 \dots N$ denotes the measurement number within the scan performed at angle φ . The value on the left-hand side represents the integral of the lattice spacing over the entire cross-section, and must remain constant. Hence, the expression on the right-hand side (denoted $A(\varphi)$ and formed from the experimental data) must also remain constant for different values of angle φ . This postulate provides a useful validity check for the experimental data set. Deviation of the measured data from constant value indicates the presence of systematic errors that must be corrected prior to tomographic reconstruction.

"Strainogram" generation

So far the specifics of tomographic "pencil" beam acquisition procedure have been presented and the *overall lattice spacing* tomographic formulation in Eq. (4.3) has been validated setting the base for the experimental implementation of diffraction strain tomography. As described in Chapter 2 a typical processing stage, following data acquisition in an absorption tomography mode, is the creation of the *sinogram*. The paraphrase in the title indicates DST apparent lattice variation data, following tomographic acquisition, are given in an analogous formulation named "*strainogram*", which contains the average lattice parameter information for each scan point (x , ϕ). This section describes the diffraction data acquisition and formally addresses the geometry effects and error correction according to Baimpas et al. [23].

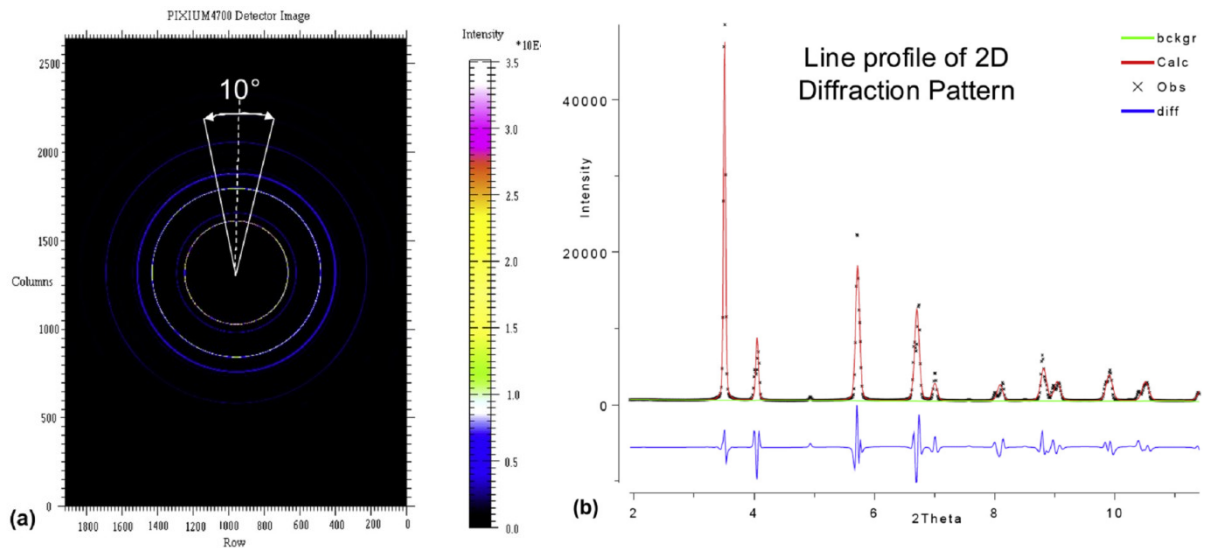


Figure 4.2:(a) The pattern of Debye–Scherrer rings recorded from yttria-stabilised ZrO_2 sample. (b) The equivalent line profile (intensity vs. scattering angle 2θ). The pattern was extracted from the diffraction pattern by "caking" over a 10° "slice" (see text below) illustrated in (a). The Pawley fit to the data is illustrated by the red curve.

The high energy monochromatic beam impinging on the sample at different positions results in scattering into a series of Debye–Scherrer rings registered by a 2-D detector, as illustrated in Figure 4.2. Each ring corresponds to a statistical distribution of grains at a specific crystal orientation within the illuminated bulk that fulfils the Bragg condition for a specific wavelength of incident beam. The increasing angle of diffraction is analogous to the decrease

of reciprocal lattice spacing calculated according to $d_{hkl} = a/\sqrt{h^2 + k^2 + l^2}$ for simple bcc and fcc crystal lattice, where hkl are the corresponding Miller indices.

The diffraction patterns (Debye–Scherrer rings) registered by the detector are then analysed using Fit2D (2-D crystallography diffraction data analysis software, ESRF [144]) for the purposes of radial-azimuthal binning that is commonly referred to as “caking” (Fig. 4.2 (a)) [142, 143]. “Caking” involves identifying a “slice” in the pattern (in the example shown in Figure 4.2, between -5° and 5° from the vertical direction). The scattering vector defined as the bisector between the incident and diffracted beams corresponded closely to the vertical (y) laboratory axis, so that the strain component measured was in the direction along the axis of rotation of the sample. The data within the selected azimuthal range is then binned into radial ranges to obtain a 1-D radial line profile, and the scale on the horizontal axis was converted to represent the 2θ scattering angle (Fig. 4.2 (b)).

In order to determine the apparent value of the lattice parameter at every (x, φ) position and respective gauge volume, Rietveld refinement is carried out on the 1-D diffraction patterns using GSAS (General Structure Analysis System, [142]). Figure 4.2 (b) provides an illustration of the GSAS fit to a diffraction pattern corresponding to Yttria-stabilized ZrO_2 diffraction pattern. By substituting the calculated lattice parameter (α) into Eq. (1.8) using the reference “strain-free” lattice parameter (α_0), the apparent elastic strain at position x and rotation angle φ can be calculated. Several procedures can be used for determining α_0 which are discussed at each section for specific case studies.

Systematic sources of pseudo-strain (apparent change of lattice parameter) may be present in the experimental setup due to the sample position with respect to the rotation axis, and also due to the effects of sample shape. Correct strain reconstruction requires that these geometric effects be considered, and appropriate error corrections applied. For illustration of how the geometric corrections are calculated and applied, consider a C263 nickel-base superalloy turbine blade.

The complex shape of the blade cross-section poses a number of problems with respect to the data acquisition, as well as calibration and analysis. The blade cross-sectional shape illustrated in Figure 4.3 corresponding to the approximate “envelope” of $\sim 22\text{mm}$ (*width*) $\times$ $\sim 6\text{mm}$ (*depth*).

In order to be able to determine strain within engineering components, precise knowledge of the sample detector distance (*SDD*) is crucial. This is due to the fact that changes in the *SDD* result in the change in the reflection position on the detector. This would lead to apparent strain emerging in the data interpretation, unless appropriate correction is applied. It has already been demonstrated by experimental evaluation and from numerical simulation that, in the absence of secondary collimating slits, the lattice parameter deduced from the interpretation of the diffraction pattern (with the *SDD* defined by the centre of gravity of the scattering volume) corresponds to the *average* value over the illuminated volume [22]. The curved blade shape means that, as the sample is translated in the x -direction, the distance between the centre of gravity of the diffracted volume and the detector continuously changes according to the shape of the blade (Fig. 4.3). Given the size of the blade, this change causes a significant apparent peak shift on the detector. This shift is of purely geometric origin not related to sample strain, and must be accounted and corrected for as part of the *SDD* calibration procedure.

Figure 4.3 defines two geometric parameters that are useful for subsequent analysis. Position A is the centre of rotation used as a global reference. This point defines the nominal Sample Detector Distance, for which the value of SDD_{nom} was obtained from calibration data analysis. Consider the situation illustrated in Figure 4.3, which shows the cross-section centre of gravity (B) being displaced from the stage rotation axis (A). At the reference rotation position ($\varphi = 0^\circ$), let the coordinates of the centre of gravity with respect to the rotation axis be given by $x_c = r_{SDD} \cos(\varphi_0)$ and $y_c = r_{SDD} \sin(\varphi_0)$. If r_{SDD} is non-zero, then the entire cross-section precesses around the rotation axis during data collection. In fact, at an arbitrary rotation angle

φ , the coordinates of the cross-section centre of gravity are given $x_c(\varphi) = r_{SDD} \cos(\varphi_0 + \varphi)$ and $y_c(\varphi) = r_{SDD} \sin(\varphi_0 + \varphi)$.

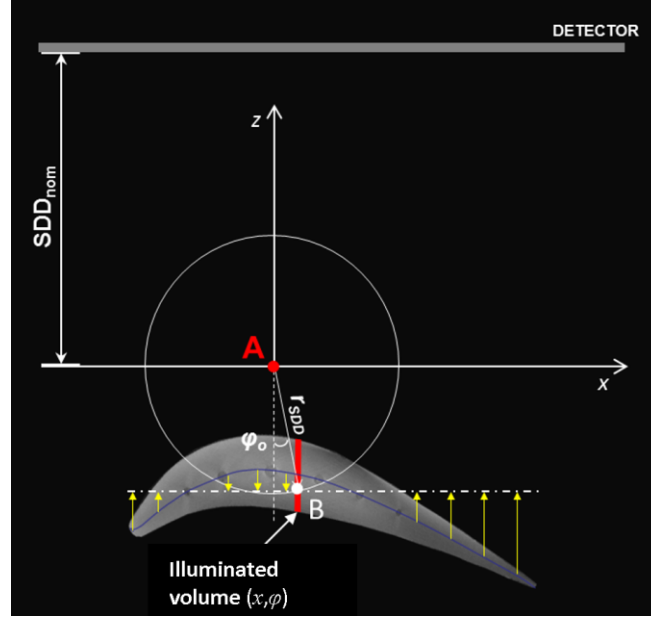


Figure 4.3: Schematic representation of the orbital movement of the Si calibrant (fixed at point B on the blade) the around centre of rotation (point A) showing the initial phase φ_0 , the radial offset with respect to the centre of rotation r_{SDD} , and the sample detector distance SDD_{nom} . Yellow arrows indicate the displacement of the centres of gravity of individual gauge volumes illuminated consecutively during sample translation.

Moreover, at each given combination (x, φ) , the actual centre of gravity of the illuminated gauge volume (B) is offset (as indicated for $\varphi=0^\circ$ by the arrows pointing at the continuous curved line), meaning that $SDD(x_n, \varphi)$ is different from $SDD_{nom} + x_c(0^\circ)$. For that reason an additional line $f(x, \varphi)$ which at each rotation increment φ joins the centres of each gauge volume across the specimen for every x_n position shall be taken into consideration. At different rotation increments the centre of gravity line is recalculated due to the change of the illuminated volume with respect to the incoming beam.

Therefore, the actual sample-detector distance SDD is different from the distance SDD_{nom} from the rotation axis:

$$SDD = SDD_{nom} + x_c(\varphi) + f(x, \varphi) = SDD_{nom} + r \sin(\varphi_0 + \varphi) - f(x, \varphi) \quad (4.7)$$

Analysis of Bragg's law shows that failure to account for this effect would cause a relative change in the apparent lattice spacing α' (pseudo-strain) of:

$$\frac{\alpha' - \alpha}{\alpha} = \frac{r \sin(\varphi_0 + \varphi) + f(x, \varphi)}{D} \quad (4.8)$$

This result represents a systematic shift of the apparent lattice spacing as a function of angle φ :

$$\alpha' = \alpha \left(1 + \frac{r \sin(\varphi_0 + \varphi) + f(x, \varphi)}{D} \right) \quad (4.9)$$

The cross-section lattice spacing integral function $A(\varphi)$ introduced in Eq. (4.7) will be similarly affected, so that:

$$A(\varphi) = A(0) \left(1 + \frac{r \sin(\varphi_0 + \varphi) + f(x, \varphi)}{D} \right) \quad (4.10)$$

The calculation of the cross-section lattice spacing integral function $A(\varphi)$ for each set of measurements at angle φ allows the determination of parameters r_{SDD} (offset of cross-section centre of gravity from rotation axis) and φ_0 (offset angle).

The above discussion illustrates how gauge volume displacement effects on the apparent deduced lattice spacing must be accounted for and corrected for each rotation angle φ and scan position x .

Matrices $\langle \alpha(x, \varphi) \rangle$ and $L(x, \varphi)$ derived from the experimental data after correction for gauge volume centre position are illustrated in Figure 4.4. The data depicted came from the cross-section of the Ni superalloy turbine blade. Figure 4.4 (b) shows the contour plot in the angle-position coordinate axes of the *average lattice parameter* extracted from the diffraction patterns collected. This is readily converted to the average strain along the beam path for each configuration using the usual formula involving the unstrained lattice parameter. Figure 4.4 (a) shows the contour plot (on the same axes) of beam path length $L(x, \varphi)$ extracted from the cross-section shape found from parallel beam synchrotron based tomographic acquisition. Eq. (4.2) provides a recipe for constructing the input sinogram for strain reconstruction.

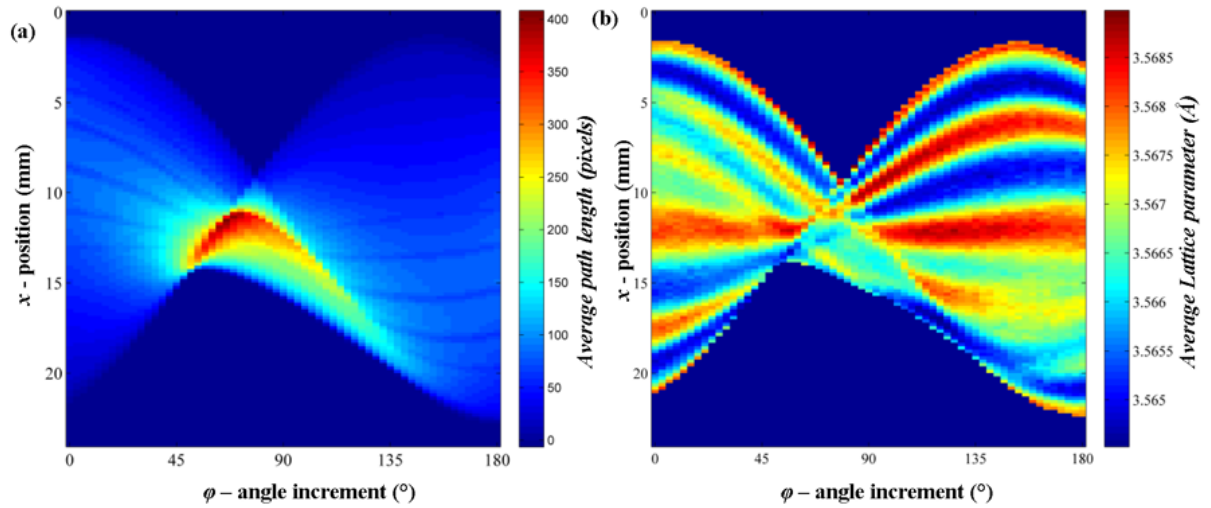


Figure 4.4: (a) Path length diagram ("thickogram") constructed on the basis of conventional tomographic imaging. The horizontal axis represents angular position ϕ ; the vertical axis represents the path length through the sample in the units of mm. (b) Contour plot of the average lattice parameter variation for the Ni blade sample. The horizontal axis represents angular position ϕ ; the vertical axis represents the sample position number and corresponds to the meaning of "pixel" in conventional tomography.

Strain Reconstruction formulation

The linear relationship between average lattice parameter and elastic strain is established by the definition given in Chapter 1 for elastic strain calculation. The apparent lattice elastic strain is computed from the definition $\varepsilon = (\alpha - \alpha_0) / \alpha_0$, where α is the apparent strained lattice parameter and α_0 is the reference "strain-free" value.

Herein after the application of Diffraction Strain Tomography (DST) is illustrated on three case studies with parallel experimental and FE modelling validation of the results. Each case study tackles different aspects of the method applicability and show cases the method use in successful practical implementation of this principle to the study of residual elastic strain distributions and applied strains in different materials and components.

- Firstly, the residual strain distribution in a **Ni-based superalloy turbine blade** fabricated by DMLS (Direct Metal Laser Sintering) is reconstructed using DST. The experiment was conducted on beamline I12 (JEEP) at Diamond Light Source, UK. The aim of the experiment was to obtain detailed information about the sample shape, structure, and state confirming the design and assessing the mechanical processing technique. Tomographic reconstruction along with residual elastic strain deformation and texture information were analysed from the imaging

and diffraction data collected during the experiment. The results were cross-correlated using contour method for residual stress determination, as described in Chapter 2, to confirm the overall residual deformation profile within the specimen.

- Secondly, a **natural hierarchical mineralised tissue structure** consisting of human dental tissues was considered. Namely, the sample contained regions of dentine and enamel, each tissue being a two-level composite structure. The sample was investigated using a multi-phase implementation of the DST analysis. The internal structure of the sample has been previously non-destructively imaged in cross-section using X-ray diffraction computed tomography. Understanding the complex mechanical behaviour related to this hierarchical structure is essential for predicting the effects of microstructural alterations on the performance of dental tissues and their artificial replacements. The experiment was conducted on beamline I22 (JEEP) at Diamond Light Source, UK. The study focused on a structural analysis of human dental tissue via non-destructive synchrotron based experimental DST on Wide Angle X-ray Scattering data (WAXS) and the observation of the out of plane strain variation separately within the enamel and dentine. In addition, a combined semi-destructive FIB-DIC approach was used to assess the in-plane residual strain distribution across the sample at the same cross section. Lastly, a tomographic Small Angle X-ray Scattering (SAXS) dataset also allowed to reconstruct mean HAp crystallite thickness of the dental tissue characteristic nanostructure, further enhancing the knowledge upon grain size distribution within the tooth using the already analysed tomographic reconstruction framework.

- In the third case study, bending strains in sintered polycrystalline zirconia used in the fabrication of **multi-unit fixed prosthetic dentures** were investigated. The experiment was conducted on beamline ID15 (JEEP) at European Synchrotron Radiation Facility (ESRF), France. *In situ* bending loading was applied to a sample of complex shape and practical interest, namely, a multi-unit fixed prosthetic denture (FPD) made from sintered polycrystalline zirconia ceramic. The applied strain was reconstructed at a cross-section of the "bridge" specimen where bending load was considered maximum using DST. Furthermore lab-based CT was used for

data calibration and direct meshing was applied to the tomographic reconstruction that allowed FE modelling and subsequent diffraction data validation for the complex shaped geometry.

4.3. DST on Ni superalloy direct laser deposition (DLD) turbine blade

4.3.1. Materials and applications

Direct Metal Deposited (DLD) parts of nickel-base superalloy C263 are used to replace complex castings and to allow rapid prototyping development of machine elements in industrial applications, particularly in aeroengine manufacture [156]. C263 is the nickel-base alloy that is most commonly used for the application of this method, often also referred to as “Direct Metal Laser Sintering” (DMLS). In Toyserkani et al. [157], the advantages and disadvantages of the major metal deposition techniques were considered, including welding, cladding and thermal spraying. Direct Laser Deposition techniques are very effective in creating very strong bonds between the deposit and substrate, with relatively low levels of distortion. Nonetheless, high thermal gradients are generated during laser heating. These, together with the difference in the coefficients of thermal expansion between the deposited materials and the substrate, lead to the generation of residual stress. Frozen-in plastic strains (eigenstrains) are also present due to the reduction of the yield stress caused by the increased temperature.

The sample is shown in Figure 4.5 mounted on the I12 beamline positioning table. The specimen of the study was a turbine blade made from C263 nickel-base superalloy via the Direct Metal Laser Sintering (DMLS) additive manufacturing route by Materials Solutions Ltd (Worcester, UK), using EOS M270 (EOS GmbH, Munich, Germany). The sample was built up in the vertical direction (as shown in Figure 4.5) onto a steel supporting plate using powder bed procedure. Thin layers ($\sim 20\mu\text{m}$ nominal thickness) of C263 powder (typical particle size $\sim 20\mu\text{m}$) were repeatedly spread over the existing structure. The outline of the area to be deposited at each step was first created by line sintering using a focused solid state laser beam. Subsequently the interior of the outline shape is filled in by repeated “hatching” by the laser

beam, resulting in the solid sample height increment. The built up sample is then lowered by the incremental layer height ($20\mu\text{m}$), the powder layer is re-spread, and the process repeated.

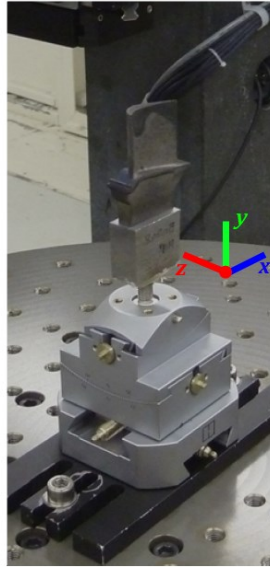


Figure 4.5. Direct Laser Deposited C263 blade mounted on I12 JEEP test bed at Diamond Light Source (DLS).

The resulting material microstructure consists of grains elongated in the direction of sample growth during build-up, i.e. through the individual deposited layers. This is likely to be associated with the fact that at each stage of sintering the newly solidified grains are likely to adopt the crystal orientation associated with the underlying grains solidified at the previous stage of the layer-by-layer build-up process. Overall, objects built-up using DLD additive manufacturing procedures displayed preferred crystal orientation (texture). The $[100]$ pole figure of the sample studied presently is shown in Figure 4.6. This result was obtained by the analysis of monochromatic diffraction data obtained in the experiment described below. The pole figure was derived from “one-shot” measurement by the construction and projection of the Orientation Distribution Function (ODF) [158]. The calculations were carried out using MTEX toolbox for MATLAB[®]. The pole figure shows preferential alignment of $[100]$ plane normal with the growth direction, i.e. the y-axis with respect to the coordinate system defined in Figure 4.5 [159].

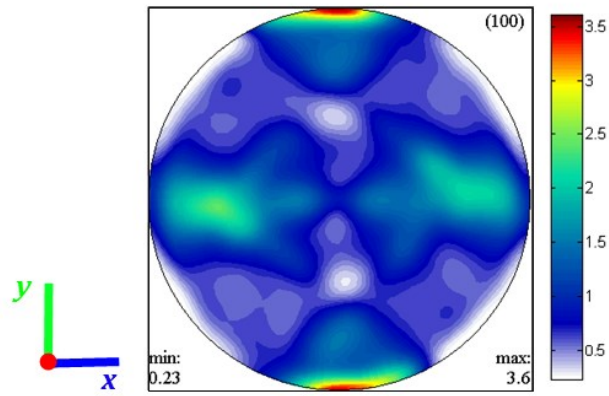


Figure 4.6. Polar figure of preferred orientation for (100) crystal plane on x-y lab coordinates. As a result of the layer after layer manufacturing process, the grain evolution across slices demonstrates severe preferred orientation throughout the bulk of the blade.

The absorption tomography analysis reported below was aimed at establishing non-destructively whether significant isolated pores were present in the built component (it is known that overall density of the material exceeds 99%), and at assessing the surface quality of the inner cooling passages within the blade.

The purpose of DST data collection was to determine, non-destructively, the internal residual strain distribution within the DLD-built blade. As the first step towards tomographic reconstruction of strain, the geometry-induced aberrations caused by the sample detector distance variation are discussed and corrected.

4.3.2. Diffraction and imaging measurements and interpretation

Hard X-ray micro-tomography experiments were carried out at beamline JEEP (I12), the Joint Engineering, Environmental and Processing beamline at Diamond Light Source (DLS, Harwell Oxford, UK). The beam was monochromated to 80keV. For high resolution tomography, a PCO 4000 camera was used with the 4008×2672 pixel matrix.

Four different scintillators and magnification optics (modules) can be used to convert the X-rays to visible light, giving great flexibility of setup in terms of the resolution (pixel size) and field of view. In the present study, Module 1 was used for imaging, with the nominal pixel size of $12.5 \times 12.5 \mu\text{m}^2$ and the field of view of $48.8 \times 32.2 \text{mm}^2$. The size of the blade in the horizontal direction was the reason for choosing this resolution, since it allowed the field of view to accommodate the full width of the blade (21.4 mm) at all rotation increments. Overall,

3600 projections were acquired by rotating the sample in steps of 0.1° over 180° . Parallel monochromatic beam was used, and 2.75s exposure time per image was chosen to match the 16-bit dynamic range of the detector. The 3-D volume reconstruction was performed using filtered back-projection algorithm implemented in MATLAB[®]. The resulting reconstruction was rendered by introducing an intensity threshold, applying Sobel filter, median filter, and masking of the blade using edge detection approaches. The results are illustrated in Figure 4.7.

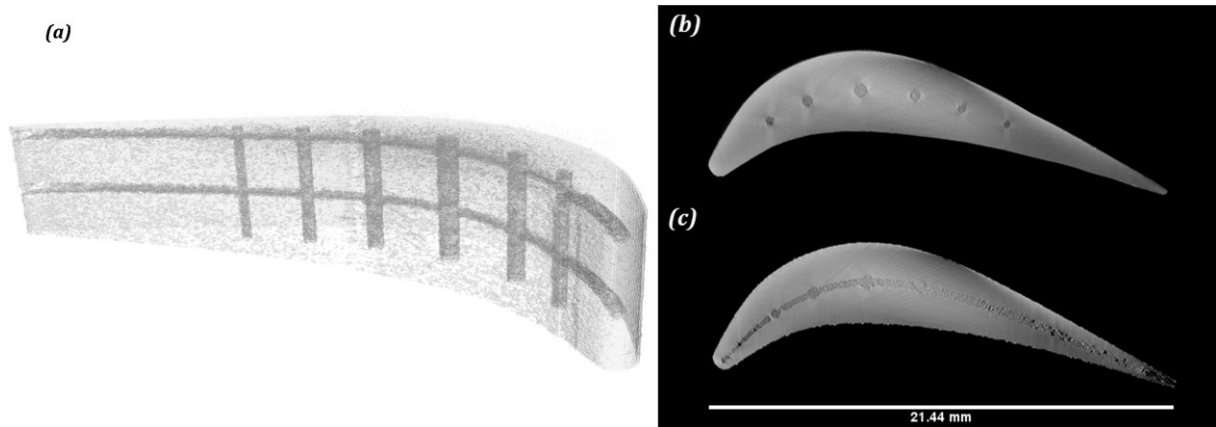


Figure 4.7: (a) 3-D image rendering of the inner channel structure of Ni Blade sample. The inner cooling channel structure is highlighted whereas the overall blade shape is given in an opaque representation. (b) Filtered back-projection tomographic reconstruction of transverse section of the blade showing the cooling channels across the blade, after smoothing and masking operations are applied. Signs of trapped C263 powder residues from the sintering process are present after the component sintering. (c) Tomographic reconstruction slice at the height of a cooling channel running transverse to the height of the blade. A rough surface finish is shown across the channel, particularly towards the thin end of the blade where the powder deposition results into considerable deviation from the original design in the form of voids and internal pores.

The inner structure of DLD-built blade revealed in Figure 4.7 provides improved insight into the dimensional accuracy and surface quality of the objects produced by the DLD process. Figure 4.7 (c) reveals the presence of internal pores (dark areas), particularly in the area close to the blade trailing edge. Furthermore, the rough nature of the internal surfaces produced by DLD can be resolved.

The flexible setup of I12 JEEP allows smooth changing between imaging and diffraction experiments, while staying in the monochromatic mode. The imaging and diffraction detectors are mounted on separate moving translation stages. The beam stop is mounted downstream of the sample separately from the detectors, on a bracket that is translated independently from other experimental components. A large area 2-D Pixium RF4343 detector was used for diffraction pattern acquisition, with a 2880×2881 pixel matrix and a $148 \times 148 \mu\text{m}^2$ pixel size.

The beam was collimated to $0.2(hor) \times 0.4mm(ver)$ with the same energy of 80keV. For the DST acquisition the sample was rotated between 0° and 180° in steps of 3° , i.e. through the total of 61 rotation angle increment. At each rotation angle, the sample was scanned across the beam in steps of 0.2mm equal to the horizontal size of the beam. To account for the irregularity of the blade shape, the scan range across the sample (and hence the total number of scan steps) was adjusted every 15° , i.e. every 5 rotation increments, as shown in Figure 4.8. The acquired image numbers were then assigned to their position within a sinogram along with the integrated diffracted intensity. A total of 5832 diffraction patterns were collected using the exposure time of 1s to ensure sufficient statistic at all (x, φ) positions.

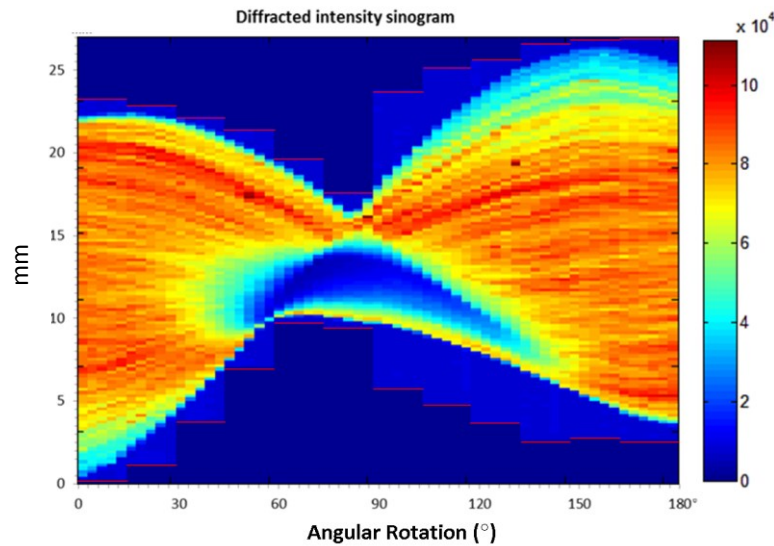


Figure 4.8: Integrated diffracted intensity sinogram. The vertical axis x is given in mm corresponding to the overall specimen width. The horizontal axis is in degrees showing the rotational increments around φ axis. The intense absorption at angular increments where the specimen long axis orientation is close to the incoming beam direction is shown in the graph. The resolution of the sinogram i.e. pixelated elements correspond to the integrated diffracted intensity at each (x, φ) scanning position.

A particular calibration approach was developed and used to determine the accurate sample-detector distance at different rotational increments. A thin capillary (0.1mm $\varnothing$) filled with NIST Si powder was used as a calibrant mounted on the table in the exact position where the holder for the specimen was mounted. Photographs of the sample and calibrant were acquired by a camera mounted in a fixed position directly above the sample table, allowing the calibrant's position to be related to that of the sample. The beam was collimated to 20mm (*hor.*) $\times$ 0.4mm (*ver.*) to make sure that the calibrant always is within the field of view. Calibration

diffraction patterns were acquired at 15° increments from 0° to 180°. The movement of the Debye-Scherrer ring pattern centre (as a result of the sample's sinusoidal movement with respect to the detector), together with the pattern scaling due to the change in calibrant (i.e. sample) to detector position, provided a means of obtaining the information about the circular orbit of the calibrant on the rotation stage.

The complex shape of the blade cross-section posed a number of previously unaddressed problems with respect to the data acquisition, as well as calibration and analysis. The blade cross-sectional shape illustrated in Figures 4.7 (b) and (c) corresponded to the approximate envelope shape causing intense absorption at angles close to 90° rotation increments resulting in the diffraction signal to become extremely weak. In the integrated diffracted intensity sinogram in Figure 4.8 this phenomenon manifests itself by the blue “blade-shaped” region of low intensity. Subsequent analysis was carried on the collection spectra according to the previous section in order to extract the apparent lattice strain.

The curved blade shape means that, as the sample is translated in the x -direction, the distance between the centre of gravity of the diffracted volume and the detector continuously changes according to the shape of the blade (Fig. 4.3). Given the size of the blade, this change causes a significant apparent peak shift on the detector. This shift is of purely geometric origin and disproportionately large compared to the subtle shifts registered by apparent lattice variation. Therefore the geometry error analysis described in the previous section is of particular importance in order to extract the most accurate amount of information regarding the residual elastic strain distribution within the Ni blade.

From the rotation of the calibrant around the axis of rotation, its sinusoidal course around a centre located at position **A** on Figure 4.3 is determined. This point defines the nominal Sample Detector Distance, for which the value of $SDD_{\text{nom}} = 1061.17$ mm was obtained. Moreover, the position of **B** with respect to **A** in terms of radius ($r_{SDD}=0.75\text{mm}$) and phase ($\varphi_o=8.93^\circ$) at each rotation increment was also determined by the calibration procedure. The

circular orbit of point **B** on the sample with respect to the centre of rotation at point **A** was calculated by fitting both the transverse movement of the Debye-Scherrer ring centre (direct beam), and by determining the SDD for the calibrant powder, to match the peak shift on the detector at 15° increments from 0° to 180°.

Two strategies were followed in order to verify calculations and to demonstrate the effect of pseudo-strain distributions, in contrast with the actual strain variation that are the objective of the final reconstruction procedure. The first strategy was to plot the line profiles of the calculated strains for the 0° and 180° slice. Since these two scans correspond to the measurements of the same physical strains in the sample, different only in terms of the sample orientation, the two profiles ought to appear symmetric. The plots display the strain profiles calculated before and after applying the *SDD* correction. The results are shown in Figure 4.9 (a). The dashed blue and red lines give the strain variation obtained when a single *SDD* value is used throughout the calculation for both the 0° and 180° line scan results. Solid blue and red lines are used to show the effect of the sample detector distance (*SDD*) correction strategy described above. Evidence of residual strain variation in the examined slices at 0° and 180° can be readily identified after the applied correction procedure as they are illustrated in Figure 4.9 (a). For direct correlation the 180° scan line is flipped and positioned on the same range as scan line at 0°. The results also provided the opportunity to assess the level of noise in the strain measurement ($\sim 2 \times 10^{-4}$) after removing geometric aberrations; and to conclude that, within the accuracy of the measurement, the first strategy results in a correction procedure that overcomes fully the geometric aberrations caused by the experimental setup.

The second correction strategy adopted was to follow the tip of the blade located at the right hand side in Figure 4.7 (b) through the sequence of angular positions where it is not obscured by the blade bulk (from 0° to 60° and 120° to 180°). Based on the assumption that the measurement always interrogates the same gauge volume at different rotation increments, the strain deduced after the correction should remain constant. Any remaining errors in the sample detector distance correction are bound to have an acute effect on the apparent strain, since the

blade tip undergoes most significant translation along the incident beam direction at different rotations, thus leading to changes in the SDD. The solid red line in Figure 4.9 (b) showed the uncorrected strain variation, whereas the blue solid line shows the strain calculated after the correction was applied. It is apparent from Figure 4.9 that the SDD correction applied was successful in eliminating geometry-induced aberrations.

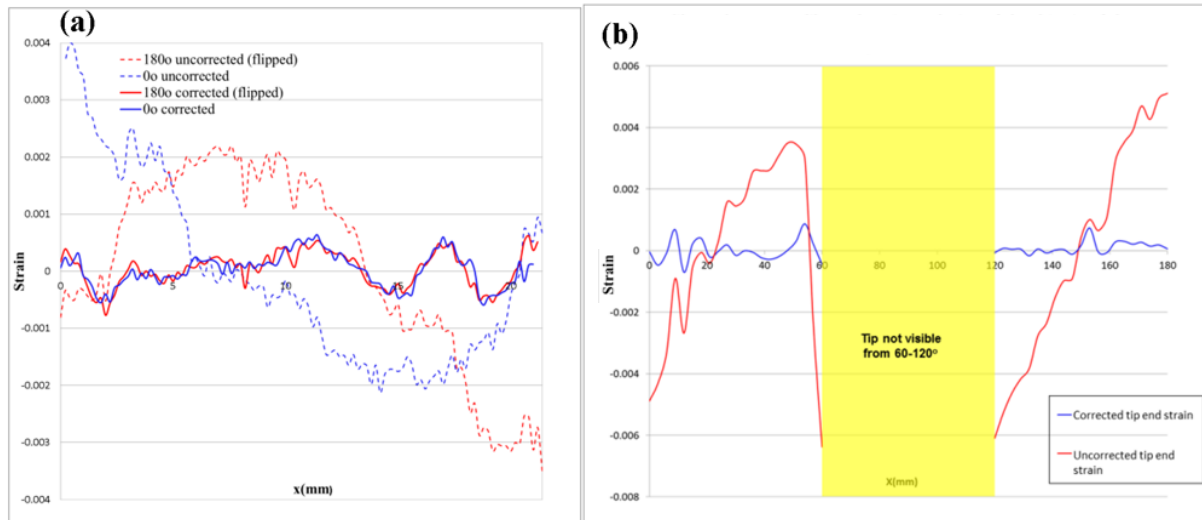


Figure 4.9: (a) Plots of nominal strain variation across the blade at 0° and 180°. Dashed lines indicate uncorrected calculations, whilst solid lines indicate strain profiles computed after applying the correction to the sample detector distance. (b) Plot of generated pseudo strain distribution from 0° to 60° and 120° to 180° in red line versus equivalent corrected sample detector distance calibrated strain in blue line.

4.3.3. Contour method

The specific steps of the contour method implementation are discussed in Chapter 2. Electric Discharge Machining (EDM) was used to cut the specimen through the same section where the diffraction experiment was carried out. The final sectioning of the sample and measurements were done at 10mm distance from the top free surface of the blade. The bottom part of the sample (i.e. blade base, DLD steel substrate) 20 mm below the top surface was initially removed. The remaining blade was moulded into tin based alloy to allow for a rectangular outer shape of the sample to be cut in the final section. The 20 mm high moulded specimen was cut using EDM into two halves to create two parts with identical geometry for further measurements. After this process the sample tin mould was removed.

The 2-D cross-section on both sides of the specimen was measured using the Mitutoyo Coordinate Measurement Machine (CMM) based in HEXLab, Department of Engineering

Science, University of Oxford. A 1mm diameter probe end was used at the CMM for the measurements and 2-D mapping technique a 2 μ m increment was used to characterise the local specimen topography, registering the height variation. The information of the CMM measurements was further processed with MATLAB[®] eliminating “bad” points outside the periphery of the specimen and at the region of cooling channel holes. Both mating surfaces were then averaged in order to cancel out shear stress effect on the measured displacements. The noise is typically removed from the data by fitting the averaged displacements to a smooth function; a cubic spline fit has been used for this purpose.

A 3-D FE model for stress calculation was built for one half of the specimen using ABAQUS 6.10. The sample geometry was meshed with linear hexahedral elements with reduced integration (C3D8R), and 236K elements were used for this analysis. It was assumed that each specimen was isotropic and linear elastic, and the moduli and Poisson’s ratios are 220GPa, 0.33 respectively. The mesh application on the FE model is shown in Figure 4.11 (inset). Displacements obtained from the cubic spline interpolation of the averaged data were applied to the model as displacement boundary conditions, and linear elastic stress analysis was performed to calculate the stress field normal to the plane of sectioning.

The 2-D cross-sectional map of the normal residual stress component was obtained, and compared with the residual elastic strain obtained from the DST reconstruction.

4.3.4. Results and conclusions

The results of the two methods are shown in Figure 4.10 for the FE based contour method implementation and in Figure 4.11 for the DST method. The nature of the direct laser sintering process is known to introduce significant residual stresses variations in the bulk of the specimen. The layer-after-layer material deposition at high temperatures and the consequent temperature drop results in freezing-in strain mismatches due to uneven contraction during solidification.

In the case of the contour method the 2-D cross-sectional out-of-plane (s_{33}) is shown in Figure 4.10. The results show a consistent thin (~ 0.5 mm) tensile region at the boundary of the specimen periphery. The s_{33} magnitude reaches a maximum of 400 MPa and an inner balancing layer of a compressive stress is noticeable. A maximum 500 MPa compressive stress is located towards the trailing end of the blade. It appears that this surface tensile to bulk compressive transition is more evident in the thinner part of the blade, particularly in the positions where there are no cooling channels. Contrary to that, close to the sample core, there are noticeable tensile stresses present between the cooling network channels.

The calculated path length diagram (“thickogram”) constructed on the basis of conventional tomographic imaging is multiplied by the apparent lattice variation diagram (“strainogram”) according to Eq. (4.3), before using a filtered back-projection in MATLAB[®]. The result is then converted to strain variation (out-of-plane strain component ε_{33}) using the mean value of the calculated lattice parameters in each (x, φ) increment. The estimated lattice parameter for the Ni 263 alloy was calculated at 3.5669Å, which in good agreement with the results reported in literature.

Obviously, the 3-D nature of the object and elastic strains within it means that the stress calculation must include the contributions from all strain components. However, it is also clear that the axial stress values are predominantly sensitive to the axial strains. Therefore, the “nominal” stress distribution was also calculated for the case of DST Ni-blade reconstruction by multiplying the axial strain values by the Young’s modulus of the material (220GPa). The reconstructed map of the axial stress component s_{33} is shown in Figure 4.11, after applying thresholding and masking. Tensile stress regions can be readily observed in the near surface regions of the specimen which can also be observed in the contour data results for s_{33} at approximately the same locations. This value is similar to the contour result for the same region. Furthermore, some features of the internal stress distribution within the cross-section are captured, e.g. the location and magnitude of compressive strain towards the trailing edge of the blade, as shown in Figure 4.10. Aberrations related to the back-projection reconstruction

technique used and the limited number of rotation increments (61) resulted in some line artefacts seen in the reconstruction in Figure 4.11. It is worth noting that the small concentrations of tensile stresses inside the blade section were not captured by the DST reconstruction. However, no evidence of such tensile regions could be obtained from the consideration of the “strainogram” slices at 0° and at 180° , shown in Figure 4.9 (a).

The application of DST to the analysis of residual stresses in a DLD blade manufactured from Ni-superalloy C263 presented a number of previously unaddressed challenges. The significant values of “pseudo-strains” that arise due to sample’s large aspect ratio and rotation required very precise characterisation and accounting for the blade movement around the axis. A separate calibration procedure was introduced to provide input for the calculation of the geometrical data correction. This was done on the basis of the transmitted intensity sinogram collected during tomographic imaging acquisition. The large specimen size in its long dimension (21.4mm) meant that the beam was almost totally absorbed, so that now useful diffraction patterns could be recorded at several positions x for angular increments (φ) from 80° to 100° .

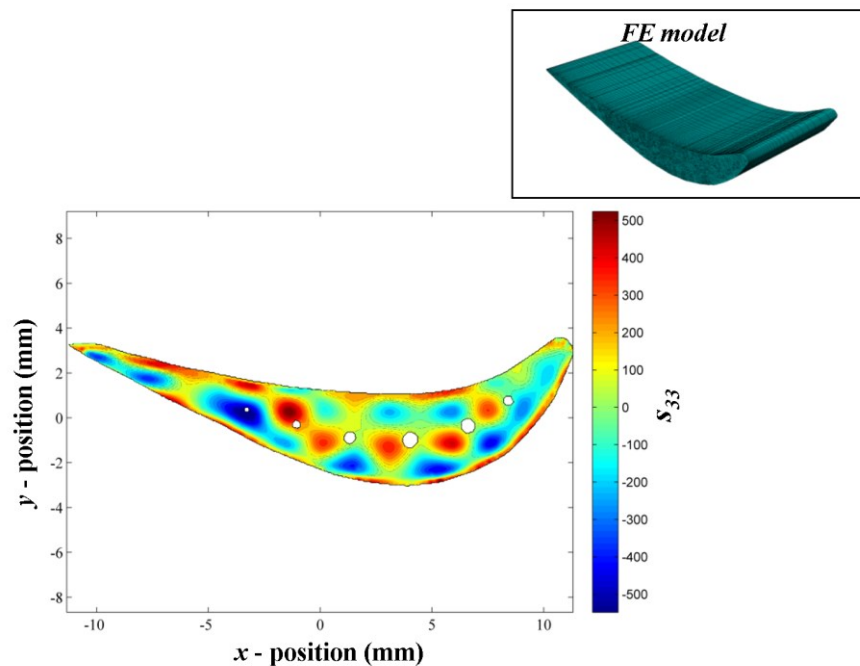


Figure 4.10: Result of s_{33} out-of-plane stress component using the contour method. An overall tensile stress profile is dominating the outline of the specimen. The tensile stress in the near surface regions, is balanced in the bulk by regions of compressive stress. Further interactions are observed in the intermediate regions between the cooling fins network. The inset shows the meshed FE model as prescribed by the CMM measurements.

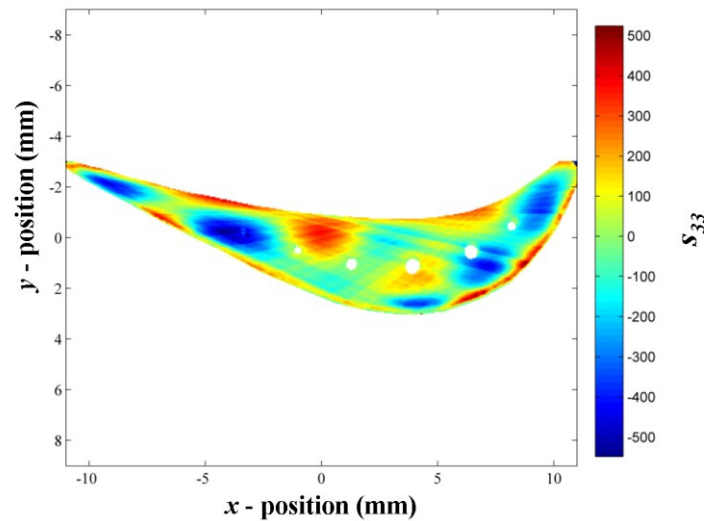


Figure 4.11: Result of nominal s_{33} out-of-plane stress component using the DST method. This method also captures the boundary layer of tensile stress counteracted by a compressive region in the bulk. The registered stress variation in the intermediate cooling channel region is not matching the equivalent stress shape variation registered by the contour method.

4.4. Multiphase DST on human dental tissue

4.4.1. Materials and applications

The human tooth is a hierarchical mineralised tissue composed of collagen-rich dentine and highly mineralised enamel which formulate a two-level composite structure. Both tissues are calcified biological composite structures incorporating biological mineral hydroxyapatite (HAp) nano-scale crystals and organic protein. The hierarchically structured dentine and enamel are different in terms of the volume fraction of mineral (50% in dentine and 80-90% in enamel by volume) [160, 161], crystal size and shape of HAp crystals, as well as crystal perfection [162-164]. The interface region between them is commonly referred to as the dentine-enamel-junction (DEJ) and demonstrates remarkable resistance to existing failure mechanisms within the oral cavity. This stands in stark contrast to the interfaces between artificial dental restorative materials and dentine [165]. DEJ constitutes a superb lesson from nature on how to achieve strong, durable bonding between very dissimilar materials: hard, brittle enamel and softer, but tougher dentine. Visualizing and understanding the internal architecture of these materials is a challenging task that cannot be readily accomplished by conventional microscopy methods alone.

One suitable technique for the study of nano- and sub-nano-scale structure is SAXS and WAXS, which are advanced non-destructive techniques widely used to reveal quantitative multi-scale information about the microstructural and mechanical properties of mineralized tissues [166-169]. WAXS technique is able to reveal quantitative information about the crystal lattice parameters, while SAXS enables the characterisation of the orientation and degree of alignment in crystalline and amorphous materials. Understanding the complex mechanical behaviour related to this hierarchical structure is essential for predicting the effects of microstructural alterations on the performance of dental tissues and their artificial replacements. Both SAXS and WAXS have been applied only recently to the study of mineralized biological composites, such as bovine and human teeth [170-175]. None of these studies were performed in 3-D, of which the critical insight into the microstructure and crystallographic characteristics inside the bulk specimen is still lacking.

In a combined mode of operation, X-ray diffraction can be used in conjunction with tomographic techniques to achieve micron spatial resolution within the bulk and non-destructively map internal microstructure and lattice parameter distribution in the sample. Diffraction Strain Tomography (DST/WAXS) is sensitive to the phase transition between low mineralised dentine and highly mineralised enamel, thus it can be selectively used to map the distribution of apparent lattice variation in each tissue individually. In addition to the wide angle scattering data, small angle scattering can provide additional information regarding crystallite size and orientation. Tomographic SAXS acquisition has been proposed and applied to axially symmetric samples by Schroer and Kuhlmann [176]. However, the internal architecture upon the two phases of biological mineralised tissue has not yet been sufficiently described.

In this section a multi-scale analysis of human dental tissue, combining two non-destructive synchrotron-based experimental techniques, wide and small X-ray scattering (WAXS/SAXS) was attempted. Quantitative spatial distribution maps were obtained for various parameters in the human dental tissues such as mean HAp crystallite thickness by the interpretation of SAXS patterns and lattice parameter distribution across the DEJ by DST-WAXS analysis.

A freshly extracted normal human third molar (ethical approval obtained from the National Research Ethics Committee; NHS-REC reference 09.H0405.33/ Consortium R&D No. 1465) was washed and cleaned in distilled water to eliminate residues, so that the possibility of contamination or other chemical effect was excluded.

Two millimetre thick disk (including dentine, enamel and DEJ) was cut from the same tooth using a low speed diamond saw (Isomet Buehler Ltd., Lake Bluff, Illinois, USA) were further prepared into smaller bars. Diamond cutting and subsequent grinding down to 10 μ m lapping film was used to refine the final 2 $\times$ 2 $\times$ 2mm cube. The sample was kept for a maximum of 7 days in distilled water in a commercial fridge at 4°C until the experiment was performed.

4.4.2. Multi-phase DST implementation and tomographic imaging

Synchrotron based SAXS/WAXS tomography setup

Simultaneous SAXS/WAXS tomography of human dental tissue cubes was carried out on I22 beamline at Diamond Light Source (DLS, UK) with monochromatic beam at the photon energy of 18keV. The beam was collimated to the spot size of 65 $\times$ 120 μ m² on the sample. To record the SAXS/WAXS tomogram, the sample was scanned through the beam in the translate-rotate mode, and a single layer scan of SAXS/WAXS patterns near the top of sample surface were collected. Each line scan across x on the specimen was done in 43 increments at a 65 μ m step (defined by the size of the beam) allowing for the entire surface of the sample transverse to the beam to be covered at all rotations. After each line scan was completed, the sample was rotated by 3° and the line scan repeated (total of 61 rotation increments from 0° to 180°). The diffraction measurements were conducted on the top surface of the sample to facilitate further investigation using FIB-DIC technique.

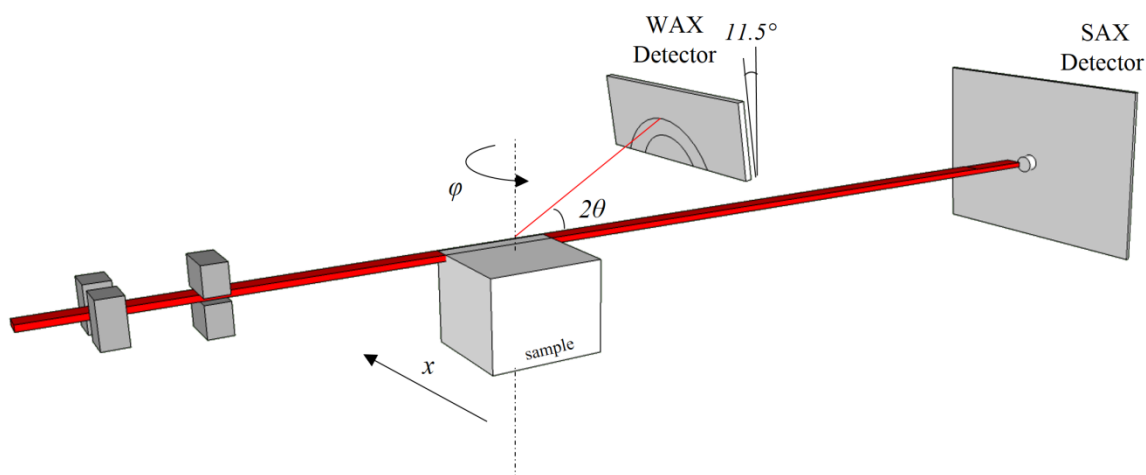


Figure 4.12: schematic diagram of experimental simultaneous SAXS-WAX tomographic acquisition set-up in I22, DLS.

In order to record both the WAXS and SAXS patterns at the scanning location, the WAXS detector (Pilatus 100K, Dectris, Baden, Switzerland) was mounted at the angle of 11.5° to the incident beam, so as to leave the SAXS detector exposed (Fig. 4.12). A diode was mounted on the beam stop of the SAXS detector (Pilatus 2M, Dectris, Baden, Switzerland) in order to collect the incident and transmitted beam intensity. The setup is shown schematically on Figure 4.12 including the sample representation (exaggerated), scanning points, collimation, and detector positioning. A lightly compacted disk of NIST standard silicon powder was used for the WAXS data calibration, and a disk of Silver Behenate powder was used for the SAXS data calibration.

Lab based computerised tomography (CT)

A commercial X-ray micro-computed tomography (micro-CT) system was used to obtain a 3-D reconstruction of the sample structure after the SAXS/WAXS tomography experiment. The scan was carried out at $7.47\ \mu\text{m}$ resolution using 40 kV voltage, 120 μA current and a 0.5mm Aluminium filter (SkyScan 1172, Bruker microCT, Kontich, Belgium). The resulting 480 projections were reconstructed using the SkyScan NRecon[®] software package using a fan beam approximation, and subsequent 3-D models were created using Fiji imaging software [177].

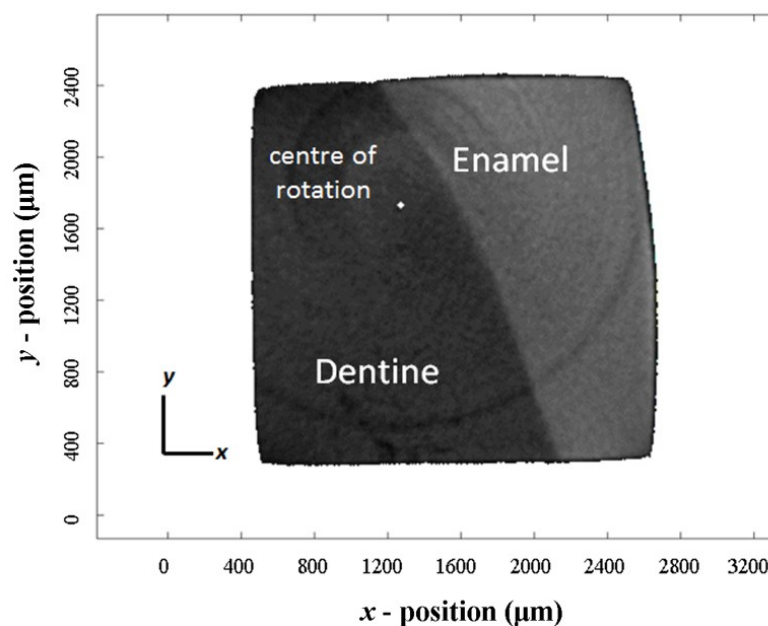


Figure 4.13: Tomographic reconstruction of the cross-section examined for the purpose of the diffraction data acquisition. Dentine and enamel regions are highlighted together with the associated centre of rotation used to determine the geometrical corrections strategy and inform the peak-fitting algorithm.

For the purposes of tomographic acquisition the specimen was kept mounted on the same sample holder as in the synchrotron experiment in order to preserve the alignment. The aim was to keep the top surface of the sample perpendicular to the axis of rotation by employing the same sample alignment procedure for both tasks. Using the phase sensitive WAX integrated intensity and the transmitted intensity recorded by the diode the sample initial orientation was deduced at $\varphi=0^\circ$. The reconstruction is shown in Figure 4.13 was used as a guide to segmenting phases and calibrating sample-detector distance at each scan position (x, φ) .

WAXS data analysis

2-D WAXS patterns were processed into 1-D intensity plots, as detailed in the previous section. Subsequently, the 1-D diffracted intensity versus 2θ profiles of the [002] peak in dentine determining the c lattice parameter of the HCP lattice (principal axis) and [210], [211] and [112] peaks in enamel were fitted determining a and c lattice parameters of the HCP hexagonal lattice. Figure 4.14 illustrates a corresponding multi-peak fitting on a diffraction pattern obtained from a pure enamel region on the specimen at $x=28$ lateral position and $\varphi=0^\circ$ using Gaussian functions and background subtraction. The algorithm was implemented as a MATLAB[®]

function using the least squares algorithm to obtain the best fit, similar to a Pawley refinement method.

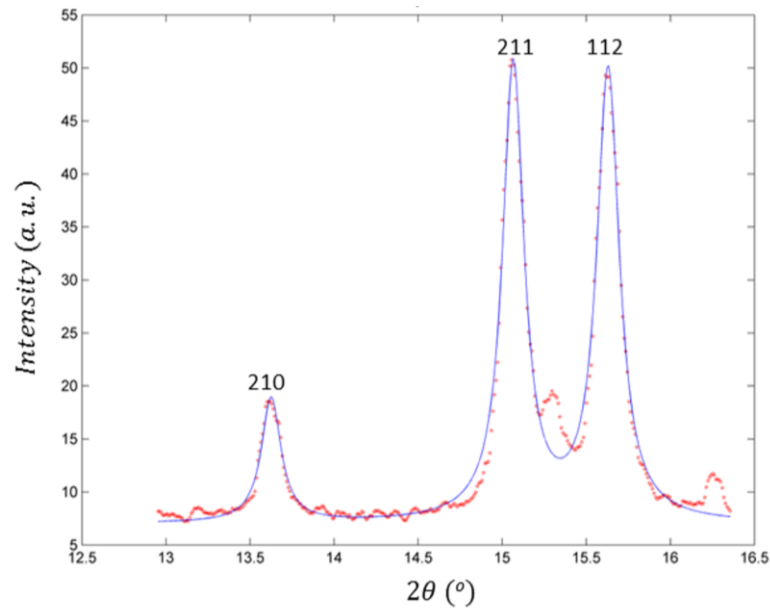


Figure 4.14: Representative multi-peak fitting on enamel using MATLAB[®] implementation. The fitted peaks are [210], [211] and [112]. The software was able to adjust the fitting parameters at each in order to discriminate between dentine and enamel diffraction patterns informed by the tomographic reconstruction of the tooth.

The same procedure was used for fitting dentine [002] diffraction peak. The decrease in crystalline content in dentine causes severe peak broadening which has not been used for lattice spacing determination. Figure 4.15 shows a diffraction pattern acquired from a pure dentine region on the specimen at $x=5$ lateral position and $\varphi=0^\circ$, and provides an illustration of the peak fitting accuracy. Preferred data orientation analysis was not performed on the WAXS due to the diffraction detector setup which allowed 60° - 100° azimuthal angle of the Debye-Scherrer ring spectrum to be registered. The same calibration procedure as for the DLD blade was used to determine the axis of rotation (illustrated in Figure 4.3).

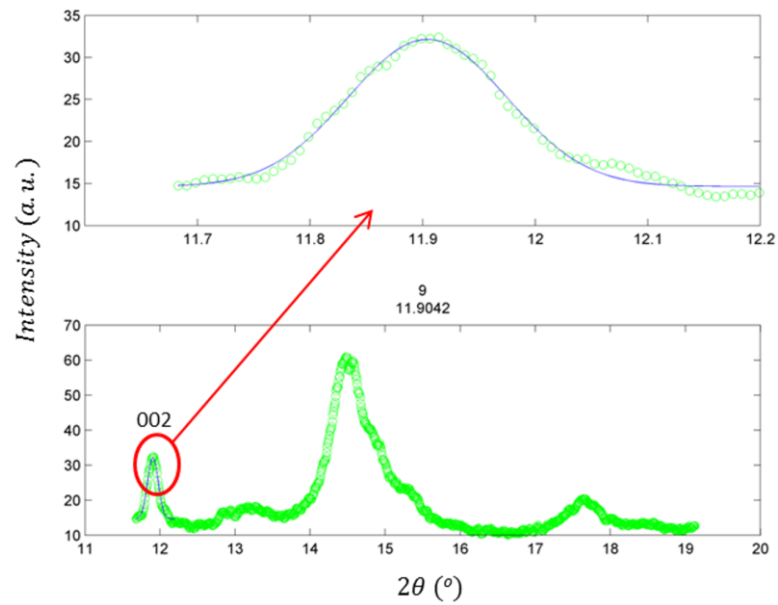


Figure 4.15: Representative multi-peak fitting on dentine using MATLAB® implementation. The peak fitted for dentine was [002].

The multi-phase algorithmic implementation allowed for dentine and enamel diffraction signal to be fitted individually even in positions where the patterns were overlapping. This was accomplished with the help of the high-resolution tomography imaging data at each (x, φ) position. This input allowed the 2θ fitting range to be prescribed in the interpretation procedure. At scan points where the amorphous content of dentine contributed a strong background signal, the [211], [112] enamel crystalline peaks displayed reduced intensity above the background. The fitting routine then used a 50% increased weight for the [210] peak to improve the precision of fitting. The average strain error related to peak centre uncertainty was found to be ~ 350 microstrain for single peak fitting for dentine and ~ 210 for multiple peak fitting for the enamel.

SAXS data analysis

Quantitative interpretation of SAXS patterns provides insight into the mean thickness (smallest dimension) and degree of alignment of HAp crystallites. In order to determine the mean crystal thickness, the 2-D SAXS patterns were converted into 1-D intensity profiles and processed using Fit2D software package. The 1-D SAX profile pattern was created by integrating scattering intensity $i(q, \theta)$ radially at each (x, φ) position over the entire range of the azimuthal

angle θ . A function of $I(q)$ was then obtained where q represents the scattering vector [178]. The crystal mean thickness T can be defined as the Porod chord length based on Porod's law valid in a two-phase system:

$$T = \frac{4}{\pi} \frac{Q}{P} \quad (4.11)$$

where P is the Porod constant given by $I(q)=Pq^{-4}$ (Porod's law at large q range), which can be determined from the $Iq^4 \sim q^4$ plot. Q is the integrated area under the $Iq^2 \sim q$ plot. Note that the definition of T was used without any assumption about the particle shape. However, in the case of needle or platelet shape crystallites, T can be interpreted as the average measurement of the smallest dimension of crystallites. The actual mean crystalline thickness can be further calculated based on T and other factors like volume fraction [169]. Quantitative information can be also extracted from the SAXS pattern regarding the degree of alignment (ρ) of HAp crystalline particles [179, 180]. In the present investigation and for the purposes of tomographic reconstruction the framework for reconstructing vector quantities (such as orientation) has not yet being implemented.

4.4.3. FIB-DIC

Following X-ray diffraction and imaging data collection the sample was mounted on an SEM sample holder using conductive silver paint as adhesive, with the orientation preserved. Part of the top surface of the sample was also covered with silver paint. The surface was further subjected to Au sputter coating in vacuum at 10mA for 90s. This technique minimises charging effects and the 10nm Au layer improves SEM imaging contrast as shown in Figure 4.16 (inset). Micro-ring-core FIB-DIC technique [55] was performed using a Tescan LYRA3 SEM-FIB. FIB-DIC procedure was carried out for 18 annular trenches and “islands” of 4 μ m diameter across the central part of the top surface of the tooth, 9 annuli in the dentine and 9 in the neighbouring enamel region (Fig. 4.16). The distance between adjacent annuli was set to 100 μ m. The “island” diameter of 4 μ m was selected as a balance between strain spatial

resolution and providing sufficient surface area to produce a reliable estimate of strain relief. A trench width of $0.5\mu\text{m}$ and a milling rate ($\text{nm}/\text{sec}\cdot\text{mm}^2$) of 0.2 was used for enamel and 0.4 for dentine to reduce the impact of material re-deposition. In order to improve surface contrast and topology, a beam voltage of 15kV was used to image the sample and over 340 high resolution micrographs were captured.

Analytical explanation of the FIB-DIC data implementation is given in Chapter 2. The output result of the FIB milling analysis is the in-plane residual elastic strain components (ϵ_{11} , ϵ_{22}) on a line across the sample surface. The advantage of the FIB-DIC approach is that it provides a measurement of residual elastic strain that is not dependent on the knowledge of strain-free lattice parameter, and can be applied with equal to (poly)crystalline, amorphous, and complex structured, hierarchical composite materials. In the present case the method helps to determine the nature and source of lattice parameter variation at the surface of the sample.

In the case of the examined specimen the minor residual strain relief on the specimen would suggest that the recovered lattice parameter variation by the diffraction experiment is not associated to mechanical deformation. Other reasons related to the material structure, such as HAp content, chemical agents embedded in the tooth lattice during its lifetime, natural growth of the dental tissue, can also have an effect on the measured lattice parameter variation.

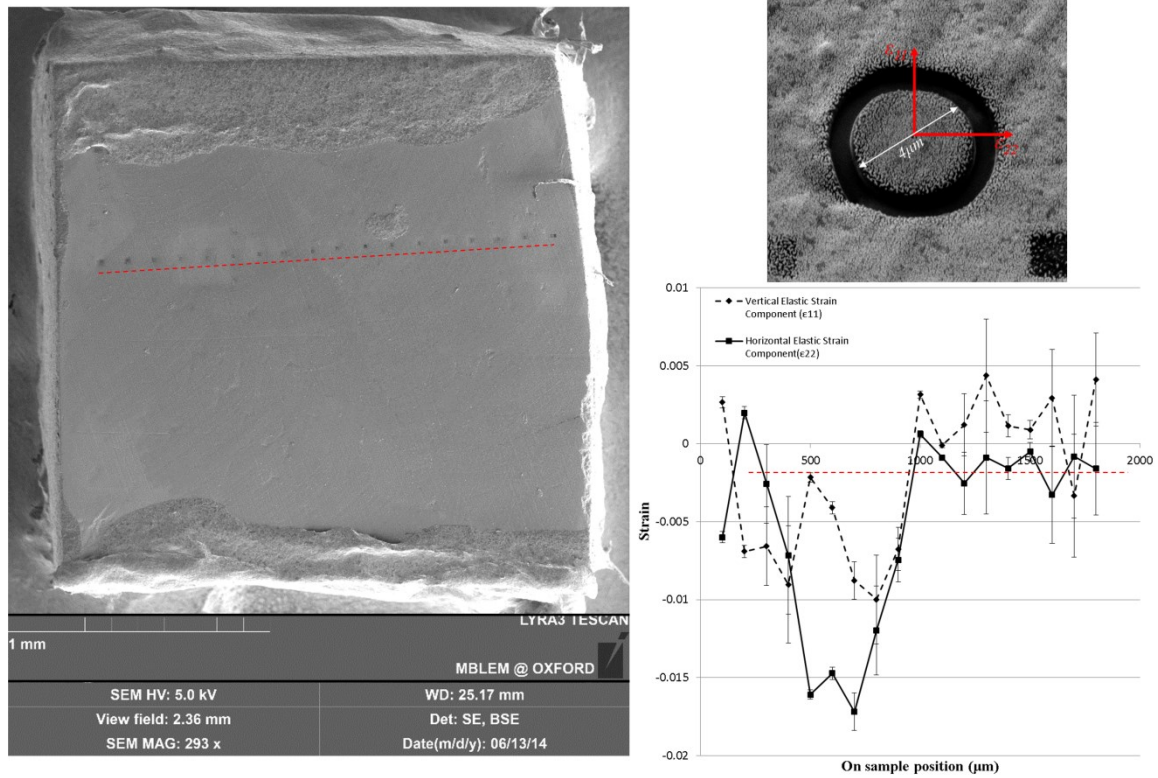


Figure 4.16: Tomographic reconstruction of the cross-section examined for the purpose of the diffraction data acquisition. Dentine and enamel regions are highlighted together with the associated centre of rotation used to determine the geometrical corrections strategy and inform the peak-fitting algorithm.

4.4.4. Results and conclusions

The spatial residual strain variations of HAp crystals in the dentine and enamel are visualized in Figure 4.17 (a), where the colour code is used to denote the positive (red), negative (blue) and intermediate (green, yellow) values on the 2-D contour plot. Separate colourbars are used for the dentine and enamel regions respectively due to the significant difference of strain magnitude observed. Although 2-D mapping provides an improved understanding of the distribution, the detailed information of the changes across the DEJ can be more readily deduced by considering a line profile. The strain variation along the red dashed line in Figure 4.17 (a) is shown in Figure 4.17 (b).

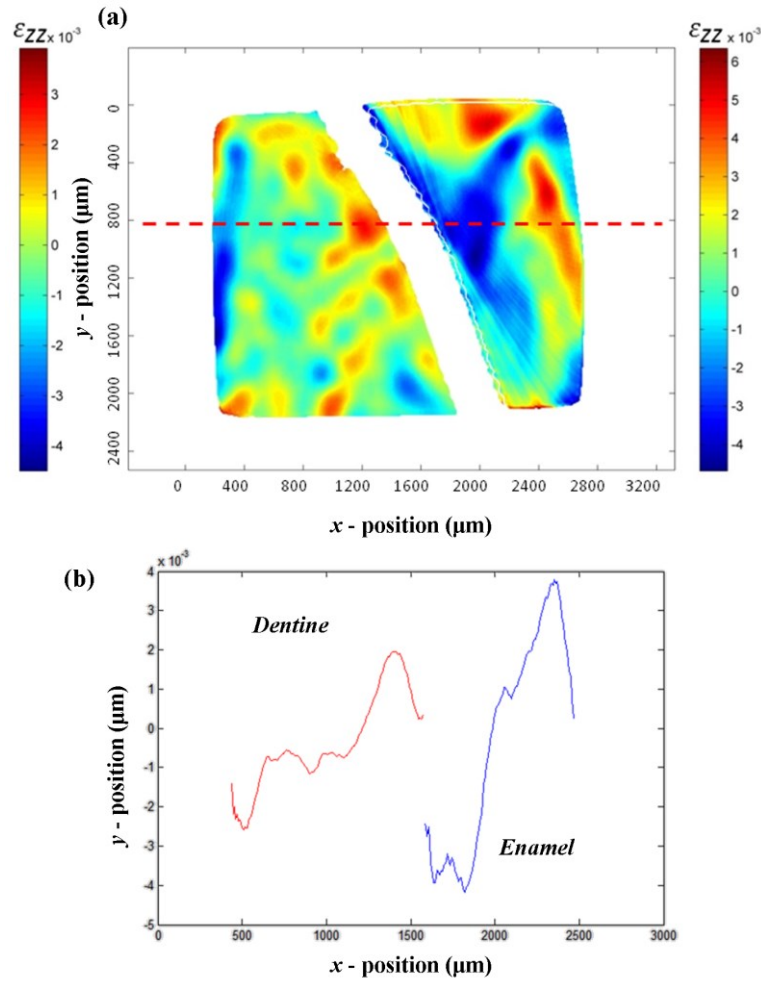


Figure 4.17: (a) DST reconstruction of ϵ_{33} strain variation in dentine and enamel regions. The strain variation across the dashed line is drawn in (b) where dentine is highlighted in red and enamel in blue.

The strain variation of ϵ_{33} component in dentine seems to have significant fluctuation towards tensile strain close to the DEJ interface. The same gradient in approximately the same position (i.e. close to the interface with enamel) is shown for the ϵ_{11} and ϵ_{22} component variation in the FIB-DIC results. However, the FIB-DIC variation shows strong compressive stains from points 4 to 8 which suggests the average calculated lattice parameter (c) used for strain calculation in the DST is underestimated. Since both techniques were applied on the surface where complete out-of-plane stress relief ($s_{33} = 0$) can be assumed the reconstructed ϵ_{33} is directly influenced by the in-plane stress variation (Poisson effect: $\epsilon_{33} = \frac{1}{E}(s_{33} - \nu(s_{11} + s_{22}))$). Therefore the average 1.2% in-plane strain in dentine ($\nu = 0.44$) recorded by FIB-DIC can be associated to the 0.5% out of plain reconstructed strain variation. Furthermore, compression is shown towards the free-end boundary of dentine on DST reconstruction. The ϵ_{22} result on the

same region (i.e. point 1) shows compression but the spatial resolution of the FIB-DIC acquisition did not allow measurements at the very end of the specimen.

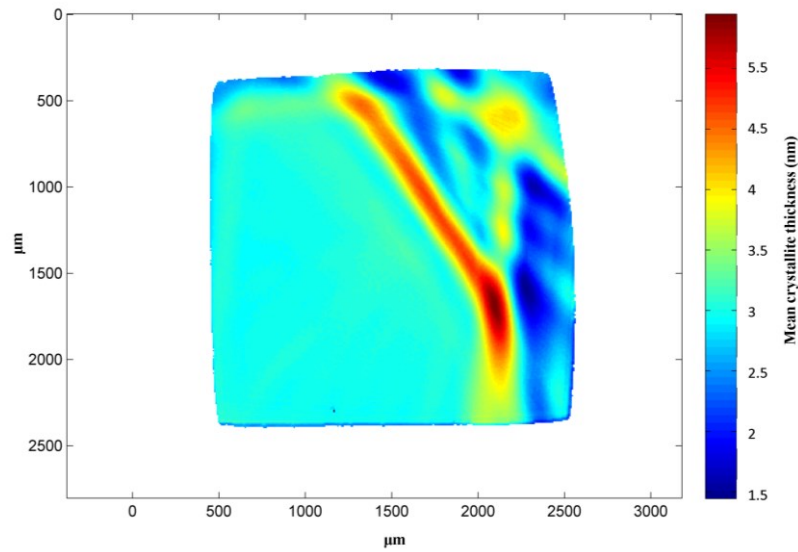


Figure 4.18: SAXS Tomographic reconstruction of the examined cross-section.

A comparison of the DST results on enamel against FIB-DIC technique identified significant differences. The in-plane strain components showed no significant variation whereas in the out-of-plane strain reconstruction significant fluctuation was observed. A steep compressive strain was reconstructed in the bulk at a parallel to the DEJ section and sharp tensile strain regions were recorded closer to the outer regions. At this stage it was assumed that the steep average lattice parameter variation recorded by WAXS-DST is not associated to mechanical origins.

This hypothesis, that the apparent lattice variation is linked to structural variation of the crystallite structure in enamel seems to be confirmed by the mean crystallite thickness reconstruction illustrated in Figure 4.18. The SAXS tomographic reconstruction revealed a uniform region of high mean crystallite thickness ($\sim 5.5\text{nm}$) in the region close parallel to the DEJ interface in enamel. The SAXS tomography results in terms of magnitude can be correlated to 2-D SAXS mapping on the DEJ interface in a comparative dental slice sample studied by Tan et al. [178]. However, the resolution of the mapping procedure was at $200\mu\text{m}$ which did not demonstrate the steep near-DEJ mean crystallite thickness variation in enamel.

Overall, the results of the combined multi-scale SAXS-WAXS approach with the contribution of FIB-DIC semi-destructive method illustrate the complex nature of dental tissue structure and mechanical characteristics. In order to overcome aberrations related to the X-ray methods high resolution X-ray SAXS-WAXS 2-D mapping over an equivalent region will be able to validate the reconstructed cross-section data. Furthermore, a FE approach incorporating mechanical state information together with multi-scale structure-related information will be able to uniformly correlate the data collected via different experimental routes, thus giving more detailed information. Earlier studies in the group such as the one by Tan et al. [181] have shown promising results in the potential of application of an improved multi-scale Eshelby inclusion model that takes into account the two-level hierarchical structure.

4.5. *In situ* loading DST of yttria-stabilised zirconia fixed prosthetic denture

4.5.1. Materials and applications

A range of materials are used in modern dentistry for the fabrication of load-bearing framework structures, with the most important classes being noble metal alloys and engineering ceramics.

Extensive clinical experience exists relating to the use of metallic frameworks veneered with porcelain. However, in the long term the cosmetic appearance of prostheses based on metal frameworks is often unsatisfactory, with the underlying dark metallic structure becoming progressively more visible through the translucent porcelain veneer. Recent decades have witnessed the increased use of all ceramic frameworks in restorative dentistry [182].

Sintered polycrystalline zirconia provided an excellent option, as it has natural white colour, and possesses an excellent combination of mechanical properties (stiffness, strength, fracture toughness, resistance to fatigue and corrosion) with biocompatibility and good processing capability. Powder route fabrication of complex shape sintered zirconia components is well advanced, and involves CNC machining of green compacts followed by sintering (with account taken of shrinkage); alternatively, the two operations can be reversed. Dimensional

accuracy of about $10\mu\text{m}$ can be readily attained even for components possessing a complex shape.

The toughening of zirconia ceramics is achieved by alloying with yttrium, with the resulting material often referred to as yttria-stabilized zirconia (YSZ). The toughening effect is caused by the stress-induced phase transformation from the tetragonal to the monoclinic phase which is accompanied by volume expansion. As a consequence, the compressive internal stresses that are generated reduce the effective stress intensity experienced by crack tips, and promote crack closure, thus increasing the apparent fracture toughness and improving resistance to fatigue crack growth [183].

Nevertheless, the use of all-ceramic Fixed Prosthetic Dentures (FPDs) still represents a challenge, since the nature of deformation and strength of sintered ceramics is fundamentally different from that of ductile metallic alloys. To improve the level of confidence in the mechanical performance of zirconia-based veneered frameworks, better insight is required into the residual and "live" stress distributions in these systems. Residual stresses develop during fabrication of FPDs across a range of length scales. At the macroscopic scale, the residual stresses arise during sintering and cooling due to the thermal gradients and the temperature dependence of material properties. Furthermore, the application of porcelain veneer and its subsequent heat treatment causes interfacial residual stresses (due to the mismatch in the thermal expansion coefficient) that play an important role in controlling the integrity of both the porcelain overcoat and the underlying ceramic framework. At the microscopic (granular) level, residual stresses arise between neighbouring grains due to the anisotropy of crystal stiffness and thermal expansion.

"Live" stresses arise in FPDs due to mastication loads, and due to temperature changes within a patient's mouth. The principal criterion used in design is to ensure sufficient resistance to the repeated application of contact loading, taking into account the potential corrosive effect of the aqueous environment present. Cyclic contact loading causes bending stresses that reach

particularly high magnitude within connectors between units, where fatigue failure typically occurs.

A considerable amount of research has been devoted to the study of the strength and durability of all-ceramic FPDs, through both computation and stress-based statistical strength prediction [184, 185], as well as laboratory experimentation [186]. Clinical evidence of the 2-year performance of all-ceramic veneered FPDs has also been reported [187].

In the present study the focus was given on the important intermediate stage of analysis that occupies the place between computer modelling and strength prediction, on the one hand, and full-scale laboratory or clinical testing, on the other. This intermediate stage related to non-destructive experimental determination of strain (and stress) distributions within FPDs. Non-destructive evaluation of internal stresses allows direct quantitative validation of deformation models.

An analytical technique that allows non-destructive evaluation of internal stresses is X-ray diffraction as discussed previously [142]. The use of different X-ray diffraction modalities has been reported, e.g. monochromatic X-ray beams in combination with angular scanning or 1-D/2-D position-sensitive detectors (e.g. Ref. [143]), or white (polychromatic) X-ray beams with fixed energy-resolving solid-state detectors [145]. Recently, powerful achromatic X-ray optics systems have been developed that allowed the use of very small (sub-micron) polychromatic X-ray beams to perform Laue diffraction experiments from (within) single grains [153]. The use of micro-beam Laue diffraction to evaluate residual stresses underneath the veneer on ceramic FPDs has been reported [154]. The residual stress evaluation obtained in that study was based on the analysis of the statistical distribution of individual grain stresses that had an average value of about -60MPa, but with large standard deviation and ranging from -1500 to 1500MPa, thus limiting the usefulness of the result. Furthermore, the very high X-ray absorption coefficient of ZrO_2 means that the penetration depth of X-rays remained very low in those experiments, so that the measurements were confined to the zirconia–porcelain interface.

In this section the distribution of internal strain within an FPD under the application of simulated mastication load was reconstructed using Diffraction Strain Tomography (DST). The aim is to present the tomographic reconstruction of 2-D internal strain distribution within an *in situ* loaded complex-shaped object: a polycrystal zirconia ceramic FPD framework. The results of the measurement and strain map reconstruction were compared with FE modelling results for the same cross-section. The details of the data acquisition and implementation are presented and discussed.

4.5.2. FPD shape analysis and modelling

The all-ceramic dental bridge framework under investigation was made from anatomically reduced bridge copings using Wieland ZENO[®] Zr sintered zirconia ceramic material (Wieland Dental + Technik, Pforzheim, Germany). Wieland ZENO[®] Tec process was used to manufacture the framework that was left uncoated (no porcelain veneer applied) for the purposes of this study.

To assist in carrying out stress analysis of bridge deformation under simulated mastication loading, a solid CAD model was constructed. The input used for model assembly was derived from the results of conventional absorption contrast X-ray tomographic reconstruction. The input dataset for this reconstruction was collected by laboratory X-ray radiography using Yxlon[®] industrial X-ray system (160kV, 2.25kW) and a Hamamatsu[®] X-ray detector at HEXLab, Department of Engineering Science, University of Oxford, UK.

The tomography stage set-up used in the Oxford HEXLab is illustrated in Figure 4.19 (view along the incident beam). The black motorized rotation stage for locating the sample at different projection angles/with respect to the incident beam is placed in the centre, in front of the mounting frame for the X-ray detector. Careful adjustment is required in terms of stage tilts in the vertical and horizontal planes. This is accomplished through the use of linear translation stages seen at the front and on the left of the assembly. The translation stages are coupled to the plate underneath the main rotation stage through small angle wedges, so that relatively large

displacements on the translators result in relatively small changes in the stage tilts. This allows tilt accuracies better than 0.1° to be achieved. Finally, translations of the entire assembly are also possible in the horizontal plane, along the beam direction and perpendicular to it. Figure 4.19 (b) shows a closer view of the FPD mounted in front of the X-ray detector (Hamamatsu®).

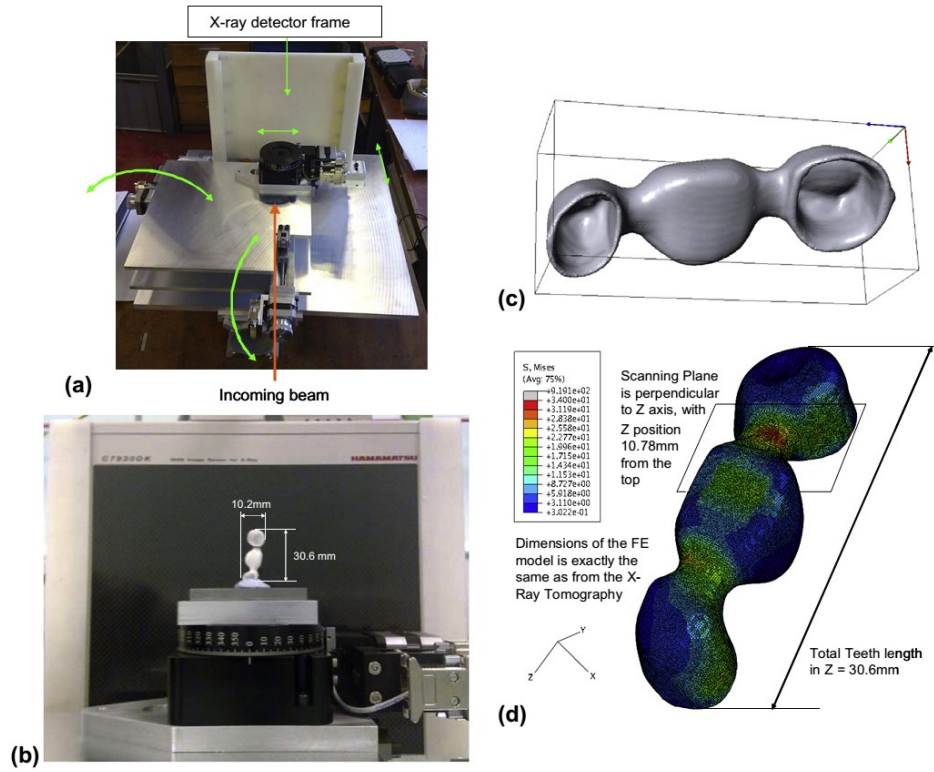


Figure 4.19: (a) Schematic illustration of the HEXlab tomographic set-up. (b) Illustration of the FDP mounted in front of the Hamamatsu X-ray detector. (c) Solid model of the three-unit dental prosthesis obtained by laboratory X-ray tomography. (d) A FE model of the three-unit dental prosthesis constructed from the solid model and showing the contours of von Mises equivalent stress under a nominal applied mastication load of 300 N [22].

Three hundred and sixty projections were acquired by rotating the sample in steps of 0.5° . The 3-D volume representation was reconstructed using a filtered back-projection algorithm (*iradon*) implemented in MATLAB®. The resulting reconstruction was rendered by introducing a density threshold, and a solid model was constructed, as illustrated in Figure 4.19 (c).

A 3-D FE mesh was generated using commercial Simpleware® software using a quadratic element type (C3D20R) directly on the X-ray CT data. A linear elastic model was implemented to simulate the behaviour of the specimen under mastication load. The FPD was treated as isotropic and homogeneous, with Young's modulus and Poisson's ratio of 250 GPa and 0.23,

respectively, i.e. within the typical range for Yttria stabilised ZrO_2 . The resulting mesh is illustrated in Figure 4.19 (d), together with the contour plot of the equivalent von Mises stress computed under the application of 300 N nominal mastication load applied on the central unit, with the support provided by fixation of the two outer units on a simulated jaw substrate.

4.5.3. Diffraction measurements and interpretation

A special three-point bending rig was designed to allow controlled application of *in situ* loading similar to that during mastication. The loading rig is illustrated in Figure 4.20 (a). For synchrotron X-ray diffraction experiments (beamline ID15B, ESRF, Grenoble, France) the dental bridge set (bridge framework and polymethyl methacrylate (PMMA) support simulating the jaw) was loaded *in situ* using the rig mounted on the beamline sample stage (Fig. 4.20 (b)).

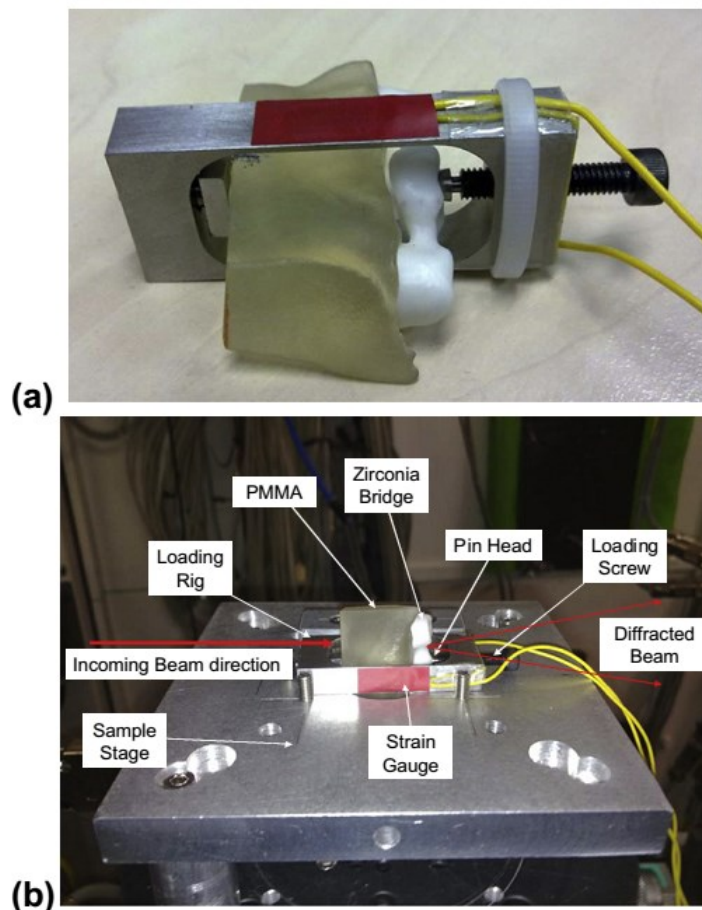


Figure 4.20: (a) *In situ* three-point bend loading device used to apply the simulated mastication force on the synchrotron beamline ID15B (ESRF). (b) Illustration of the *in situ* loading and scanning arrangement on beamline ID15B at the ESRF (dental bridge three-point bending rig mounted on the rotation and translation stage). The x and y -axes are in the plane of the table and correspond to the directions across and along the incoming beam, respectively. The z -axis is vertical (normal to the table surface) [22].

The bending load was applied via the lead screw incorporated in the rig. The screw was tipped with a Ti–6Al–4V pinhead that was reamed and push-fitted into the screw shank. The smooth circular shape of the tip ensured that the load was applied only in terms of transverse displacement causing framework bending, but the transmission of torque due to screw rotation was excluded. The strain gauges on the rig were previously calibrated so that their readouts could be readily interpreted in terms of the applied contact load on the middle unit. The loading rig was mounted on a sample stage which had the capability of applying x , y , z translation and ϕ rotation around the vertical axis. The examined cross section where bending load was applied is shown schematically in Figure 4.21 (a).

A monochromatic 2-D diffraction set-up was used to collect diffraction patterns. This is illustrated schematically in Figure 4.21. The incident beam was monochromated to a photon energy of $86.93 \pm 0.01 \text{ keV}$, and collimated to the spot size of $0.1 \times 0.1 \text{ mm}^2$. The beam traversed the entire thickness of the sample, and scattered from the zirconia bridge framework to form Debye-Scherrer rings (Fig. 4.21 (b)). 2-D Pixium detector with the pixel matrix of $1920(\text{H}) \times 2640(\text{V})$ and pixel size of $154 \times 154 \mu\text{m}^2$ [188] was used for diffraction data acquisition. Texture effects are ignored during the lattice parameter fitting procedure. Calibration was carried out using a CeO_2 stress-free powder sample to determine the precise values of the radiation energy and the sample–detector distance. The sample–detector distance (more specifically, the distance between the stage centre of rotation and the detector) was found to be $D = 731.93 \pm 0.04 \text{ mm}$, i.e. the uncertainty of sample–detector distance determination was $\sim 5 \times 10^{-5}$. The sample was rotated between 0° and 180° in steps of 3.6° , i.e. through a total of 51 rotation angles. At each rotation angle, the sample was scanned across the beam in steps equal to the beam spot on the sample (0.1 mm). Translation scans across the sample required a maximum of 57 steps.

It is worth noting here that, as expected, the presence of the PMMA support did not affect the diffraction data collected from the zirconia framework. This is due to the fact that at the beam energy of $\sim 87 \text{ keV}$ used in the experiments, the absorption (and scattering) power of zirconia exceeds that of PMMA by several orders of magnitude.

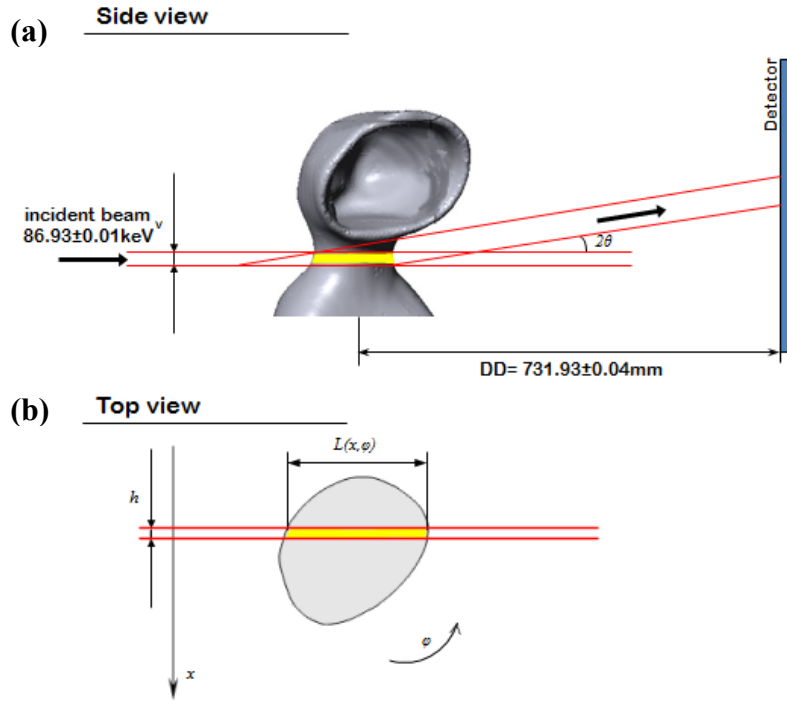


Figure 4.21: (a) Illustration of the scattering geometry used in the diffraction experiment (side view). (b) A schematic representation of the diffraction experiment (top view).

4.5.4. Results and conclusions

Figure 4.22 illustrates the comparison between the FE calculation of the bending strain distribution within the chosen cross-section through the FPD (Fig. 4.22 (a)) and the tomographic strain reconstruction obtained from diffraction data using the above approach (Fig. 4.22 (b)). To enable the validation of the experimental results, an elastic bending FE model for the prosthesis was used that was deliberately kept as simple as possible. The emphasis here is on capturing the effect of the bridge geometry on the local stress distribution and magnitude. The experimental technique captured the general nature of the bending strain distribution correctly, although some deviations from the expected distribution must be noted.

The tensile strain region within the cross-section of the connector was clearly captured in both the strain reconstruction based on experimental data, and in the FE simulation. However, the compressive region was not captured as well in the strain tomography contour plot. In fact, there even appears to be a region of tension on that side of the cross-section, which is counter-intuitive. Furthermore, the magnitude of the strain reconstructed from experimental measurements did not show a perfect match with the simulation results.

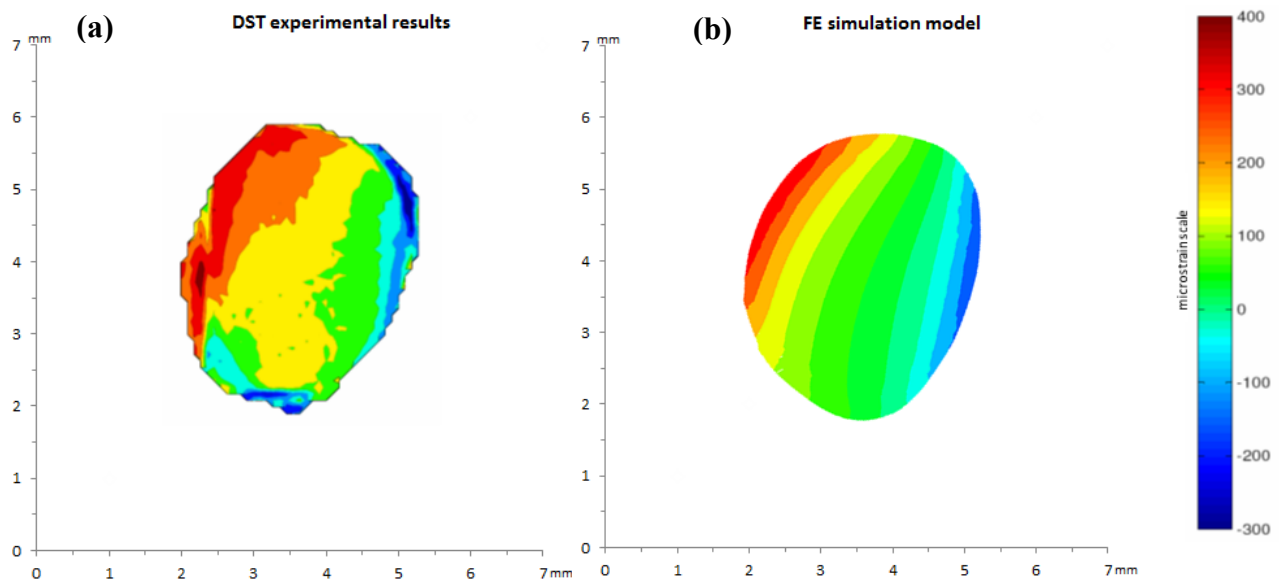


Figure 4.22: (a) The simulated bending (and tension) strain contour plot from the chosen section of the dental bridge under a 300 N nominal applied load. (b) The result of strain tomography reconstruction of the bending strain distribution within the same cross-section under a 300 N nominal applied load [22].

Several reasons for these disagreements were identified. Firstly, the precise nature of the loading conditions was difficult to ascertain. While the nominal load applied in the experiment was 300N, in practice the strain gauge signal showed some fluctuation, with the greatest deviation from the nominal value being $\pm 100\text{N}$. This may not only cause a scaling of the strain distribution, but might also affect values in different regions of the cross-section in different ways at different times. Improvements in the stability and reliability of the loading arrangements are required for future development of this work.

Furthermore, the exact nature of the support provided by the PMMA block for FPD was not known with sufficient precision. Perfect bonding was assumed in the FE simulation, meaning that three-point bending may also have induced some overall tensioning within the connector. It is difficult to ascertain whether this phenomenon was captured correctly; however, some evidence of overall tensioning could also be seen in the strain reconstruction.

The reconstruction of strain was based on a relatively small number (51) of sample rotation angles (“projections”). Filtered back-projection reconstructions based on a small number of projections are often prone to aberrations and artefacts. Increasing the number of “projections”

at the time of strain tomography data collection is an obvious further improvement recommended for future studies.

Finally, some difficulties were associated with the necessity to correlate the orientation and shape of a complex shaped sample between two sets of experimental data: absorption tomography for shape determination, and diffraction tomography for strain reconstruction. In order to overcome these difficulties, imaging and diffraction measurements should be performed as part of a single experimental session, without altering the sample position between them. In conclusion, the results obtained in the present study demonstrated the viability and potential value of strain tomography for the analysis of internal stresses in all-ceramic zirconia frameworks. It was possible to assert with confidence that DST research technique is identified for experimental stress analysis in complex, high value components used in dentistry. The method allowed (i) experimental investigation of the effects of FPD support arrangements on stress distributions, and hence strength; (ii) validation of numerical models; and (iii) experimental analysis of the effects of heat treatment, application of porcelain veneer, fatigue, corrosion, etc.

4.6. X-ray Laue Orientation tomography (LOT)

In polycrystalline materials, the orientation, shape, intragranular structure and size of individual grains, or clusters of grains, can have important implications for a range of aspects of the material behaviour. The motivation is to be able to accurately map and characterise the mechanical behaviour of a material in the non-destructive context of X-ray diffraction and imaging techniques at sub grain length scales.

The process illustrated in the present section, is based on the ability to capture and eliminate Laue patterns for an individual grain from a collected pattern of a multi-grained material cross-section. The technique is termed X-ray Laue orientation tomography, which best summarises the diffraction and tomography methods used and the output produced. A multiple-angle set of projections using transmission Laue diffraction setup and translation rotation technique is

collected. An algorithm uses the indexed Laue pattern images to distinguish the total number of grains within the bulk and separately reconstructs diffracted intensities formulating the problem at the same fashion as convention absorption contrast tomography. Improvements of the algorithm proposed by Hoffman et al. [36] are presented. Furthermore an inaugural Rodriguez vector relative mis-orientation strategy theory is deployed in order to reconstruct internal mis-orientation within each grain. The potential and limitations of this new approach are discussed, and results are illustrated and evaluated.

4.6.1. Introduction - Principles

A considerable research effort has been given, over the past fifteen years into the development of 3-D grain mapping techniques for polycrystalline materials. Early studies aimed at orientation and strain characterisation of individual grains based on 3-D reciprocal space mapping approach [189, 190].

The evolution of 3rd generation synchrotron X-ray sources offering high brilliance on the high energy X-ray spectrum allowed for real space description of bulk polycrystalline materials in terms of 3-D shapes and orientations of all grains present in the illuminated sample volume. A range of techniques have been proposed and are analytically discussed in the methods section (Chapter 2) using dedicated setups offering the ability of 3-D grain structure reconstruction relying on harvesting grain specific information.

The first set of techniques are based on full-field illumination, monochromatized beam and multi-angle data acquisition to retrieve grain shape in the bulk [42]. The amount of projections is limited to the diffraction occurrences of each grain in the bulk around 360° of rotation. Therefore the reconstructed grain shapes are usually subjected to grain dilation in order to fill the space in between grain clusters.

The second category of techniques such as "differential aperture X-ray microscopy" (DAXM) [38], obtain 3-D information by scanning the sample in three directions. The setup employs a micro-beam Laue transmission setup and involves wire scanning procedures across

each scan point in order to determine spatial distribution of grain orientations in the bulk. Practical limitations of the setup arise from a very complex wire scanning setup, which allows only for specific rear flat surface samples to be examined. Furthermore, the scanning arrangements are time expensive due to the sheer magnitude of scanning positions at each increment.

The Laue Orientation Tomography (LOT) technique proposed here relies on an alternative principle and leads to a different experimental scheme. The new approach is able to simultaneously reconstruct, truly non-destructively, the 2-D distribution of relative mis-orientation within a cross-section of a grain.

Laue Orientation Tomography (LOT) uses the translate-rotate experimental procedure to obtain the mis-orientation angle distribution sinogram. A framework that exploits all the information from the experiment is described hereinafter establishing the principles of the proposed technique.

Assuming a constant incident flux, an approximating assumption can be made that the recorded total intensity of the Laue pattern in the transmission diffraction configuration is governed by the sample path length within the scattering crystal. Absorption means that the longer the distance the diffracted beam is travelling in the sample, the lesser the intensity reaches the detector. On the other hand, the longer the incoming beam "travels" within the specific grain (crystal), the great number of crystalline planes fulfil the diffraction condition, causing the intensity of the Laue spot to increase. In the case of hard X-ray synchrotron beams ($>50\text{keV}$) the relative magnitude of the absorption phenomenon is much lower than the scattering path length effect, so that the former can be ignored.

One can assume that the integrated intensity of the Laue diffraction pattern provides a measure of the length of the beam path through a particular grain at a specific measurement position (x, φ) . If this assumption is true, the problem can be treated in a framework that is similar to that used for the absorption tomography discipline. The key difference for this "rich"

tomography application is that tomographic data analysis now should possess have grain orientation sensitivity, so that the morphology of each grain can be determined by collecting redundant, multiple-angle projections of the same object.

The data collection of LOT follows the “pencil” beam translate-rotate acquisition principle illustrated in the beginning of the chapter and micro-beam Laue transmission setup described by Hoffman et al. [24]. The differences arise by the nature of diffraction effects with respect to powder monochromatic diffraction of DST. The Laue patterns consist of a series of distinct Laue “spots” due to the multiple diffraction incidents between the multi symmetric lattice crystals and the white (polychromatic) incident collimated beam. No collimation of the scattered beams is performed and the Laue patterns are registered for all (x, φ) positions are registered on a 2-D area detector.

4.6.2. Laue pattern prediction from rotation matrices ($\mathbf{R}$)

Once the Laue patterns are collected from the experiment and indexed, a framework able to generate Laue spots on a simulated detector plane for the determined lattice orientations is used in order to harvest the Laue intensity for individual grains across the sample at each rotation increment (φ). The corollary is that by the accurate determination of the experiment axis of rotation the individual grain orientations at each φ can be associated, therefore the intensity of separate grains can be harvested into grain specific intensity "sinograms" and followingly reconstructed used algebraic reconstruction techniques (ART).

This section gives the outline of the mathematical tools used to predict the Laue spot positions on the detector on the basis of known rotation matrix ($\mathbf{R}$) that represents the crystal orientation with respect to the laboratory reference frame. The rotation matrix ($\mathbf{R}$) represents the rotation that needs to be applied to a grain in the reference configuration in order to rotate it to the real orientation state within the sample. The framework takes into account the geometry of the experimental setup in order to determine the multiple order reflection positions on the detector.

The relationship between an infinite 3-D crystal lattice, defined by its primitive vectors $\mathbf{a}_1$, $\mathbf{a}_2$, $\mathbf{a}_3$ in the crystal reference frame with the equivalent reciprocal space vectors has been defined in Eq. (1.10). The reciprocal space vector $\mathbf{d}_{hkl}^*$ has been expressed as a linear combination of the primitive h, k, l vectors in Eq. (1.9). The $\mathbf{d}_{hkl}^*$ vector prescribes the normal to the scattering plane. By multiplying the rotation matrix $\mathbf{R}$ by the $\mathbf{d}_{hkl}^*$ vector, that represents the lattice normal orientation in crystal coordinates (primitive vectors), the normal to the scattering plane ($\hat{\mathbf{n}}$) in the lab coordinates can be expressed as (shown in Figure 4.24):

$$\hat{\mathbf{n}} = \frac{\mathbf{R} \cdot \mathbf{d}_{hkl}^*}{|\mathbf{R} \cdot \mathbf{d}_{hkl}^*|} \quad (4.12)$$

The scattered beam unit vector $\hat{\mathbf{s}}$ can easily be calculated using:

$$\hat{\mathbf{s}} = \hat{\mathbf{s}}_0 + 2(\hat{\mathbf{s}}_0 \cdot \hat{\mathbf{n}}) \cdot \hat{\mathbf{n}} \quad (4.13)$$

where $\hat{\mathbf{s}}_0$ denotes the incident beam direction. The shortest distance vector $\mathbf{d}_c$ from the scattering point to the detector surface is:

$$\mathbf{d}_c = \mathbf{SDD} \cdot (-\mathbf{d}_s) \quad (4.14)$$

where:

$\mathbf{SDD}$: is the sample to detector centre distance in vector notation $\mathbf{SDD} = (0, 0, D)$

$\mathbf{d}_s$: is the normal to the detector plane ($\mathbf{d}_s = \mathbf{d}_1 \times \mathbf{d}_2$, where $\mathbf{d}_1$ and $\mathbf{d}_2$ are the horizontal and vertical axis on the detector as shown in Figure 4.23).

The Laue spot position ($\mathbf{P}_L$) in vector notation can now be determined on the detector surface relative to the detector centre position as follows:

$$\mathbf{P}_L = \frac{|\mathbf{d}_c|}{\hat{\mathbf{s}} \cdot \hat{\mathbf{d}}_c} \left[\hat{\mathbf{s}} - \hat{\mathbf{d}}_c (\hat{\mathbf{s}} \cdot \hat{\mathbf{d}}_c) \right] - \mathbf{SDD} + \mathbf{d}_c \quad (4.15)$$

Here $\hat{\mathbf{d}}_c$ is the unit vector of $\mathbf{d}_c$. The projection of vector $\mathbf{P}_L$ onto the detector axes $\mathbf{d}_1$ and $\mathbf{d}_2$ gives the position of the reflection on the detector in detector coordinates. This vector then

needs to be normalised by dividing its length by the pixel size of the detector and added the detector's centre pixel positions (x_{cen}, y_{cen}). Note, that the detector centre pixel position is not determined by the pixel width, but is determined from the experimental calibration procedure described below.

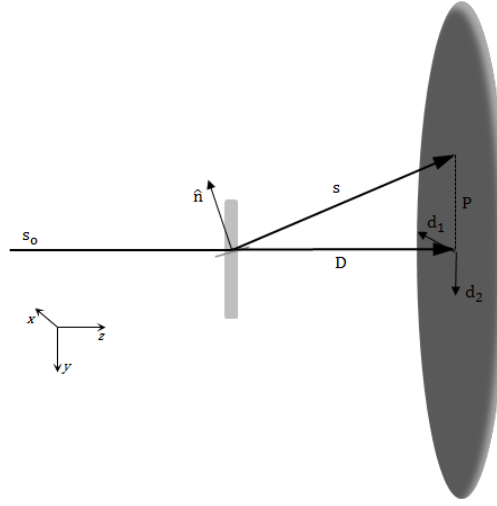


Figure 4.23: Schematic illustration of Laue spot generation on the plane of the 2-D detector defined by d_1 and d_2 . Scattering plane is defined by the scattering vector $\hat{n}$.

4.6.3. Laue diffraction data collection and implementation

The first implementation of the method was performed at the I12 - JEEP: Joint Engineering, Environmental and Processing (DLS) beamline [36]. The high-brilliance, high-energy of the beamline, together with a versatile multiple acquisition environment allows for the transmission Laue setup to be used. The reason primarily lies into the beamline available hard energy spectrum ($>50\text{keV}$), resulting into small scattering angles ($<20^\circ$) through the beam interaction with the lattice, which allows for most of the diffracted intensity to be harvested by a flat panel detector downstream the sample perpendicular to the beam direction.

The sample was mounted on I12 sample stage which offers high resolution x, y, z translation and φ rotation around the vertical axis. Another two linear translation stages were mounted aiding the alignment of the sample to the centre of the rotation. The *Laue diffraction* setup used is illustrated in Figure 4.1. The incident beam was collimated to the size of $20 \times 20 \mu\text{m}^2$ and lower energy spectrum was filtered out using 12mm copper filter limiting multiple scattering

phenomena and enhancing the quality of the acquired Laue patterns. The incident beam (by default z direction) travelled through the sample generating Laue patterns according to the orientations of the different grains illuminated. The Laue spots were recorded by the 2- D Pixium detector mounted perpendicularly to the beam. The Trixel Pixium RF 4343 detector is of particular interest for micro-beam Laue diffraction. It has a large active area of $426.24 \times 426.388 \text{ mm}^2$ which is subdivided into 2880×2881 pixels. A beam stop was used to absorb the straight through transmitted beam which can saturate or damage the detector.

Calibration was carried out in two stages to define the precise sample detector distance. Si wafer was placed in the approximate sample position, and calibration patterns were acquired. In the *second* stage, the Ni sample itself was used to generate calibration patterns, and these were acquired at three different sample positions: one with the sample upstream to the beam (z direction), one with the sample downstream to the beam and one at the nominal sample-detector distance position ($D \approx 573 \text{ mm}$). This allowed the precise determination of the sample-detector distance by triangulating the patterns acquired using XMAS [153]. XMAS also allows for the (x_{cen}, y_{cen}) position to be determined by overlapping identical Laue patterns collecting at different $SDDs$. The sample was aligned vertically on y to the desired height distance (1.92mm) from its top edge. Lastly, using the finer translation stages, the sample was optically adjusted on the transverse direction x to be at the centre of rotation of the main stage. This allows using minimum number of translation scanning steps for each rotation step.

The acquisition time i.e. exposure time of the detector, was adjusted by monitoring the intensity of the collected Laue patterns. The angular span φ from 0° to 180° was covered in 2° *rotation steps* ($\varphi_i = 1 \dots 90$) chosen as the best compromise between reconstruction function results and time duration of the experiment. On the transverse direction x at each rotation step an area of 1.2mm was scanned at 0.2mm *translation steps* determining the resolution of our reconstruction and governed by the nominal beam size ($x_j = 1 \dots 56$). Hence, 5096 Laue patterns

were collected and indexed using XMAS software. The algorithm implementation was written using MATLAB®.

For the first application of the proposed technique, a highly annealed, polycrystalline nickel sample was considered. A 500 μm wide, 15mm long strip was cut from a 300 μm thick nickel foil. After the sample was polished to 1 μm diamond paste, it was enclosed in evacuated quartz vials to prevent oxidation and heat treated at 1200°C for 4 hours, followed by a furnace cool for 24 hours. These conditions ensure complete recrystallization through thickness of the sample and adequate crystal growth. Figure 4.24 shows optical micrographs of the four sides of the sample to illustrate the shape and observe the annealed crystal structure. The chosen cross-section where the experiment was carried out is indicated.

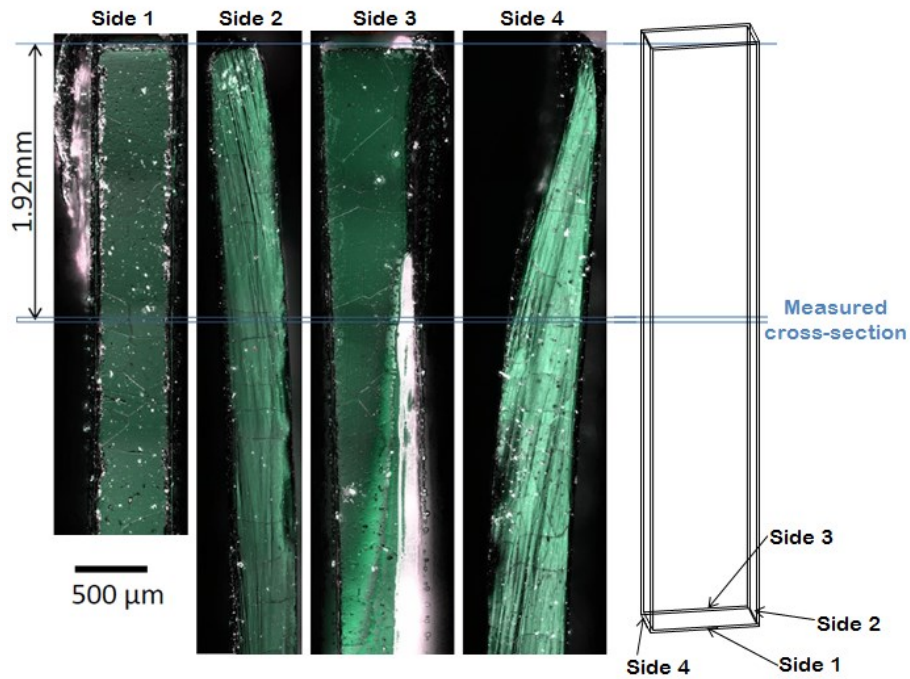


Figure 4.24: Optical micrographs recorded from four sides of the Ni sample for the LOT demonstration. The measurement plane was allocated 1.92mm from the top surface [36].

The first step following data collection is to recover information about the total number of grains within the chosen sample cross-section, and to calculate their average orientations. For that reason the first slice of indexed data i.e. for $\varphi=0^\circ$ and $x_j=1\dots56$ was used. By comparing the output of the fitting routine between adjacent patterns, represented by 3×3 rotation matrices (R) five distinct crystal orientations were identified. For the purpose of the comparison a

threshold value $\theta_{th}=2^\circ$, which represents the angular change for different lattice orientations at each x_j was set. The algorithm operates by retrieving each indexation file, for each successfully indexed position x_j , and calculating the Rodriguez angle $\theta_{Rod,j}$ for the pattern identifying a grain. The $\theta_{Rod,j}$ for j position is compared to the previous $\theta_{Rod,j-1}$ for $j-1$ position according to the following criterion. If $|\theta_{Rod,j} - \theta_{Rod,j-1}| \geq \theta_{th}$ then a new grain orientation is registered. Otherwise if $|\theta_{Rod,j} - \theta_{Rod,j-1}| < \theta_{th}$ it proceeds to the next pattern at $j + 1$, until $j = 56$.

The procedure was repeated for different rotation angles namely $\varphi=90^\circ$ and $\varphi=180^\circ$ to confirm the results for $\varphi=0^\circ$. This routine recorded the five distinct grain orientations within the sample in the form of rotation matrices R_{gr} , $gr = 1 \dots 5$. Each rotation matrix represents the crystal direction-orientation to the laboratory reference frame at rotation $\varphi = 0^\circ$.

In the next step the routine scans through all translation-rotation increments in order to “harvest” the Laue reflections from each grain separately. For that reason, masks are generated (Fig. 4.25) through a simple geometric transformation around the axis of rotation (y) using the known rotation increments, predicting where the individual spots are anticipated on the detector for individual grains according to their indexed orientation. In order for that step to be effective the initial error of rotation matrix determination is required to be very low and the lab coordinate system should be well-defined.

The input information for this procedure is produced by an algorithm that feeds back to the program all the possible diffracting planes of an fcc lattice as it was the case for the Ni sample under examination (in hkl primitive vector notation of crystal coordinates) for a chosen reflection order according to the energy flux of the beam. The energy E_{hkl} of a particular hkl reflection is given by:

$$E_{hkl} = \frac{hc|H_{hkl}|}{2 \sin \theta} \quad (4.17)$$

where h refers to Plank's constant, c the speed of light and $\theta = \frac{1}{2} \cos^{-1}(\hat{s} \cdot \hat{s}_0)$. The spots are further filtered with respect to the spanning surface of the detector i.e. neglecting the ones that fall out of the scanning surface of the detector.

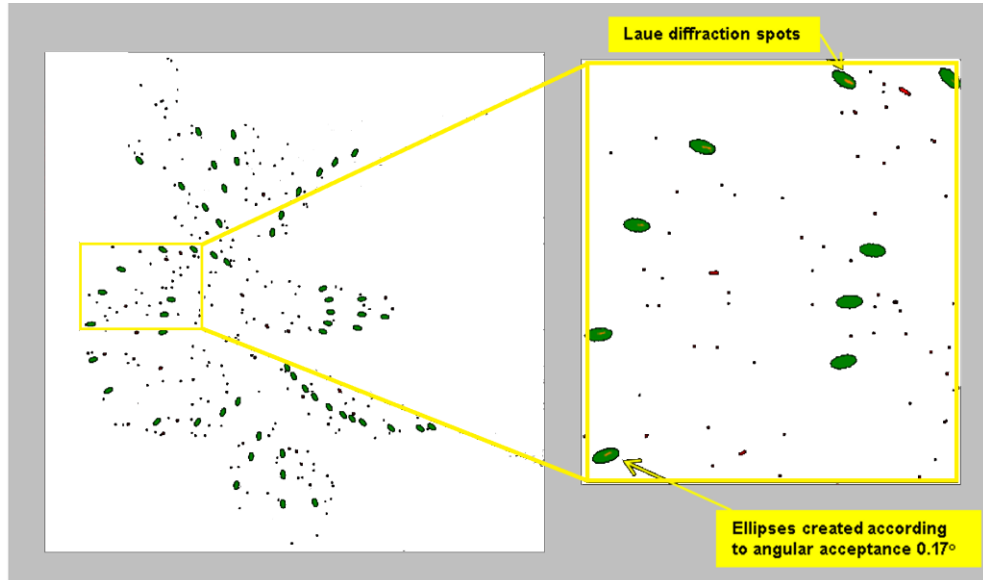


Figure 4.25: Generated mask illustrated in “green” and Laue spots in “red”. On the right a close up indicating the Laue spots that fall within the mask. Individual masks were generated for each of the five identified grain orientations at each ϕ rotation increment. Each mask is multiplying all the translation increments on x for the specific rotation increment.

For the mask construction, ellipses with the size dictated by the angular acceptance of $\pm 0.34^\circ$ in x direction and $\pm 0.17^\circ$ in the θ direction were used, as illustrated on Figure 4.26. Only reflections between 2θ diffraction angles of 10° to 15° were considered according to the scanning surface range of the detector and the sample detector distance. It has to be noted that most real life engineering materials with inherent grain distortions have elongated "streaky" Laue diffraction spots, therefore an ellipsoidal spot shape best captures the intensity compared to circular masks. Furthermore, the maximum $\pm 0.34^\circ$ degrees of acceptance was determined by taking into account how far from the anticipated position the actual Laue spot was at rotation increment and the typical pixel span. Therefore the value is relatively small if the 573mm of sample detector distance is considered.

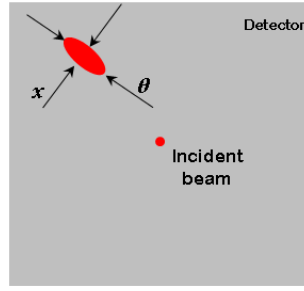


Figure 4.26: Individual ellipsoidal shape corresponding to an individual hkl reflection with highlighted x and θ directions.

The other issue that had to be addressed concerned the forward prediction of the mask formation for the consecutive rotation steps φ_i . Working in the lab reference coordinates, an additional rotation matrix R_{φ_i} was generated at each step by multiplying it by R_{gr} giving the forward predicted lattice orientation around y -axis at 2° rotation steps:

$$R_{gr,\varphi_i} = R_{\varphi_i} \cdot R_{gr} \quad (4.18)$$

where: $R_{\varphi_i} = \begin{bmatrix} \cos \varphi_i & 0 & \sin \varphi_i \\ 0 & 1 & 0 \\ -\sin \varphi_i & 0 & \cos \varphi_i \end{bmatrix}$ and $\varphi_i = 0^\circ, 2^\circ, \dots, 180^\circ$.

All the above procedures generated for each grain a set of distinct Laue patterns (masks). By applying each mask (P_{gr,φ_i}) for a specific grain orientation, at a particular φ_i to the corresponding Laue pattern ($I_{\varphi,x}$), the intensities of all the matching peaks are harvested and accumulated using determining scattered intensity $I_{gr,\varphi,x}$.

$$I_{gr,\varphi,x} = \sum I_{\varphi,x} (P_{gr,\varphi_i}) \quad (4.19)$$

The above was applied at all (φ, x) positions for each grain $gr = 1 \dots 5$ separately, formulating the intensity sinograms, somewhat analogous to the absorption tomography sinograms discussed in previous section.

Concluding the section, two principal aspects of the reconstruction procedure shall be reported.

The first is the intensity normalisation across each scanned slice, i.e. at each rotation step (φ_i). Considering the analogy between absorption tomography and Laue orientation discussed

previously, the integral of the “harvested” intensity across each slice should be equal at all φ_i . This derives from the fact that at each φ_i , over the entire x-scan, the total illuminated volume (sample) must be the same. Thus, the total scattered intensity collected was considered to be constant at all slices and proportional to $\sum_{j=1}^{56} I_{gr,\varphi,x_j}$. Hence, the normalisation was performed according to:

$$\bar{I}_{gr,\varphi,x} = \frac{I_{gr,\varphi,x}}{\sum_{j=1}^{56} I_{gr,\varphi,x_j}} \quad (4.20)$$

The second observation that significantly improves the results of the grain reconstruction is to take into account and ignore overlapping Laue reflections that carry significant intensity, thus confusing the reconstruction procedure and reducing the quality of results notably. This is usually the case at twin boundaries between adjacent grains that generate partially overlapping Laue patterns. By identifying the symmetry relation on grain boundaries, the algorithm omits the individual overlapping predicted Laue spots from the generated mask. The following picture (Fig. 4.27) shows a contour plot of all the individually reconstructed grains separated by lines around their periphery.

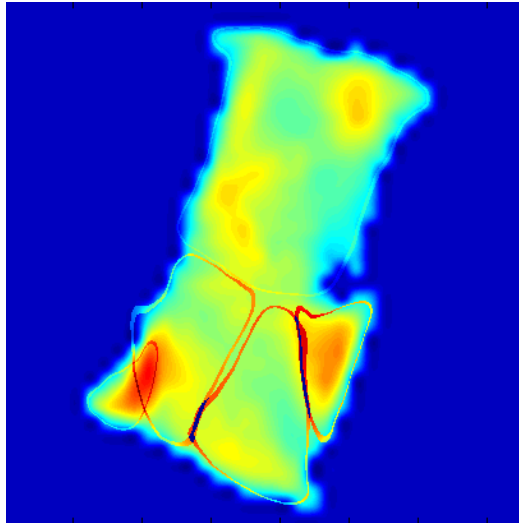


Figure 4.27: Contour preliminary representation of the grain reconstruction, showing overlapping boundaries of grains. This result indicates twining relationship between the grain boundaries.

4.6.4. Rodriguez Vector formulation

For the purpose of reconstructing the relative mis-orientation within each grain, rather than the large scale relative orientations of distinct grains a different strategy was employed. The concept discussed here is based to the property of the Rodriguez vectors (RV) according to which for small mis-orientations (i.e. $<0.1^\circ$), RV s are additive.

A convenient representation of rotations through the Rodriguez Vector formulation (RV) is given here. A rotation matrix $R_{\hat{\omega}}$ can easily be defined using a RV $\hat{\omega} = (\omega_x, \omega_y, \omega_z)$ and a Rodriguez angle according to the following relation:

$$R_{\hat{\omega}} = e^{\tilde{\omega}} \approx \mathbf{I} + \tilde{\omega} \quad , \quad -\cos \theta) \quad (4.21)$$

where: $\tilde{\omega} = \begin{bmatrix} 0 & -\omega_z & \omega_y \\ \omega_z & 0 & -\omega_x \\ -\omega_y & \omega_x & 0 \end{bmatrix}$, $\mathbf{I}$ is the identity matrix and θ the angle of rotation around the

axis of rotation represented by the unit vector $\hat{\omega}$. In the context of the specific problem of intragranular mis-orientation characterisation, the relative rotations within a grain do not exceed 0.1° , $\sin \theta \approx \theta$ and $\cos \theta \approx 1$. Then the (4.58) for RV formulation simplifies to:

$$R_{\hat{\omega}} = e^{\tilde{\omega}} \approx \mathbf{I} + \tilde{\omega} \quad (4.22)$$

The process is based on the ability, of the above algorithmic implementation, to capture and eliminate Laue patterns for an individual grain from a collected pattern of a multi-grained material cross-section. The masks are applied and collect specific Laue patterns corresponding to individual grains, saving them as separate image (TIFF format) files. Moreover these sets of images are indexed and further refined before rotation matrix output is saved in the form of RV components. This approach required several refinement steps of all five recovered grains individually at all translation-rotation positions.

The relative mis-orientation distribution (unknown parameter) can therefore be reconstructed according to the analogous approach used in DST. In this case the *2-D density*

distribution function from absorption tomography is replaced by the *2-D orientation distribution function* ($\rho_{RV}(x, y)$) in the sample cross section (*a priori* unknown).

The measured Laue patterns represent the *average path values* ($\langle R_{RV}(x, \varphi) \rangle$) of through-thickness grain orientation of the sample at each point ($L(x, \varphi)$). This is directly comparable to the confirmed hypothesis for *mean elastic strain variation* in DST. Arranging the treated data in a “sinogram” type graph an identical shaped pattern is created that contains the raw orientation information at each point and is called “*ori-gram*”. Therefore Eq. 4.2 can be rewritten as:

$$\langle R_{RV}(x, \varphi) \rangle = \frac{\int_{L(x, \varphi)} \rho_{RV}(x, y) dy}{\int_{L(x, \varphi)} dy} = \frac{\int_{L(x, \varphi)} \rho_{RV}(x, y) dy}{L(x, \varphi)} \quad (4.23)$$

Consequently, in order to prepare the data in the form suitable for filtered back-projection, the experimentally measured values must be treated as follows, by reformulating Eq. (4.3):

$$p_{RV}(x, \varphi) = \int_{L(x, \varphi)} \rho_{RV}(x, y) dy = \langle R_{RV}(x, \varphi) \rangle L(x, \varphi) \quad (4.24)$$

The same procedure is followed with respect to the calculation of the path length $L(x, \varphi)$ of the beam travelling through individual gauge volume at different (x, φ) positions.

4.6.5. Results and discussion

In this section the results of the first application of *Laue Orientation Tomography* (LOT) on a polycrystalline large grained commercially pure Ni sample are presented. The reconstruction of the grain outline and relative grain mis-orientation was captured at a cross-section 1.92mm below the top. Micrographs of the sample revealed *a priori* several grains through thickness of the particular cross-section as a result of the annealing and extensive re-crystallization. The reconstructions were carried using an Algebraic Reconstruction Technique (ART) approach implemented in MATLAB[®]. The specifics of the ART implementation are described by Kak and Slaney [19].

Grain shape reconstruction

Indexing at θ revealed 5 grains in total. Following the techniques described in previous sections, diffracted intensity "sinograms" of the individual grains were obtained. The sonogram for grain 1 presented in Figure 4.28 showed good agreement with the anticipated structure of an absorption tomography sinogram. Moreover, the enhanced intensity "harvested" techniques compared to the first application of the method, on the same data by Hofmann et al. [24] was obtained by the ellipsoidal masking procedure and the partial elimination of coinciding Laue spots from different grains during the intensity "harvesting" procedure. Only few slices (angular increments) appeared to carry artefacts that are overall insignificant to the reconstruction result. Furthermore, the outline of the sonogram was somehow blurred at the edges of each slice. This was due to second order diffraction maxima generated from the slits that define the beam size. The beam size was somewhat bigger than the anticipated due to these multiple scattering phenomena. For that reason, at the edges of the sample at each rotation step Laue spots were collected from the detector even though the straight through beam didn't come across the sample. Better definition of the beam size (addition of scatter guard upstream of the sample on the beam axis just before the sample) would give better definition of the illuminated volume.

The reconstructed shape of grain 1 illustrated in Figure 4.29 shows sharp features, with reasonably uniform intensity in the centre of the reconstructed grain shape. At the edges of the reconstruction, some blurring is observed, with the edge spreading out over up to four pixels in places. Grains 2 and 4 were examined together due to their twinning relationship. The orientation change between them corresponds to a rotation by 109.5° about the $[011]$ crystal axis. This is one of the $\Sigma 3$ twin rotations for FCC structured materials [191]. This posed a great limitation at the first stages of the handling of the data. Overlapping Laue patterns from the twins were gathered from the applied masks, thus significantly influencing the reconstruction result. Especially at the interface of twins where significant change of orientation occurred, whilst

symmetry was kept, generated individual high intensity streaking Laue spots on the detector. Eliminating these spots allowed separate definitions i.e. sinograms for these grains.

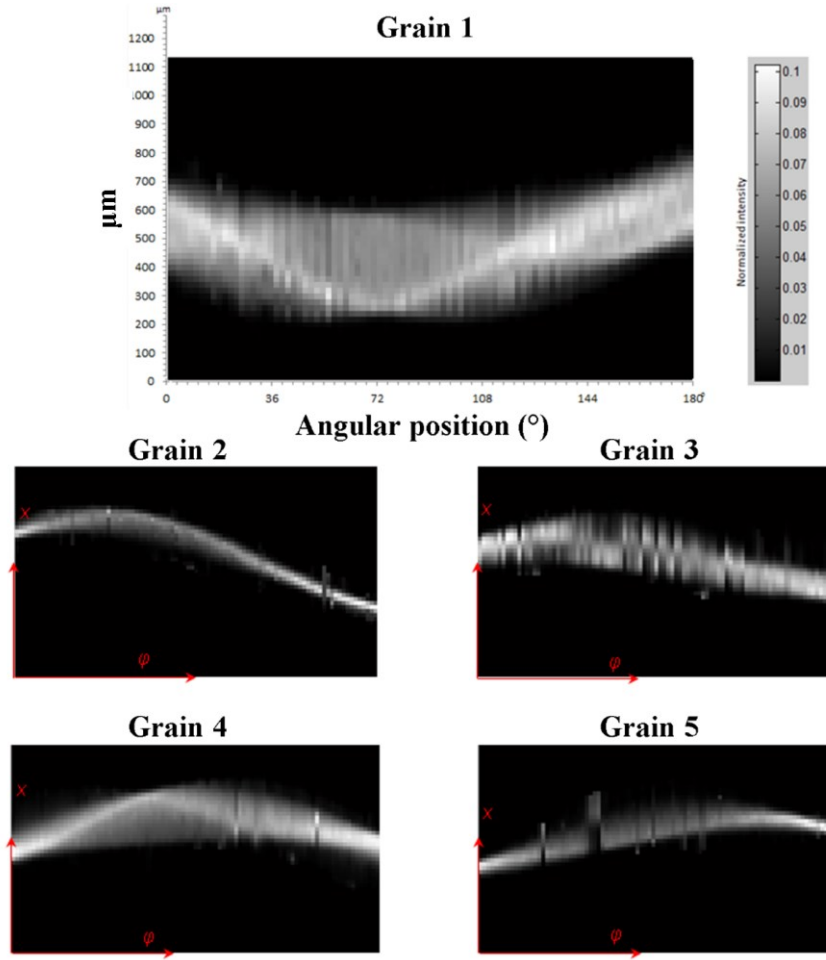


Figure 4.28: Diffracted intensity sinograms for individual grains no 1 to 5. The horizontal axis refers to angular increment. The vertical axis is scanning positions at x_j , where $j = 1 \dots 56$ in $20\mu\text{m}$ increments.

The origram for grain 2 shows remarkable accuracy with only individual slices failing to capture the correct outline (slices 162° - 164°), plus the intensity profiles demonstrate a smooth profile despite the resolution ($0.2\mu\text{m}$) of the beam. The reconstruction technique used is not sensitive to the erroneous slices, whereas the low spatial resolution (big beam-small grain) together with the small number of projections creates significant artefacts at the reconstruction. The grain structure is nevertheless captured and matches the reconstructed geometry of its twin (4th grain). The sinogram for grain 4, as shown in Figure 4.28 carries significantly stronger artefacts but still relatively limited within the outline of the reconstructed grain. This means that the overlapping twin patterns were successfully removed. The Laue spot “harvesting” procedure failed to recover sufficient intensity in regions falling within the sample outline at

individual slices-angles (ϕ). This was as a result of missing out patterns from the masking procedure. The prediction of the anticipated patterns mainly depends on the success of the initial fitting process, thus making it liable to mistakes even for small initial fitting error. Further development is needed in this direction. Reconstructing grain 4 clearly demonstrates the complementary shape of the twin boundary grains. No further masking or thresholding was applied to enhance the result in order to show the full information coming from the diffracted intensity distribution.

For grains 3 and 5, a similar twinning relationship was observed. Their orientation change corresponds to a rotation of 70.5° about the $[\bar{1}\bar{1}0]$ crystal axis. Grain 3 has a non-uniform sinogram as shown in Figure 4.28 whereas the sinogram for grain 5 shows a more successful intensity distribution along with erroneous individual slices. This error typically occurred as a result of harvesting Laue spot intensity, generated from totally different grains by the applied masks. Furthermore, their reconstructions clearly demonstrated their shared twin boundary, showing typical shape and twin boundary behaviour for the annealed commercially pure Ni sample used. Similar artefacts can be observed at the outline of the cross-section of the reconstruction (Fig. 4.29).

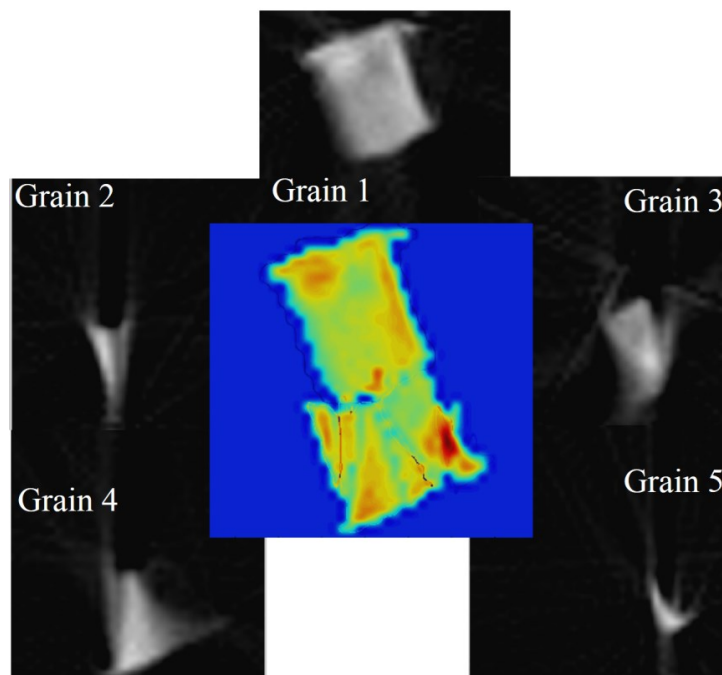


Figure 4.29: Reconstructions of grains 1 to 5 using Algebraic Reconstruction Technique (ART) approach. The inset in the middle shows the contour of added intensity contributions from individual grain reconstructions.

Relative mis-orientation reconstruction

The results of the mis-orientation reconstruction in terms of Rodriguez vector representation are also reported in this section. Following the strategy stated previously for every grain, three separate *ori-grams* were generated and processed for each component of the vector quantity being reconstructed. Each *ori-gram* contains a separate component of the Rodriguez vector as shown in Figure 4.30 for grain 1. Since the shape of the *ori-grams* is now well defined due to the individual sets of patterns collected for each grain, the attention is now drawn to the individual orientation distribution at each slice. The precision of indexing plays a significant role at this stage for being able to successfully depict the slight variations of the individual vector components of the RV as the sample goes through the intermediate rotation positions. At each slice (φ) the pattern for a specific grain is indexed monitoring the relative mis-orientation variation across the sample (x). The maximum variation of relative mis-orientation observed was less than 0.1° in all cases. This is rational according to the un-deformed configuration of the sample and the deployed mask angular tolerance set at 0.17° . Another sanity check that was successfully carried out on the *ori-grams* was the exact opposite orientation change profile of the indexed patterns at 0° and 180° .

The result from the reconstruction of all grains in the bulk RV components is illustrated in Figure 4.31 in an EBSD representation using MTEX library implemented in MATLAB[®]. Further details of the pixel-wise orientation distribution are given in the insets. The orientation of individual grains was confirmed by carrying EBSD experiments on all four specimen sides. A fine grinding procedure was also followed trying to unveil the examined surface (measured 1.92mm away from the top end) within the specimen bulk in order to be EBSD characterised for direct validation. However the steep grain formulation of grain structure and the limited control of the grinding height allowed only grain 1 orientation to be found in the bulk.

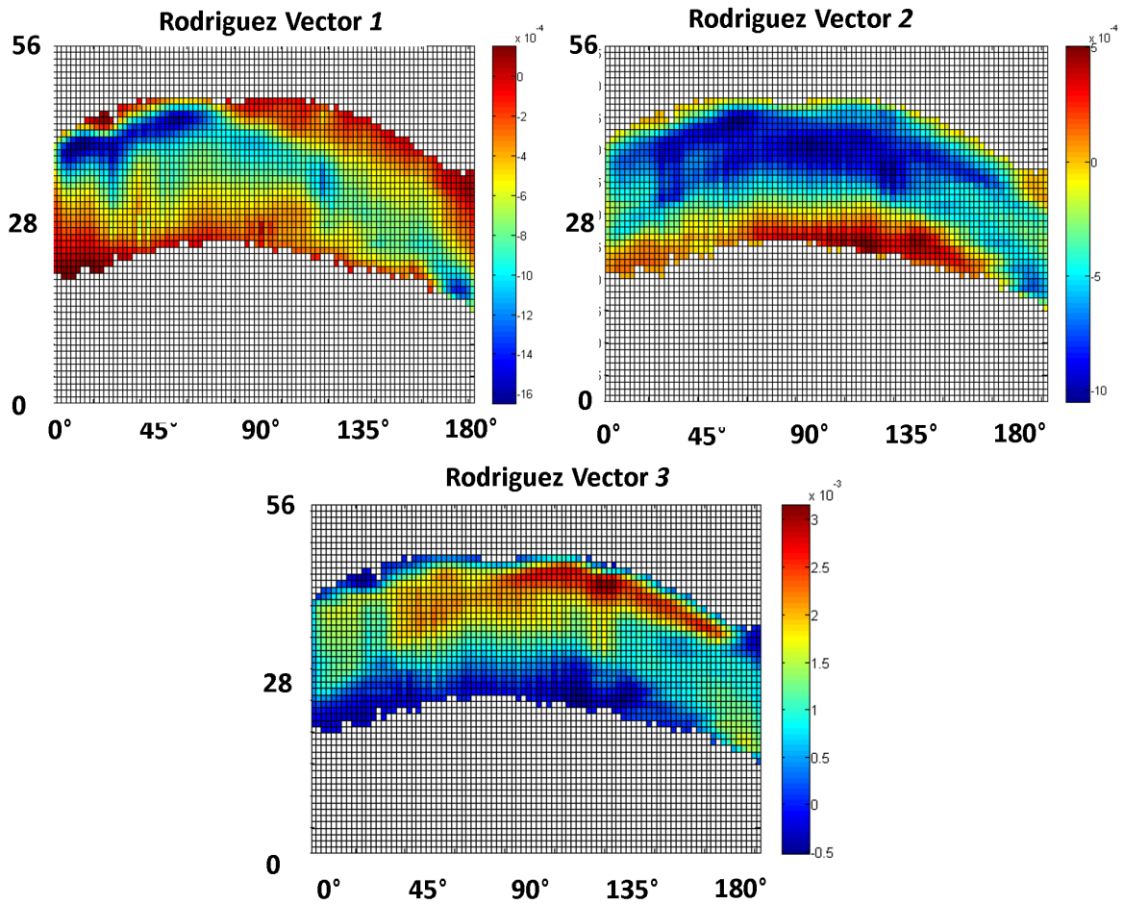


Figure 4.30: Produced “ori-grams” for individual RV components for grain 1 as a function of ϕ_i , $i=1 \dots 90$ (horizontal axis) and x_j , $j=1 \dots 56$ (vertical axis).

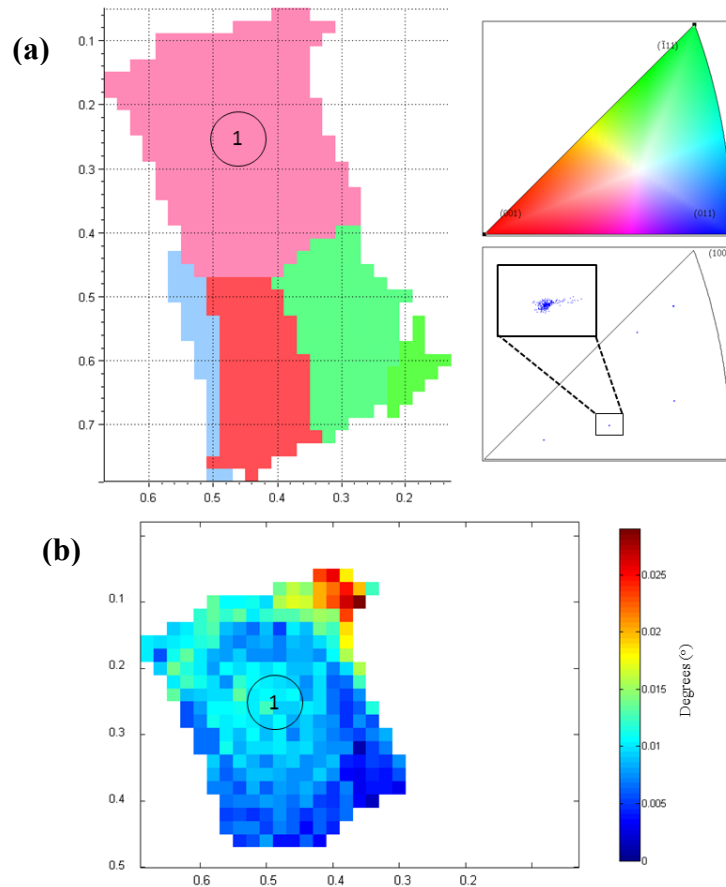


Figure 4.31: (a) EBSD map for the reconstructed slice (*all grains*). (b) Contour plot of intergranular misorientation (*grain 1*).

4.7. Conclusions on ‘rich’ tomography techniques

The Chapter reported the contribution towards “rich” tomography techniques in DST and LOT implementation.

In the case of direct laser deposited Ni-based superalloy turbine blade, a deep structural and dimensional precision examination showcased the preferred orientation due to followed mechanical process and the details of inner cooling channel network, by the use of synchrotron based parallel beam absorption tomography. Extensive analysis towards correcting geometrically imposed pseudo-strains in DST method that appear during data collection as a result of the blades shape were performed. The DST strain result was validated by the well-established contour method at the approximate height where the diffraction data were collected and applying the contour method for residual stress calculation. The result for out of plane component of strain (ϵ_{33}) for DST and stress (s_{33}) for the contour method, showed a consistent tensile stress-strain at the periphery of the blade and overall compressive behaviour in the bulk.

A multi-scale DST approach was implemented on a hierarchical mineralised human dental tissue, consisting of dentine and enamel which formulate a two-level composite structure. A multi-phase approach, by exploiting the tomography acquired data, selectively mapped dentine and enamel diffraction peaks and fitted the out of plane residual elastic lattice variation. Moreover, for the first time a SAXS tomographic acquisition approach was implemented reconstructing the crystallite size variation, confirming significant size variation in the DEJ boundary. The in plane strain component relaxation analysis, using combined FIB-DIC semi-destructive approach, revealed a consistent strain variation in dentine and a steep variation of strain in the enamel part close to the DEJ, very similar to the reconstructed data for ϵ_{33} component tackled by DST.

On the third implementation of DST method an applied load distribution during *in situ* data acquisition on an FPD tooth bridge. Parallel to the experimental implementation a FE model was developed using direct meshing of the tomographic reconstruction of the structure in order

to validate the results. A successful match in terms of strain magnitude and overall strain distribution variation was accomplished. A complete match between the experimental techniques was not possible due to the reasons identified in the section. However, it is possible to assert with confidence that DST is a valuable research technique for experimental stress analysis in complex, high value components used in dentistry. This tool opens the way to elaborate on experimental investigation of the effects of FPD support arrangements on stress distributions, and hence strength; validation of numerical models; experimental analysis of the effects of heat treatment, application of porcelain veneer, fatigue, corrosion, etc.

A Laue based tomographic approach towards precise characterisation of internal structure at a sub-micron level within the bulk of arbitrary shaped materials was proposed. In the first implementation of Laue orientation tomography an annealed polycrystalline 99% commercially pure Ni rod sample was tested. LOT is able to simultaneously reconstruct, non-destructively, the 2-D distribution of relative mis-orientation within a cross-section of a grain in a polycrystalline aggregate. Key advantage of the proposed method is comparatively faster data acquisition in comparison to the extensive wire scans in the case of DAXM, or 360° angular rotation of the sample during DCT scans at very small increments ($\sim 0.02^\circ$). The implementation of Algebraic Reconstruction Techniques (ART) further improved the reconstruction results, making 3-D intragranular lattice mis-orientation and elastic strain determination feasible. Preliminary, testing has showed that 5° increments around 180° of total angular rotation can be sufficiently reconstructed using ART, without significant information loss. Furthermore, a better collimated beam definition (addition of secondary slits right before the sample) of even smaller sized incoming beam ($\sim 10\mu\text{m}$) will dramatically increase the resolution of acquisition. In this case one always has to find a compromise between the concurring requirements of ultimate spatial resolution and available time. Spatial resolution is dependent on the size of the incident beam and the size of the beam dictates the number of scanning points across the object at each rotation step. At the moment the $20\mu\text{m}$ spatial resolution of LOT is still fairly low

compared with the 7 μm achieved with 3DXRD [192] and the lower limit of 10 μm grain size accessible by DCT [42].

The crucial advantage of LOT lies in its potential to provide intragranular measurements of lattice strain and of orientation at high angular resolution, which is currently difficult using either 3DXRD or DCT. Nevertheless, limitations of the method can be readily found on the capability of indexing software to distinguish a large number of grains diffracting at specific positions. Especially in samples with large orientation spread in the scattering volume, streaked reflections might lead to difficulties of indexation and refinement, potentially confining this technique to low deformation samples.

Chapter 5

***In situ* ‘hybrid’ strain mapping techniques**

The knowledge of multi-component, spatially and temporally varying stress-strain states and structural information is crucial for developing a thorough understanding of complex mechanical structure deformation mechanisms. The state of the art 3rd generation synchrotrons offer high brilliance X-ray beams, as well as a variety of detector devices (CCDs, EDX detectors, photon counting diodes), optical focusing and collimation arrangements for adjusting the probe size. Flexible multi-purpose sample environments, in dedicated engineering setups, allow for high complexity *in situ* and *in operando* engineering test rigs to be created and utilised. *In situ* thermal treatment, chemical reactions, as well as various modes of mechanical deformation can therefore be investigated using appropriate beamline operation modes. This offers special opportunities for direct characterisation of various manufacturing processes.

As discussed in Chapter 1, it is important to emphasise the significance of choosing the appropriate length scale, i.e. the spatial resolution one can achieve at the target sample. For the purposes of investigating large engineering structures and components the combination of penetration ability and high flux are the main requirements. The former property ensures that large metallic assemblies, such as an internal combustion engine (ICE) can be investigated in total without compromise in their structural elements and operating environment. Moreover, transmitted or diffracted X-ray beams must possess sufficient intensity for efficient detection,

providing a fast and spatially resolved measure of the internal arrangement of components and their deformation.

In parallel, examples of miniaturized engineering components commonly used in microelectronic applications (thin films, micro-electronic circuits) or microscopic reinforcing agents in engineering alloys (carbon fibres, inclusions) require sub-micron spatial resolution to enable the probing of finer volumes' lattice characteristics and texture. This is accomplished by focusing optics such as sets of Kirkpatrick-Baez (KB) mirrors or compound refractive lenses (CRL) which reduce the effective beamsize to nano-scope probes. In addition, surface discontinuities, intragranular structure and local features on macroscopic samples are the mainstay of modern engineering design and material behaviour research. For efficient and reliable designs, the study of deformation behaviour at local the microstructural scale must be incorporated in the macroscopic design.

Table 5.1: Outline of the experimental conditions for each case study.

Case Study	Beam Probe Size (μm)	Length scale of Characteristic Specimen Features	Average acquisition time (ms)
In operando mechanical analysis of engineering components	100	Single cylinder 124cc IC engine Bore $\times$ Stroke : 55.0 $\times$ 52.2 mm ²	0.35-0.5
Stress evaluation in thin films: micro-focus synchrotron X-ray diffraction combined with Focused Ion Beam patterning	30	0.8 $\times$ 0.8 μm^2 "islands" on 75 $\times$ 75 μm^2 patterned region	10.000-30.000
Nano-scale mapping of lattice strain and orientation inside carbon core SiC fibres	0.3	35 μm $\varnothing$ carbon fibre core 130 μm $\varnothing$ SiC fibre	30.000

Over the last decade or so, high spatial resolution X-ray experimental setups under *in situ* loading are becoming increasingly available at dedicated engineering beamlines at large scale facilities such as neutron scattering centres and synchrotrons. However, the capacity to accommodate full-scale engineering devices and machine testing rigs is somewhat limited. In this Chapter, "hybrid" imaging and diffraction experiments using synchrotron X-rays are presented in the form of case studies listed in Table 5.1 in the hierarchical order of increasing

spatial resolution. Nevertheless, it needs to be emphasized that the demarcation of the experimental studies according to spatial resolution is not necessarily suggesting increasing complexity. The specific nature of each case study requires the adaptation of certain engineering methods, discrete experimental setups at different instruments, as well as significant variations in the acquisition strategy. Since it is impossible to set a single common basis for a systematic approach towards high-resolution state-of-the-art *in situ* and time-resolved engineering experiments, the present Chapter presents useful case studies in this field.

The **first case study** describes the details and implementation of an *in operando* mechanical analysis experimental setup. The project investigated the feasibility of using synchrotron X-ray beams for time-resolved *in situ* imaging and diffraction of the interior components of an internal combustion engine during its operation [20]. The experiment was carried out on beamline I12 (JEEP) at Diamond Light Source, U.K. The world-first application of the *in operando* acquisition on a full-scale internal combustion engine presented a sequence of challenges that are discussed in detail below. For the purposes of the experiment, a coupled time-resolved X-ray imaging and energy dispersive diffraction setup was employed. A series of X-ray "movies" were recorded for various engine components *in operando* using stroboscopic acquisition approach. Finally, the dynamic internal strain within the moving con-rod was evaluated with the accuracy of $\sim 50 \times 10^{-6}$ at a spatial resolution of $100 \times 100 \mu\text{m}^2$.

The **second case study** addresses the challenge of identifying and mapping the strain in a Focused Ion Beam (FIB) milled region ($75 \times 75 \mu\text{m}^2$) at the surface of a nanocrystalline metallic coating of sub-micron thickness [23]. Transmission X-ray diffraction setup was used for data collection at DIFFABS beamline (Synchrotron Soleil, France). A 150nm-thick multi-layered W-Cu nano-composite thin film on polyimide (Kapton®) substrate was studied. under *in situ* uniaxial tension. Micro-beam diffraction data at a $30 \times 30 \mu\text{m}^2$ spatial resolution were collected on and away from the reference array using a 2-D X-ray diffraction scanning approach for "on-board" FIB pattern identification. It was shown experimentally that the "island" array remained approximately strain free throughout the experiment, providing an on-board do lattice spacing

reference. The elastic strain evolution during deformation on the sample was monitored and associated to the macroscopic applied strain from the rig.

In the **third case study** an ultra-high resolution nano-probe was used to evaluate the residual strain profile within unidirectional silicon carbide fibres with carbon monofilament core. A transmission X-ray diffraction setup, and optical focusing down to a $0.3 \times 0.5 \mu\text{m}^2$ spatial resolution was used for data collection on beamline B16 at Diamond Light Source, U.K. By combining synchrotron-based imaging and nano-focused X-ray beam scattering with Focused Ion Beam stress evaluation, detailed maps of structure and strain inside this material are presented [23]. Eigenstrain modelling was used to reconstruct the full residual strain state within the fibre cross-section. The joined-up experimental and theoretical approach allows extracting information about fibre structure down to the nanoscale, developing insight into its processing history, and revealing the existence of significant residual strains that have a strong effect on the performance of CFs in service.

5.1. In operando mechanical analysis of internal combustion engine by synchrotron time resolved X-ray diffraction and imaging

Internal combustion engine (ICE) is a widely used complex thermo-mechanical system which consists of numerous components. The combination of a compact design and overall output power density has ensured its undiminishing popularity for over a century of exploitation. The ignition stroke within the fuel-air mixture created in the combustion chamber is initiated by a spark. This results in the rise in the gas temperature and pressure, thus generating a force on the piston. The force generated during the reciprocating motion of the piston is transmitted through connecting rods (con-rods) to cause the rotation of the crank-shaft and producing exploitable mechanical torque. A significant amount of simultaneous interactions between different engine components gives rise to a complex environment of coupled variables that have to be determined, during engine operation, for the purposes of performance evaluation and optimisation. A thorough investigation takes into account gas cycle measurements within the

cylinder, incorporating information about flow rates, gas pressure, temperature together with stiffness and noise, vibration, and hardness (NVH) conditions. Experimental measurements are therefore important to establish comprehensive models describing the thermo-mechanical behaviour of the engine and establish a suite of useful predictive tools that can then be used for design and optimisation.

A variety of sensors, data logging and optical imaging devices are tools often used to obtain measurements of the ICE operating parameters. For example, thermocouples can be embedded in the cylinder block to record the temperature history, and strain sensors can be used to obtain measures of deformation at the surface of the assembly. Moreover, customised transparent piston-liners have been widely used in research to provide time-resolved and location-specific measurements particularly to do with the fuel-air mixture and ignition initiation propagation within the combustion chamber. However, obtaining strain-stress temporally and spatially resolved information *non-destructively* from *inside* the operating combustion engine is a much greater challenge. The harsh mechanical and thermal environment presents a significant challenge for conventional sensor systems.

An investigation presented by Baimpas et al. (2013) [20] was focused in imaging the movement of the con-rod attached to the reciprocating piston, together with the valves regulating air inlet and exhaust. Further investigation of the feasibility of mapping the spatial distribution of internal elastic strains in the moving con-rod was conducted. The specification of the engine used for the implementation of the experiment is shown in Table 5.2. Evidently, this kind of spatially resolved measurement also has to be sufficiently fast, since the stress distribution varies continuously over the period of crank-shaft revolution. It is important to emphasise that the principal benefit of the proposed methodology is that it can serve model validation and design optimization for internal combustion engines, ultimately helping improve fuel efficiency, power output, reliability, and also reduction of the amount and weight of material used.

In service, a single con-rod experiences repeated loading that runs to a high number of cycles (10^8 - 10^9). The type of loading ranges from compression induced by the pressure of combustion gases on the piston to tensile and bending loads due to inertial phenomena. FE analysis [193] has been carried out to predict the dynamic multi-axial stress response of con-rods in service. However, the fidelity of such models depends strongly on the availability of correct boundary conditions and other data, such as the time-dependent temperature and pressure of combustion products. A number of further unknown parameters remain, namely, those related to friction, the engine's response to external load (e.g. the forces that arise from road interaction) and the complex coupling between the engine's mechanical performance and ancillary systems (cooling, oil and fuel circuits). These aspects of the problem introduce further challenges for FE simulation and necessitate additional characterisation via dynamic experimental measurements in operating IC engines. Strain gauges can be attached to the con-rod and used to acquire continuous deformation data to be captured and transmitted or stored [194-196]. However, this approach suffers from the limitations associated with strain gauges being sensitive to temperature and thermal gradients, as well as the engine vibrations and the difficulty of connecting signal wires to data loggers at high revolutions per minute (RPM) [197].

Table 5.2: Honda ATC single cylinder engine specifications

Engine type	4 stroke, Air cooled
Displacement	124cc
Bore x Stroke	55.0 x 52.2 mm
Compression ratio	8.8 to 1
Compression	156 - 198 psi
Transmission speeds	4 speed with Dual range
Final drive	Chain

Source: Honda

The non-destructive nature of X-ray imaging and diffraction is a highly effective means of visualising internal structures at different spatial resolutions [198], and for mapping internal residual and “live” strain distributions [145]. Alongside the classical powder diffraction mode (see Chapter 2), various approaches have been proposed for probing inter- and intra-granular deformation within crystalline materials [41, 153]. The variety of techniques and arrangements that have been put forward [34, 199-203] allows the selection of length scales (from a few mm down to sub-micron) and obtaining information about various material properties (local or

statistical crystal orientation, crystallographic phase composition). In particular, high energy polychromatic beam configuration for elastic strain measurement allows fast data acquisition and lends itself well to the mapping of large sample areas [145]. At DLS I12, a Canberra 23-element Germanium energy dispersive X-ray detector (EDXD) is used for polychromatic powder diffraction measurements.

X-ray imaging, microscopy, topography and tomography techniques have also witnessed rapid development in recent years, due to the advent of improved beam conditioning optics and detectors offering better efficiency and resolution. *Radiography* as well as tomography techniques make use of the penetrating ability of X-ray beams that show differential attenuation depending on the amount of absorption along the beam path. Micron resolution imaging can now be achieved routinely using scintillators coupled to magnifying optics. Achieving nano-scale ($\sim 100\text{nm}$) resolution requires special setups making use of curved (divergent) beam geometries [204]. In terms of temporal resolution, sub-millisecond time resolution can be achieved using rapid imaging [205], for example through using imaging devices such as Vision Research Phantom v7.3 CMOS high speed camera [206].

I12 JEEP Experimental Hutch 2 (EH2) key facts

The ability of high-energy synchrotron X-ray setups to investigate multi-component, spatially and temporally varying stress-strain states and structural information is a significant tool towards assessment of the mechanical behaviour of engineering components and assemblies. The EH2 at I12 JEEP was designed and implemented in order to allow for large scale engineer assemblies to be accommodated. It incorporates a 2 ton capacity sample stage, with a variety of translations at $10\mu\text{m}$ resolution, $\pm 2^\circ$ tilting for alignment of virtual rotation axis for tomography and continuous rotation around the axis perpendicular to the sample stage with 0.0005° resolution for tomographic data acquisition (Fig. 5.1 (b)). A three module detector table similar to that available in EH1 (see Chapter 3) can simultaneously provide support for various counting devices for X-ray, diffraction and imaging and allows for swift interchange between

different detectors in the course of the experiment (Fig. 5.1 (b)). An optics table is also mounted upstream the sample stage right after the beam entry window to allow for optical and alignment devices to be installed for beam definition purposes. The overall hutch dimensions are 11m (*long*) $\times$ 7m (*wide*) $\times$ 4m (*high*). A 5 ton capacity overhead crane and direct access from the outside of the synchrotron building serve the needs of manipulating large components, such as IC engines and pre-assembled test rigs, which can be directly transferred and installed in the experimental hutch. The experimental hutch has also been designed to accommodate large capacity (1500 L/min) air venting for evacuating exhaust fumes at temperatures up to 300°C, large capacity air-conditioning, water cooling, and three-phase power supply in order to facilitate *in operando* experimental studies.

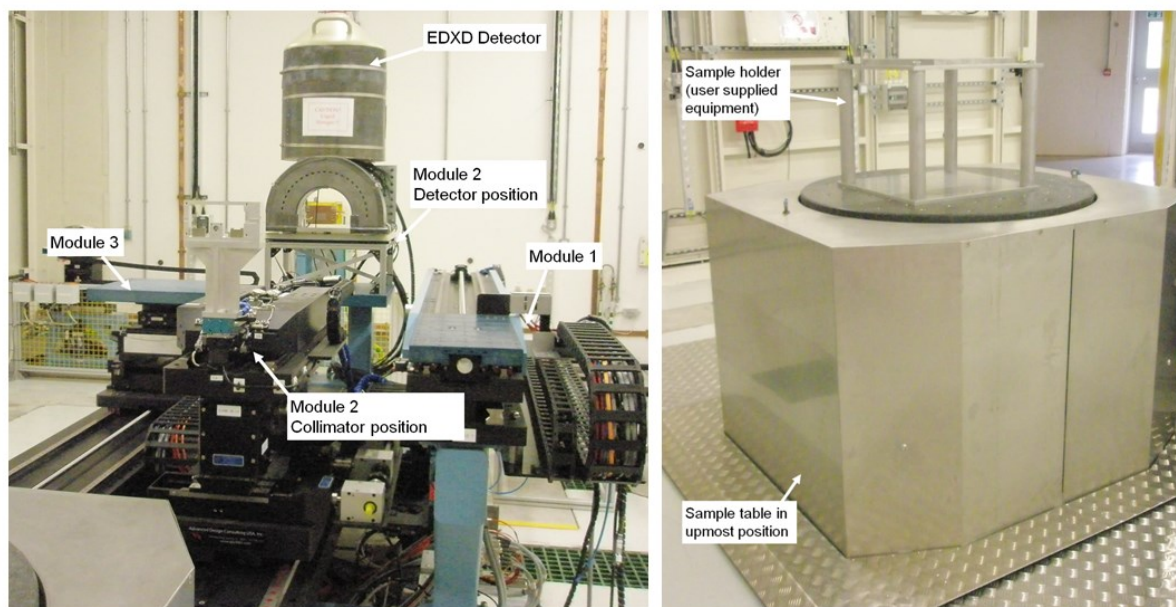


Figure 5.1: Experimental Hutch 2 at I12 JEEP Diamond Light Source; (a) Stage module for accommodating simultaneously different detectors according to the setup; (b) Sample stage [20].

The high energy X-ray spectrum of the beamline allows the useful range of X-ray energies between 50-150keV to be used, over which the beams show sufficient transmitted signal through millimetre-thick layers of materials. The high flux ensures that it is possible to “see” through relatively large metallic assemblies, such as an internal combustion engine (ICE). More specifically, X-ray beams that have been transmitted through or diffracted from internal components must possess sufficient intensity for efficient detection, providing a fast and

spatially resolved measure of the internal arrangement of components and their mechanical behaviour. The beamline acquisition setup also allows manual electronic gating of the detector shutter via TTL pulse generation at high frequencies that is incorporated to the GDA control system and allows for time resolved measurements and stroboscopic acquisition on repeatable processes for improved statistics.

5.1.1. Experimental Setup – EH2 I12, DLS

At the initial preparatory stage of the experiment, composite ("mosaic") radiographic images of the engine block assembly were collected at different crankshaft rotation angles (0° - 120°) while the engine remained stationary such as the one shown in Figure 5.2. This allowed evaluating beam attenuation by the engine assembly, and establishing the conditions for imaging.

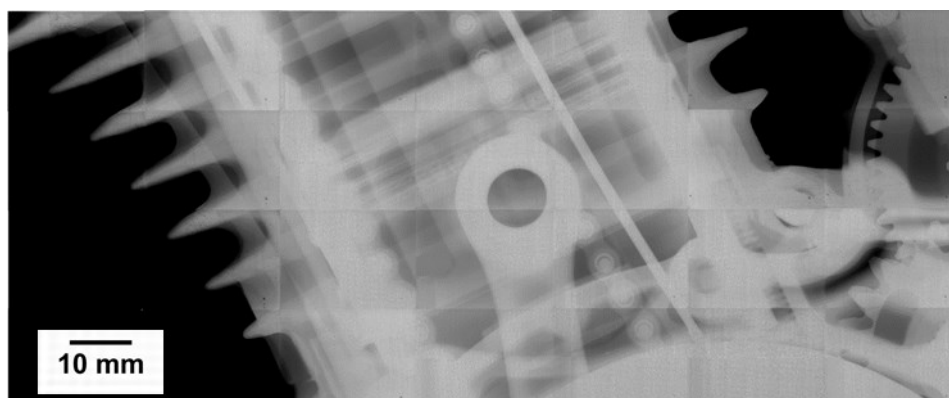


Figure 5.2: Mosaic of engine cross section assembly at 120° crankshaft rotation (inverted greyscale colourmap).

The possibility of carrying out the experiment was contingent on resolving a number of previously unaddressed issues.

X-ray beams undergo strong attenuation when passing through metallic assemblies. Successful data collection requires the engine to be sufficiently radiographically “transparent” for the beam to penetrate simultaneously the engine block, the piston skirt and the con-rod, whilst allowing the exposure times to remain sufficiently short ($<10\text{ms}$ for a single-shot exposure). The engine side view must not be obscured by other moving components, such as the flywheel, timing chain or the complex cooling water circuit channels. For the purposes of

con-rod bending stress analysis, the incoming beam should be parallel to the pin end of the piston, so that the translation of the assembly across the beam would allow high spatial resolution across the con-rod for strain scanning. A typical single cylinder, air-cooled motorcycle engine does not have any of the above limitations, and has an overall lightweight construction. It was chosen as a system that satisfies all the above requirements. The cooling fins forming the engine block outer shell are made from cast Al alloy with a low absorption coefficient. The steel piston skirt has a relatively small thickness ($< 5\text{mm}$), and the con-rod with the I-beam cross-section has a maximum width of 7mm at the side brackets. The configuration of the Honda ATC engine chosen for investigation is shown in Table 5.2. The attenuation of the X-ray beam during its passage through the engine is illustrated on a cross section of the engine in Figure 5.3 (a).

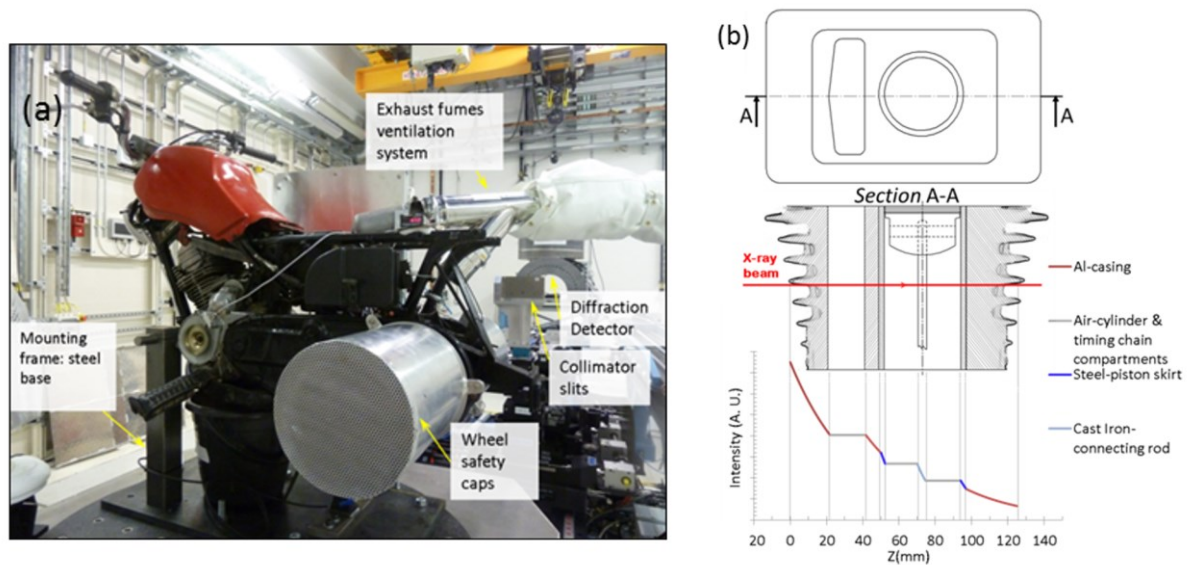


Figure 5.3: (a) Running motorcycle frame mounted on the high capacity sample positioning table of the rig inside EH2, I12, DLS. (b) Beam attenuation through the engine cross section. The position of the cross-section A-A is demonstrated at the top of the figure. The cast Al attenuation is shown in red, the steel-piston skirt attenuation in blue, the cast iron con-rod in cyan and air attenuation in grey. The μ absorption coefficient for each metal is taken by NIST for energy 150keV and the attenuation is according to Beer-Lambert law.

The engine was securely mounted on the translation table available in the external hutch (EH2) of the JEEP instrument (Fig. 5.3 (a)), using a rigid support structure and minimising the vibration effects that would cause blurring during image acquisition and uncertainty in the gauge volume position during diffraction measurements. The configuration was made ready for assembly within the restricted time-frame of the experimental beamtime allocation. For these

reasons, a complete motorcycle configuration was selected, mainly as it already accommodates important ancillary components of the engine (exhaust, fuel tank, electrics etc.). A tricycle Honda ATC 125M was used, together with the specially designed supporting frame that provided a convenient base for mounting on the test bed of the hutch. Additional bracing of the motorcycle frame and shielding the engine moving parts ensured minimal vibration effects and safety in the hutch. The engine mounting arrangement on the steel base frame on top of the beamline test-bed is shown in Figure 5.3 (b), together with the wheel safety covers.

The time-resolved nature of the experiment required precise monitoring of the reciprocating piston movement and the ability to trigger data acquisition (in both imaging and diffraction modes) at specific crank angles, with a high degree of repeatability over consecutive strokes. Signal accumulation was used to achieve sufficient intensity. To synchronise detector triggering with the crankshaft angular position, electronic gating arrangements were put in place. A proximity sensor, connected to a tachometer, was mounted on the motorcycle crankshaft and emitted a TTL square signal pulse every time the piston reached the top dead centre (TDC). The tachometer was able to register the input frequency up to 7.5 kHz and delivered a pulse to a waveform generator that emitted a new TTL pulse with (a) specific delay time, (b) specific pulse width and (c) total cycle period. The delay time was determined from the frequency of rotation of the engine (rpm) according to the desired shaft angle for observation on the con-rod. A specific location was chosen for detailed analysis, corresponding to the distance from the pin end of 20mm. The width of the TTL timing pulse was determined by the desired exposure time of the detector. This was chosen to ensure that the displacement of the con-rod during exposure amounted to less than three pixels of the imaging camera (nominal pixel size 30.30 μ m). This was adopted as a good compromise between achieving sufficient accumulated counts and accuracy of position and gauge volume definition. Due to the engine being a 4-stroke, the detectors were triggered at half the frequency of crankshaft rotation, to ensure consistent sampling of the engine cycle. A schematic diagram of the triggering configuration is shown in Figure 5.4.

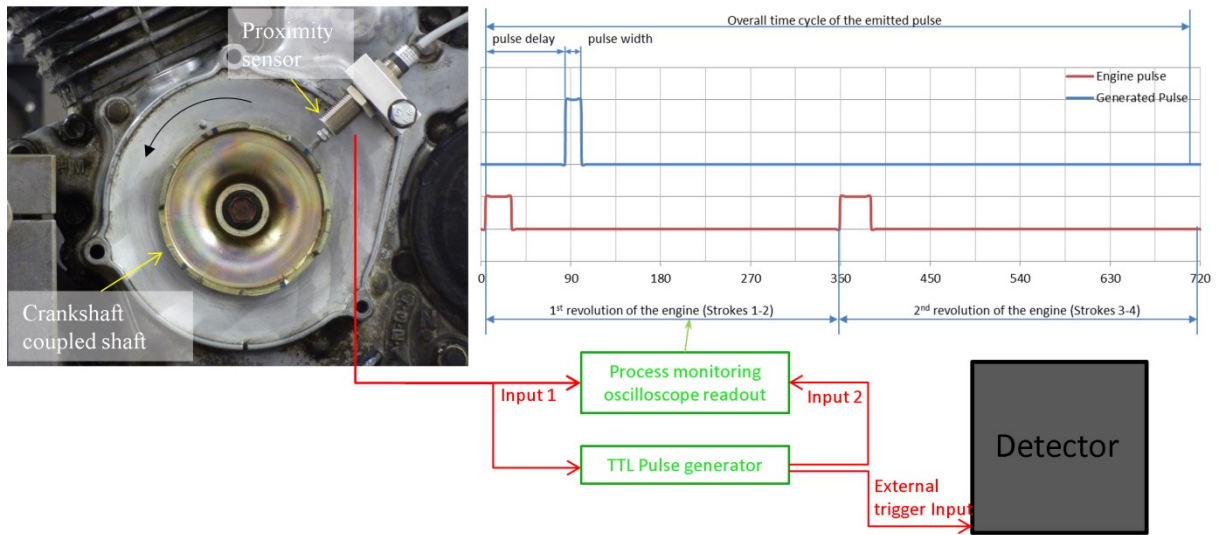


Figure 5.4: Schematic illustration of detector triggering setup. A pulse is triggered every 360° , by the proximity sensor mounted on the rotating shaft mounted on 1:1 ratio to the crankshaft. The pulse is fed to the TTL pulse generator and the process monitoring oscilloscope. The parameters, such as Pulse Delay (P.D.) that controls the angular position we are hitting the con-rod according to the RPMs i.e. phase and the Pulse Width (P.W.) which controls the exposure time i.e. the time that the shutter of the detector remains open on every cycle, are adjusted.

The overall time cycle of the emitted pulse is set just under the duration of 2 overall crankshaft revolutions to ensure triggering on the same stroke each time.

The motorcycle setup as illustrated in Figure 5.3 was installed in EH 2 of I12 JEEP. Exhaust fumes were pumped to the outside of the hutch through the purpose-built ventilation system described earlier. A flow regulator (damper valve circuit) was used to regulate the system intake when the engine was not producing the amount of exhaust fumes that the pump was extracting, thus allowing natural flow of the engine outtake stroke. On similar engine test rigs, the fume extractor is usually not attached to the engine, as the engine typically operates in a dedicated isolated chamber. However, in this experiment the vent was fixed to the exhaust system to protect the equipment in the experimental hutch.

For imaging, a broad spectrum white beam with a cross section of $18\text{mm} \times 24\text{mm}$ was directed at the engine. The transmission image through the engine was registered using an X-ray camera consisting of a scintillator (CdWO_4) that was coupled to visible light optics and a high-speed CMOS camera (Vision Research Phantom V7.3). A standard resolution imaging set up used corresponded to the nominal pixel size (accounting for point spread function) of $\sim 30\mu\text{m}$ and the field of view of $17.8 \times 23.8\text{mm}^2$.

For diffraction, the 23-element energy-dispersive “horseshoe” detector was used. The individual detector elements on the detector are arranged at equal azimuthal steps on a half

circle. The detector was aligned with respect to the incident beam to ensure the equal scattering angle of 10° for all elements and two sets of slits provided secondary collimation (as described in Chapter 2 [34]. The incoming beam together with the set of slits define a gauge volume of approximate dimensions $0.1 \text{ mm} \times 0.1 \text{ mm}$ across the beam and $\sim 2.1 \text{ mm}$ along the beam (Fig. 5.5 (a)) [35]. At the set-up stage, the con-rod in the plane perpendicular to the incident beam was aligned using the imaging mode of the beamline.

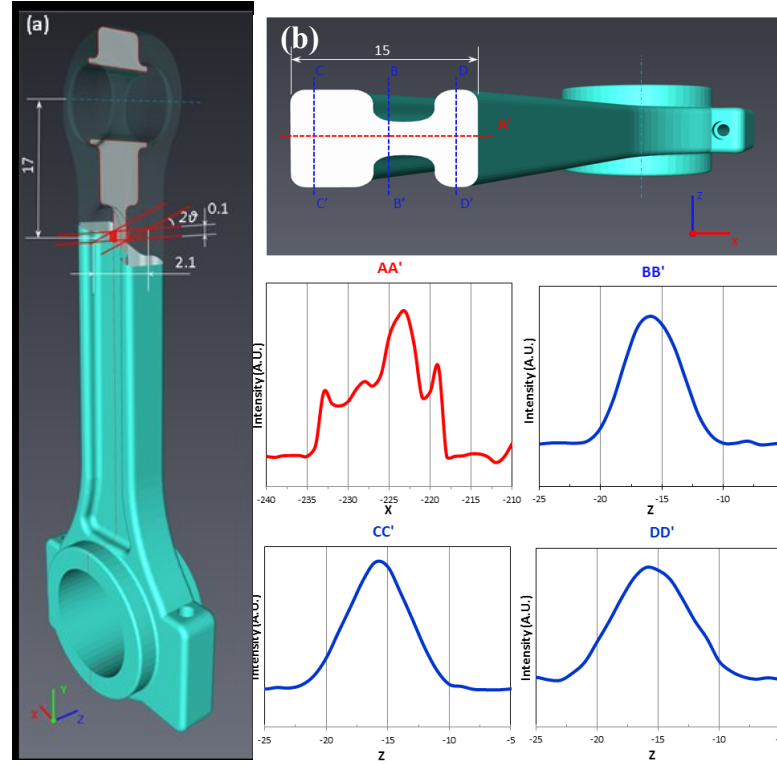


Figure 5.5: (a) Schematic of the con-rod diffraction setup. The inclined cross-section shows the angle at which the con-rod stands with respect to the Y-axis of the setup in I12. Highlighted in red the cross-sectional definition of the gauge volume defined by the double set of collimation slits and the incident beam cross-sectional size ($0.1 \times 2.1 \text{ mm}^2$), (b) Cross-section of the plane analysed by diffraction 17mm below the pin end of the con-rod. Highlighted in red is the x-scan used to determine the overall width of the section and blue –indicates the z-scan to locate the gauge volume in the middle of con-rod. Scan in -x- (AA') and -z- (BB', CC', DD') showing integrated intensity from all channels of the EDXD detector. The observed intensity variation comes from the combination of absorption and scattering power (polycrystalline structure) of the material within the gauge volume.

For the subsequent local strain measurements, the beamline was configured for diffraction. The correct position of the gauge volume with respect to the con rod was confirmed through prior X-ray imaging and through 3-D alignment scans in diffraction mode. The final alignment in diffraction mode was carried out by scanning through the entire cross section of the con-rod along the directions AA', BB', CC' and DD', Figure 5.5 (b). The scan results were used to

confirm that the gauge volume position and the beamline positioning arrangements were in good agreement with the engineering model of the engine.

5.1.2. Compound radiography inside an Internal Combustion Engine

“Live” imaging of the cylinder operation was initially carried out in “one-shot” mode. Since the number of photons available for imaging in this case is severely restricted by the short exposure time, the signal-to-noise levels are high. In Figure 5.6, the inherent background noise structure of the Phantom camera can be distinguished. The larger compound image in Figure 5.6 shows reduced contrast and resolution. The outline of the piston pin is barely visible, suggesting that stroboscopic accumulation of images is likely to be required for the collection of high resolution data.

Figure 5.7 shows a sequence of cylinder valve images during engine operation. In the sequence taken at intervals of 10° phase angle, the valve enters the field of view from above, reaches the lowest position near the middle of the image, and is withdrawn again upwards. Acceptable spatial resolution of the image was obtained, sufficient to distinguish the valve profile.

Fast radiographic image collection was carried out in stroboscopic accumulation mode. To achieve this, the triggering procedure described above was used to control the Phantom high speed camera, and the images were added to improve the signal to noise ratio (SNR). Due to the limited field of view compared to the size of the object and the region of interest, “tiling” of images was required in order to build up an overall picture of the operating engine. Some variation in beam intensity and detector response with time was observed, i.e. different sub-images had different overall intensity. Some beam profile non-uniformity across the field of view was evident.

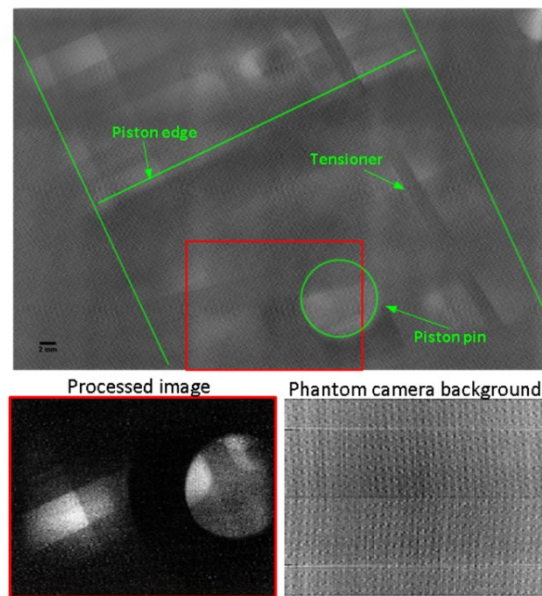


Figure 5.6: Mosaic of engine's cross-section map at 120° crankshaft rotation at 1850 rpm (delay time 10.8ms, exposure time 0.5ms).

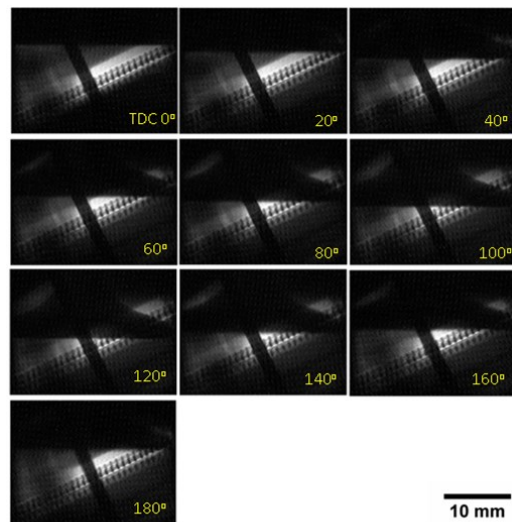


Figure 5.7: Image sequence from the intake valve starting at the TDC piston position at 20° crankshaft rotation increments, showing significant wear of the camshaft lobes at 1850rpm (exposure time: 1ms).

In order to build up the overall radiographic picture of the engine at a particular phase instance during its operation, some image intensity correction was applied. This was accomplished by edge matching between individual adjacent tiles, seeking to ensure a consistent compound image. The overall composite (“mosaic”) image is shown in Figure 5.8. It displays excellent contrast, good sharpness (absence of blurring) and therefore good spatial resolution of engine internal features. The details of the timing chain, gears, internal threads and also the moving crankshaft are all clearly distinguishable.

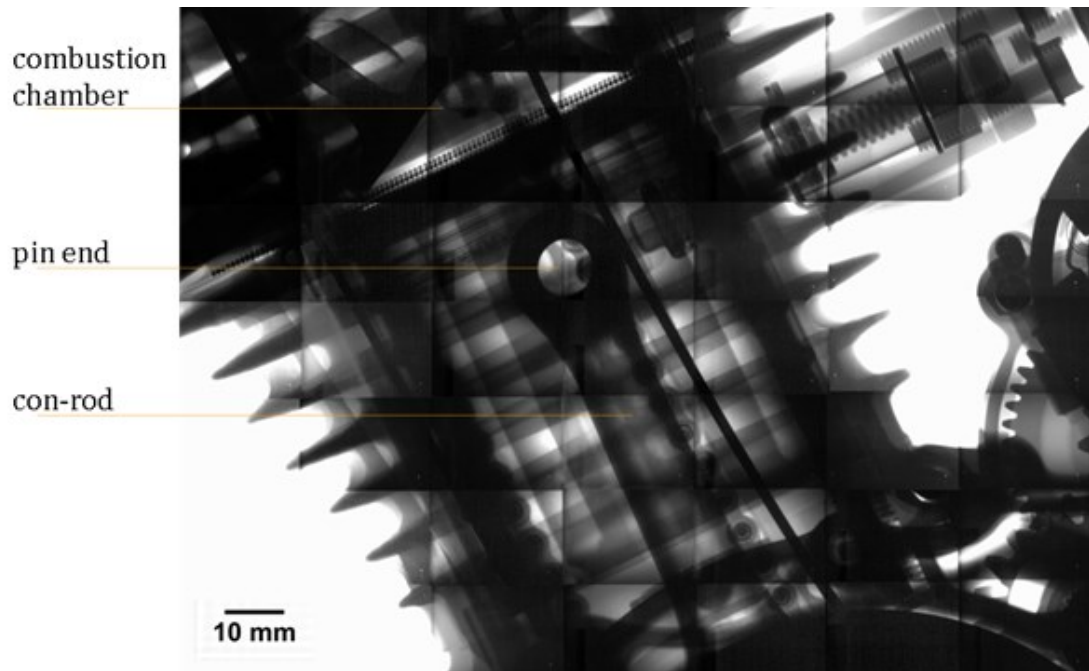


Figure 5.8: Mosaic of engine cross section assembly close to 0° crankshaft rotation (top dead centre, TDC).

5.1.3. *In operando* stress quantification inside Internal Combustion Engines

As discussed previously diffraction of hard X-rays from polycrystalline materials is well-established as a truly non-destructive spatially and directionally resolved measurement method for elastic lattice strain, allowing the macroscopic and microscopic components of stress to be deduced. The ability of energy dispersive X-ray diffraction (EDXRD) and the use of double collimation for the definition of the “gauge” volume across the scattered beam path, managed sufficient levels of diffracted beam transmission through the entire engine block. Whereas a monochromatic setup is limited in terms of the maximum energy bandwidth, due to limitations related to the tilting angles of the double crystal silicon monochromator, EDXRD makes use of the harder spectrum of the beam at energies $>150\text{keV}$ and is able for sufficient penetration through the engine.

“Live” strain measurements carried out in the present study had a spatial resolution $<0.1\text{mm}$ and a time resolution $<1\text{ms}$). The beam was collimated to the size $0.1\text{mm} \times 0.1\text{mm}$ located at the position on the crankshaft shown in Figure 5.9, lying 17mm below the piston pin axis. “Live” strain state at a single point on the engine connecting rod was then evaluated by a stroboscopic accumulation technique applied to diffraction data [207], through synchronisation of the fast

electronic shutter of the EDXD detector with the engine crankshaft angular position, and accumulating the signal over multiple engine cycles. For the cast iron material of the con-rod ($I-m3m$ crystal structure, unstrained lattice parameter $\sim 2.852\text{\AA}$) four peaks $[200]$, $[211]$, $[220]$, $[310]$ were analysed.

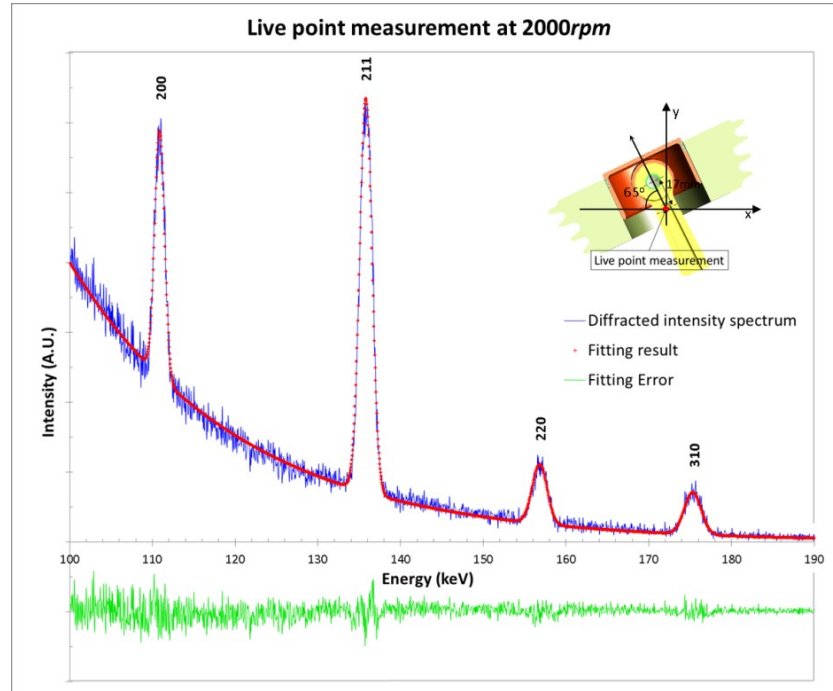


Figure 5.9: Illustration of multi peak fitting result together with the fitting error shown by the bottom curve. CAD schematic representation of engine assembly cross-section with the point where the diffraction measurement was taken is highlighted in red.

A stationary reference residual stress linear profile across the con-rod was collected in advance on the pre-warmed engine con-rod. The *in operando* pattern showed no additional peak broadening compared to the data collected from a static engine. Figure 5.9 illustrates the result of diffraction profile fitting using Pawley refinement method. The background was fitted with a second order polynomial. The green line below the plot in Figure 5.9 indicates the fitting error, confirming that satisfactory agreement was achieved. The derived strain was found to be ~ 630 microstrains (compressive) at 0° during the ignition stroke at $\sim 2000\text{rpm}$.

Figure 5.10 shows detail of fitting for the $[211]$ peak of ferrite located at 135.7203keV ($d_{211} = 1.1634\text{\AA}$). The curve for the stationary con-rod pattern corresponds to the data points shown by “+” red markers. The $[211]$ peak for the “live” measurement had the centre located at 135.8061 keV ($d_{211} = 1.1627\text{\AA}$), and corresponds to the data points shown by “+” blue

markers. The peak positioning error for the pattern taken from the stationary engine was only $\sim 30\text{ppm}$ (30×10^{-6}). A long exposure time of 3000s was used for this measurement to achieve maximum precision.

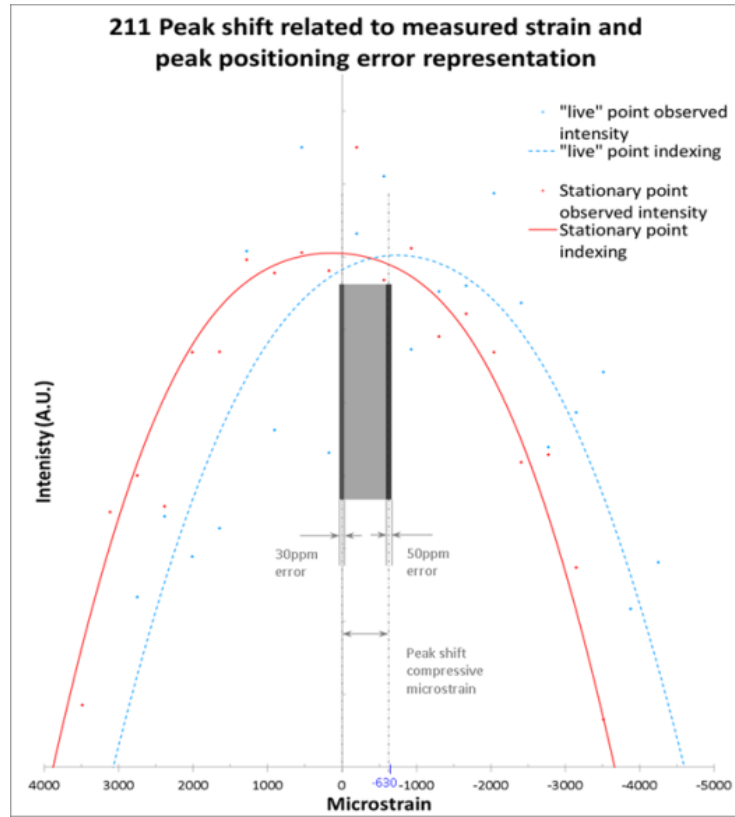


Figure 5.10: Close-up illustration of [211] peak fitting result for “live” measurement in blue and stationary measurement in red. The peak intensities are normalised so that they have the same intensity height distribution. The peak positioning error for the stationary measurement is 30ppm and for the “live” measurement is 50ppm. The measured compressive strain (630 microstrain) represented by the peak shift is highlighted in light grey colour and the peak positioning error margins are highlighted in dark grey colour.

The peak shown in Figure 5.10 was extracted from the diffraction pattern collected when the engine was running. The exposure time for each revolution was 0.289ms with total accumulated exposure time amounting to $\sim 24\text{s}$ during 80 minutes overall engine running time. The accuracy of determining the peak centre position was better than 40ppm (40×10^{-6}). The strain calculation from interplanar lattice spacings computed on the basis of peak centre positions in the reference and “live” patterns (a_0 and a , respectively) is accomplished using Eq. (1.3). The strain in the con-rod during operation was found to be compressive and equal to 630 microstrain (unit of strain measurement equal to 10^{-6}). The accuracy of experimental strain evaluation was thus consistently better than 50 microstrain (50×10^{-6}), as illustrated in Fig 5.10. This corresponds to the nominal stress evaluation result of $\sim 120 \pm 10\text{MPa}$. The small error of

only ~10MPa provides the proof of feasibility of “live” stress-strain analysis in a running ICE engine.

5.1.4. Conclusions

The results of the feasibility study presented concern the combination of imaging and scattering from internal components of an operating internal combustion engine for non-destructive determination of component motion and strain. The results open at least two avenues for further strain analysis. One makes use of the contrast achieved in the imaging mode, and in principle allows displacement mapping to be performed by Digital Image Correlation [208]. Differentiation of displacement maps with respect to time allow speeds to be measured, whilst differentiation with respect to position provides data on the distribution of total strain in engine components. However, since the elastic strains are typically small (<1%), this method is likely to be best suited to situations when plastic deformation is observed. The second approach particularly important for the determination of microscale residual stresses is polycrystalline diffraction that relies on elastic strain evaluation through precise determination of interplanar lattice spacing from X-ray scattering intensity.

5.2. Stress evaluation in thin films: micro-focus synchrotron X-ray diffraction combined with Focused Ion Beam patterning

Nanocrystalline metallic coatings of sub-micron thickness are widely used in modern microelectronic applications. In X-ray diffraction experiments to determine both the residual and applied stresses in nanocrystalline coatings, one difficult challenge that comes up invariably is the determination of the strain-free lattice spacing d_o . As discussed, residual stress distributions are present in most engineering materials and components. In many cases they strongly influence the static and dynamic response of structures, thus affecting their integrity. In thin films bibliography, the residual stresses are separated into two categories: extrinsic and intrinsic. Extrinsic residual stresses are thermal stresses that arise due to the incompatibility caused by (unequal) thermal expansion coefficients of the film(s) and the substrate [209, 210].

At any temperature T , there is a linear relationship between the temperature and the extrinsic thermal σ_T stress:

$$\sigma_T \propto (T_{Dep} - T) \Delta\alpha \quad (5.1)$$

Here T_{Dep} refers to the (effective) deposition temperature and $\Delta\alpha$ is the difference of the coefficients of thermal expansion. If the product of $\Delta\alpha$ and the temperature difference is large, the thermal stress can exceed the yield point of the metal layer in the system and cause plastic flow.

The second category for thin-film stresses is the intrinsic stress generated during the deposition of the film on the substrate. Factors such as oxidation and incorporation of impurities may affect the intrinsic residual stress. It has been shown that at the nano-microscale the intrinsic residual stress dominates the thermal stress, contrary to the macroscopic situation [211].

Nanometer-sized or nano-structured material volumes show some remarkable differences in their deformation behaviour compared to their larger and more coarse-grained counterparts. This is due to the obstacles to dislocation motion presented by the grain boundaries, and the proximity of the free surface and the interface. Particularly, stratified samples have attracted much attention, since the individual layer dimensions can be carefully controlled down to the nano-scale, leading to interesting combinations of electronic, magnetic, optical and mechanical properties for novel applications. The tungsten-copper (W-Cu) multi-layer composites considered in the present study are such nano-coatings. They are promising materials used mainly in micro-optics and micro-electro-mechanical systems (MEMS/MOEMS), offering flexibility and cost-effective production. The combination of their electrical properties and strength make them effective for high voltage contact applications [212, 213]. Furthermore, the high thermal conductivity of copper together with the low thermal expansion coefficient of tungsten renders W-Cu composites useful for thermal management applications, such as heat sinks for high density integrated circuits [214, 215]. Finally, they show remarkable mechanical

properties resulting from the compromise between the high strength of W and the ductility of Cu, along with size effects due to their nanoscopic grain size and stratified structure [216, 217]. W–Cu composites have attracted a lot of interest as coatings for use under harsh service conditions [218, 219].

The evaluation of residual stresses across the scales, from macroscopic to micro and nano-scale is of great importance for these systems, in the context of device design and optimization. One of the most versatile and powerful methods of stress evaluation is X-ray diffraction [14, 220] of microfocus X-ray beams that allow achieving sub-micron spatial resolution [221]. A number of destructive and semi-destructive analysis methods have also been developed based on the material removal and the measurement of the associated strain relief [222] in mechanical and materials engineering applications. The introduction of new traction-free surfaces causes measurable relief of the elastic strain that can be measured and related to the pre-existing stress magnitude by numerical deformation modelling [55, 223].

Recently, an approach to residual stress evaluation at the micro-scale has been proposed based on strain relief due to sample milling [55]. Detailed consideration of the stress state modification induced by cutting allowed it to be argued that an initially stressed small material volume that is created by careful, minimally disturbing machining, should become completely relieved of strain, even if it remains attached to the substrate on one side. The above hypothesis was put at the basis of an experimental procedure that made use of a combination of tools: FIB for incremental milling of “trenches”, to create new free surfaces that cause stress relief, as well as for patterning the surface to provide contrast; SEM for high resolution digital imaging of the sample surface before cutting, and at each stage of incremental milling; DIC for interpreting the sequence of images collected in this way in terms of strain changes; and FE method that allows the interpretation of observed incremental strain changes in terms of the residual stress in the coating [55], including depth profiling. FE analysis has also been used to demonstrate that for equi-axed islands (circles or squares), complete strain relief is achieved at the milling depth that is approximately equal to the “island” diameter.

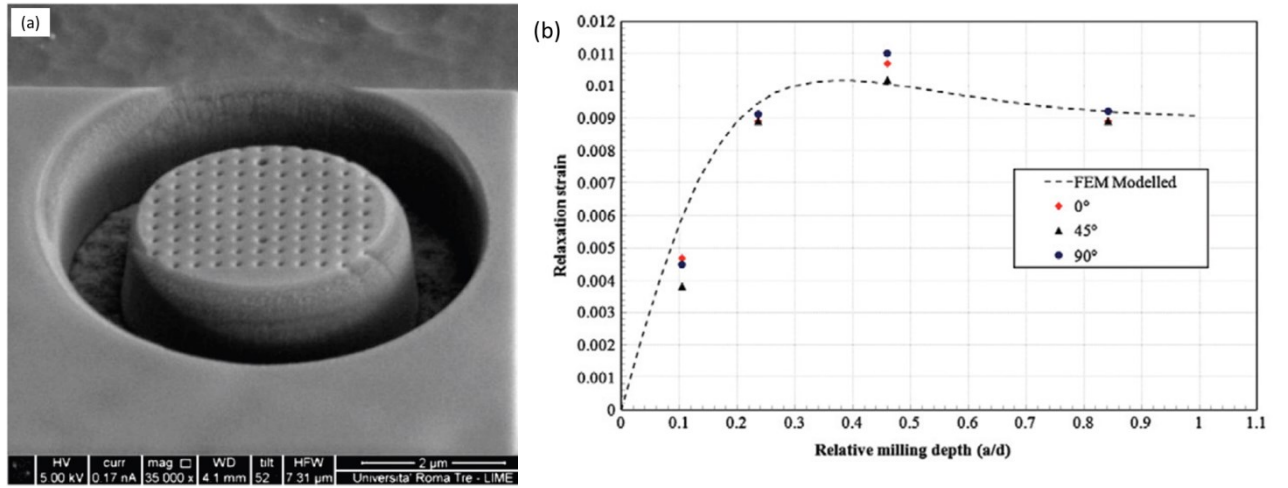


Figure 5.11: (a) Illustration of FIB ring drilling geometry with Pt sputtered surface and regular dot pattern (b) Plots of FE simulation results (dashed line) with FIB ring drilling DIC measurement (markers) [55].

It was established (by theoretical analysis and numerical simulation) that the relationship between the total strain relief $\Delta\epsilon^*$ and the residual stress σ^* is given by:

$$\sigma^* = -\frac{E\Delta\epsilon^*}{(1-\nu)} \quad (5.2)$$

A dual-beam FIB-SEM instrument allows using FIB for surface patterning and annular trench milling, and SEM for imaging and strain evaluation.

The sample under examination was 3.8 μm TiN CAE-PVD coating on a thick WC/Co substrate. Figure 5.11 (a) illustrates the FIB ring drilling configuration used in [55], consisting of the annular trench and the circular central “island”. A thin layer of Pt was deposited prior to FIB drilling to enhance contrast. A regular array of shallow dots was milled to assist the strain determination by DIC. The ring trench was cut incrementally to give through-depth resolution. A FE simulation approach was used to simulate residual deformation states during incremental ring drilling. Figure 5.11 (b) illustrates the FE simulation result (continuous curve) plotted alongside the FIB drilling-SEM imaging-DIC analysis experimental results (markers) for the strain relaxation against the relative milling depth, i.e. the trench depth h normalized with respect to the “island” diameter, d . For drilling depths exceeding the island diameter, the strain relief value approaches a constant, as the residual stress in the ‘island’ becomes fully released [55]. The residual stress can then be easily computed from the complete elastic strain relief.

The purpose of the present study was to show explicitly that the island(s) created in the surface coating by FIB-milling remain free of strain even in the presence of externally imposed substrate deformation. This furnishes conclusive proof of the assumption that underlies the FIB ring-core method [55], namely, that milling leads to strain relief in the island independently of the substrate stress state. In the context of X-ray stress analysis this is also particularly useful as an aid to determining the sign and thus the nature of the residual stresses in the coating (tensile-compressive). Using the d_o (the strain-relieved lattice parameter), the residual (or “live”) strain is calculated from the measured lattice parameter in the coating d using Eq. (1.3).

5.2.1. Experimental Setup – DIFFABS, Soleil Synchrotron

For the purposes of X-ray lattice strain measurements, a laminate composite formed by polycrystalline W and Cu thin films was used. Thirty eight periods of 3nm W layers and 1nm Cu layers were deposited by ion beam sputtering in a NORDIKO 3000 device at room temperature, onto a polyimide substrate (Kapton[®]), forming a 0.150 μm thick coating. In this study, the polyimide substrate was 127.5 μm thick and was cleaned with ethanol before deposition. Dogbone samples were then cut to the dimensions shown in Figure 5.12.

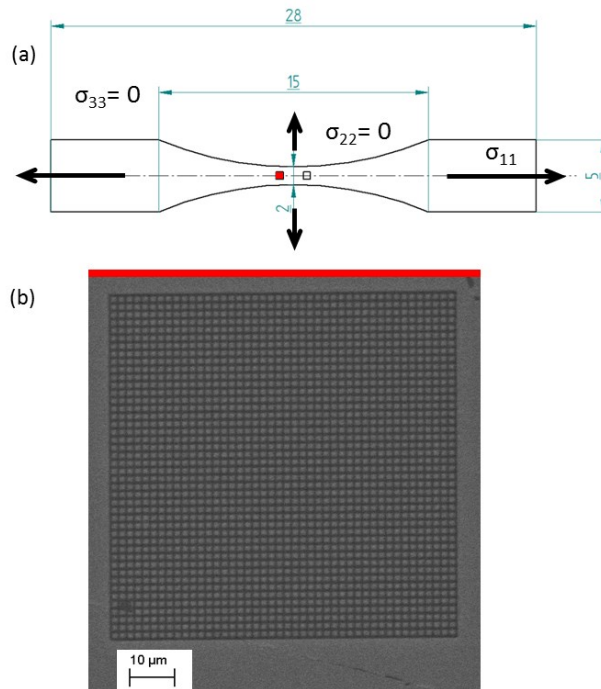


Figure 5.12: (a) Uniaxial dogbone sample dimensions (in mm), and (b) FIB “island” grid pattern milled on the nanocoating surface region highlighted in red.

The dogbone samples were then placed in a single beam FEI FIB 200 system, which was operated at a voltage of 30kV and with a milling current of 300pA. The key requirement of the investigation was to create an “on-board” strain-free patterned region to be evaluated at different load increments. The challenge was to be able to mill “patches” consisting of an array of “islands”, ensuring minimum exposure under the beam to avoid the damage and destruction of the nano-coating. An AutoFIB script was used initially to produce 45 horizontal trenches, each 75 μm long, with a 1.7 μm step in the selected region. Then a second script was used to create line trenches of the same dimensions, running vertical and perpendicular to the first set. Milling depth was set to 0.5 μm , but the process was interrupted for all the samples once the substrate was exposed, identified by the appearance of a darker contrast when imaging. An area 75 $\times$ 75 μm^2 was milled to ensure the provision of a stress-relieved region that would contain fully the X-ray beam spot used in the X-ray diffraction experiment (<50 $\times$ 50 μm^2).

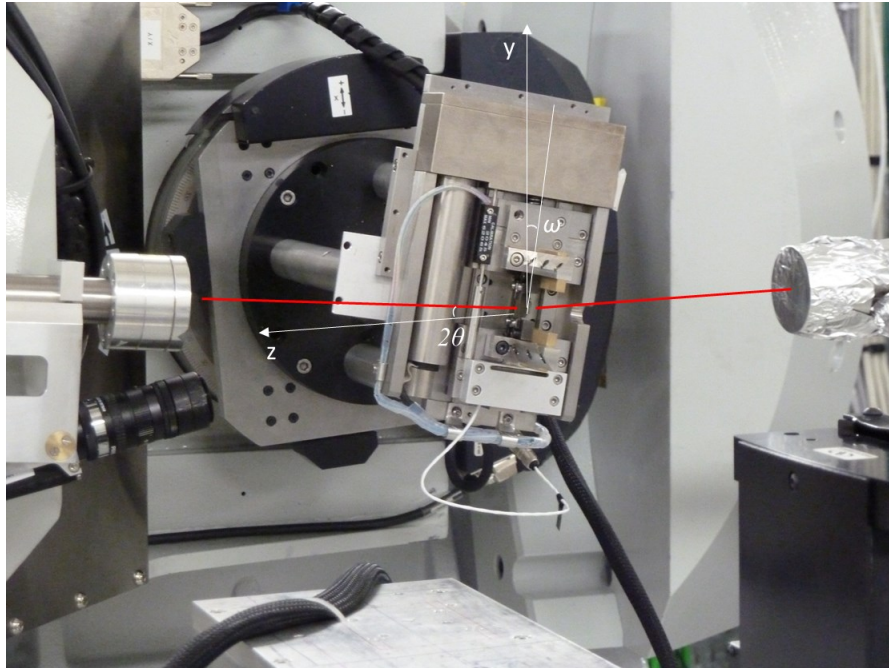


Figure 5.13: Experimental Setup on DIFFABS beamline Synchrotron Soleil, France. The sample was mounted on the uniaxial loading device and placed at the centre of the diffractometer. The incoming and diffracted beam paths are illustrated.

The island grid pattern is illustrated in Figure 5.12 (b). The size of each island on the grid was $\sim 0.8 \times 0.8 \mu\text{m}^2$. The value of the relative milling depth for the island-patterned region in this case was $\sim 0.19 \mu\text{m}$ (milling depth of $h = 0.15 \mu\text{m}$ equal to the coating thickness over the island diameter of $d = 0.80 \mu\text{m}$). Referring to Figure 5.11, we note that this corresponds to a "sweet

spot" on the strain relief plot, when the strain increment upon milling is equal to the value that would arise at saturation.

X-ray diffraction experiments were performed in transmission geometry illustrated in Figure 5.13. The XRD measurements for the present study were performed at the DIFFABS beamline of at synchrotron SOLEIL (Saint-Aubin, France). The X-ray beam used was monochromated to the energy of 17keV and focused to the spot size on the sample of $30 \times 30 \mu\text{m}^2$. A point detector was used for the 2θ scans. The angle ω was fixed at 11.7° for the uniaxial loading set-up. The $[110]$ family of lattice planes in W was analysed with the scattering angle $2\theta \sim 18.87^\circ$. The Bragg angle θ (found from peak fitting) was monitored during *in situ* deformation of the substrate, and the lattice spacing d_{110} computed.

A Deben[®] uniaxial loading rig was used, with custom-made sample supports, allowing access for the incident and diffracted beams, and with the sample surface parallel to the scattering vector. A 660N calibrated load cell was used to measure applied load, and a displacement transducer was used to deduce the macroscopic sample strain. Samples were monotonically stretched to failure at a total strain of 0.1. Larger number of increments were executed in the elastic region, and fewer at higher loads. This is illustrated in Figure 5.14, where the stress-strain curve extracted from the Deben[®] software is shown, together with the experimental measurement points.

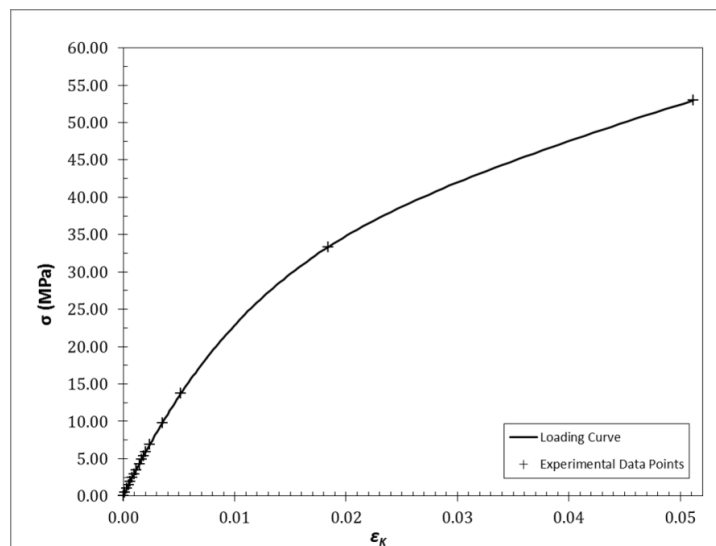


Figure 5.14: Loading stress-strain curve illustrating the loading increments and measurement points.

The location of the FIB-milled array on the sample surface was first found approximately using a microscope aligned to the centre of the diffractometer. At each loading increment, a positional X-ray beam scan across the sample surface was performed using a fixed 2θ position of the point detector. As illustrated on Figure 5.15, once the beam was located within the milled array, a significant reduction (50-60%) of the diffracted intensity at peak centre was observed. This reduction of intensity is mainly due to the reduced volume of material being illuminated by the beam (approximate calculation shows that the square grid pattern preserves $\sim 22\%$ of the original coating volume). Interestingly, our initial attempts to locate the milled arrays using transmission mode using an imaging detector were unsuccessful, due to the dominant contribution to sample absorption of the $127.5\mu\text{m}$ -thick polyimide substrate. The island pattern positions were identified using that procedure manually making a total of three scans at each increment. Subsequently, a θ - 2θ scan was executed for each value of applied load for the grid pattern and for the undisturbed region of original coating.

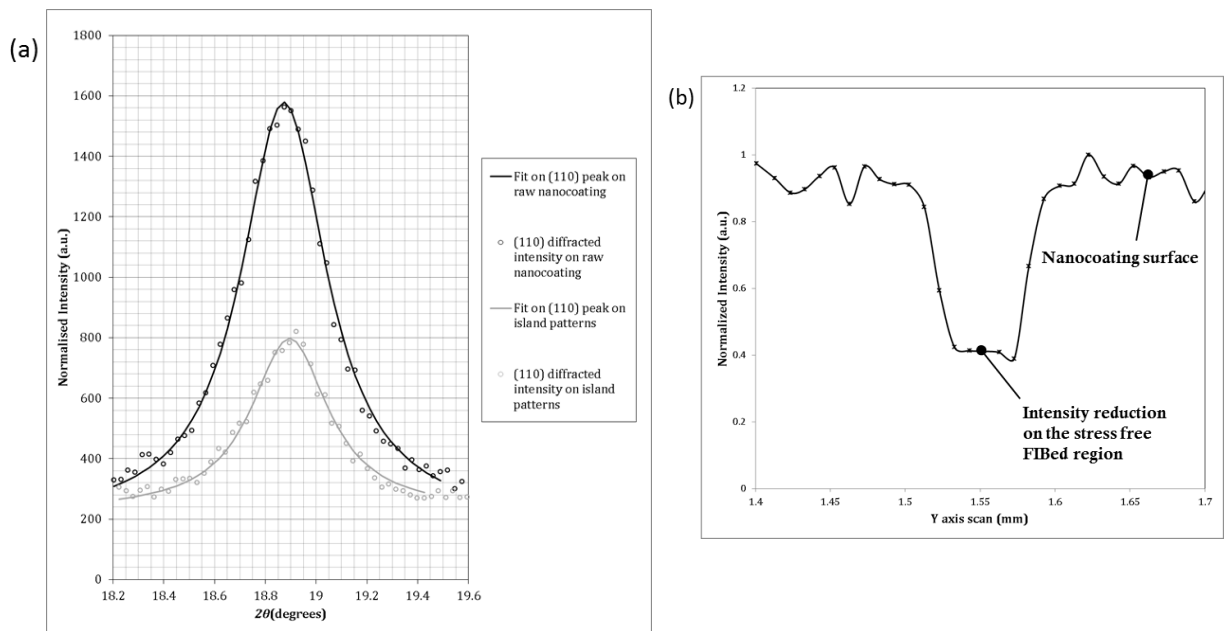


Figure 5.15: The determination of the location of the patterned arrays: (a) 2θ scans together with peak fitting results for the $[110]$ peak of W from the original coating surface (dark) and from the patterned area (grey), and (b) a line scan profile at fixed 2θ angle to determine the position of the “patch” showing significant reduction in the accumulated intensity.

5.2.2. Results and discussion

The results for the loading sequence applied to the W/Cu 3/1 coating are given in Table 5.3. The macroscopic response of the coated polyimide dogbone sample was recorded by the loading rig. Lattice strain was evaluated by diffraction from the $[110]$ family of planes in W from positions on the raw (unpatterned) coating surface, as well as in the region patterned by FIB milling. The strain error in all measurements was <200 microstrain (see Table 5.30).

The strain response of the rectangular grid patterned region is shown by the dashed line in Figure 5.16. The strain within this region remained approximately unchanged through all loading increments. The continuous curve in Figure 5.16 plots the raw coating lattice strain obtained by averaging the measurements from two distinct regions on the coating surface. The experimental data points are indicated by markers, and the continuous curve is obtained using splines. Clearly, the multilayer W-Cu structure has the capacity to incorporate “frozen-in” short range (nanoscale) residual stresses that exist over the length scales commensurate with the multi-layer period. However, at the microscopic scale probed in the experiment, the initial state of W-Cu within the multilayer can be considered to correspond to the equivalent d_o for these materials. The experimental findings allow the determination of both the strain-free (fully relieved) lattice spacing (d_o) from the patterned region, as well as the residual strain in the raw coating. This leads to the compressive strain figure of $(2806 \pm 100) \times 10^{-6}$ in the coating, as calculated from the reflection from W $[110]$ family of planes. The published values of the elastic constants of W are Young’s modulus $E = 411$ GPa, and Poisson’s ratio $\nu = 0.28$). According to Eq. (5.2), this is equivalent to the compressive residual stress of ~ 1.6 GPa that was induced during cooling from the coating deposition temperature. It is important to note that significant difference was observed between the residual strain values obtained at different locations in the coating, due to the high spatial resolution (beam spot size $30 \times 30 \mu\text{m}^2$) afforded by the setup. The origin of extrinsic residual stress in the coating is the thermal expansion coefficient mismatch between the polyimide substrate and the multi-layered W-Cu coating. The residual

stress results are broadly in agreement with those reported by Girault et al [224] for similar coatings on Si wafer.

The small deviations recorded in this experiment lie within the accuracy range of X-ray data interpretation – note from Figure 5.15 that the recorded peaks were relatively wide due to the broadening induced by the grain nanostructure. Thus, to the limit of precision of our measurement we can state that islands milled at the coating surface remain in the strain-free state throughout substrate deformation.

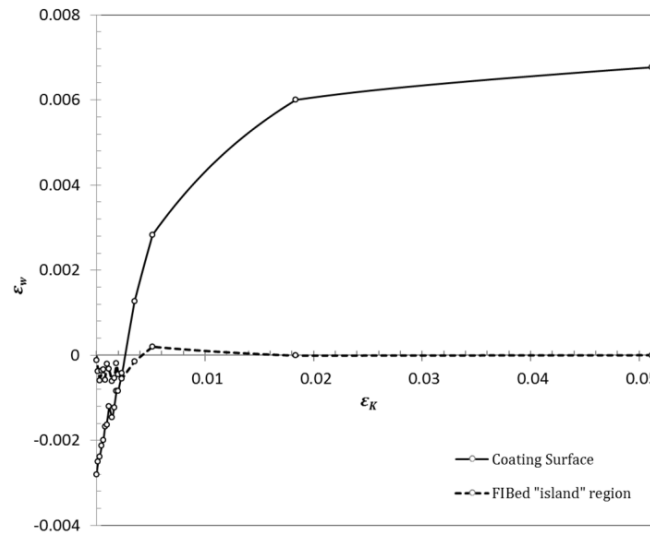


Figure 5.16: Strain-strain representation of the free surface of the coating response under loading versus equivalent response of the “patch” of FIBed islands.

Table 5.3: Loading increments, with equivalent stress, strain results and strain error for W/Cu 3/1 sample.

Loading Increment W/Cu 3-1	Stress (MPa)	Macroscopically measured strain (ϵ_{11}^K)	Microstrain on nanocoating surface ($\epsilon_{11}^{W,Raw}$)	$\langle 110 \rangle$ W peak fitting error on nanocoating surface given in microstrain	Microstrain on FIBed island region ($\epsilon_{11}^{W,Det}$)	$\langle 110 \rangle$ W peak fitting error on FIBed island region given in microstrain
1	0.00	0.000000	-2806.65	126.03	174.29	172.95
2	0.49	0.000157	-2506.00	89.24	-82.82	152.05
3	0.98	0.000316	-2380.80	94.47	-298.02	193.99
4	1.47	0.000476	-2132.73	89.25	-67.08	173.02
5	1.96	0.000639	-1993.79	89.25	-30.32	162.60
6	2.45	0.000803	-1671.79	73.48	-282.27	188.89
7	2.94	0.000968	-1632.99	84.01	100.95	194.08
8	3.43	0.001136	-1195.22	99.75	-14.57	188.97
9	4.31	0.001442	-1467.52	78.73	-313.76	220.41
10	4.90	0.001650	-1224.13	83.98	-240.29	152.22
11	5.39	0.001825	-836.33	99.76	111.45	204.75
12	5.88	0.002003	-839.07	99.74	-151.07	152.24
13	6.86	0.002364	-427.24	78.73	-245.54	183.80
14	9.80	0.003502	1260.71	68.26	148.22	189.39
15	13.73	0.005169	2829.99	84.05	500.24	179.10
16	33.33	0.018378	5999.89	99.78	290.05	195.47
17	52.94	0.051186	6766.46	73.53	300.55	222.18

5.2.3. Conclusions and remarks

The uniaxial deformation behaviour of a nanostructured W/Cu metallic film deposited on a polyimide substrate, using synchrotron XRD technique was investigated. A strain-relieved region ($\sigma_1 \approx \sigma_2 \approx 0$) in the pattern of rectangular grid of islands was milled on the sample surface using FIB, and its deformation was monitored at different loading increments, together with that of the original, untreated coating. The data analysis and representation are based on the strain-strain approach, i.e. considering the variation of the lattice strain measured by diffraction versus the total strain imposed on the substrate via the loading device. This allowed us to consider mechanical response of the thin film to applied loading. The experimental results provide clear demonstration that the island-patterned region remains under the conditions of constant strain independently of the deformation of the substrate. The method proposed in the present study is therefore particularly suitable for accurate determination of the unstrained lattice parameter in thin films and nanocoatings.

5.3. Nano-scale mapping of lattice strain and orientation inside carbon core SiC fibres

The strongest fibres available today are carbon-based, made from carbon nanotubes (CNTs) or reduced graphene oxide flakes (RGOFs). In practical use, CFs are typically incorporated into larger scale systems that require multi-scale characterisation. Much of the present understanding of the morphology and texture in carbon fibres derives from TEM investigations that reveal the lattice properties and arrangement at the nanoscale [7-9]. In practical use CFs are incorporated into larger scale systems, e.g. as cores of silicon carbide fibres for aerospace composite applications or woven into fabric reinforcement in high performance polymer matrix composites for motor sports, etc. The ability to characterise the hierarchical, multi-scale structure of carbon fibres that serve as the reinforcement basis in composite systems remains a challenge. Synchrotron X-ray beams offer an excellent combination of flux, brightness and energy for nanoscale imaging alignment and spatially resolved diffraction mapping [225].

However, the co-existence of crystalline and amorphous content in CFs makes nanoscale characterisation using a single technique insufficient [226].

CFs can be made via different routes that depend on the choice of precursor. PAN-based carbon fibres are made by pyrolytic reduction of polyacrylonitrile fibres. The resulting carbon fibres contain a mixture of amorphous and crystalline material, with the maximum crystalline volume fraction of 50% [227]. Alternatively, CFs can be produced by carbonization of mesophase pitch (MPP), a carbonaceous precursor with a highly oriented crystal structure [228]. Both PAN and MPP precursors are subjected to subsequent processing including spinning (melt-wet-dry), coagulation bath, drawing and curing in the as-spun state. Control over process parameters such as fibre elongation (draw-down rate) results in fibres with different morphologies (different void content and preferred orientation), with the degree of carbonization (crystalline content) reaching up to 80% (>2000°C) [229, 230].

The principal distinction between the nanostructure of PAN-based and pitch-based CFs is the degree of crystallinity and preferred orientation. PAN-based CFs with random crystal orientation and significant amorphous content display high strength and moderate modulus [9]. The highly aligned MPP-based fibres have the combination of high modulus and moderate strength. In the present study, focused X-ray beam mapping of the carbon monofilament core of SCS-6 silicon carbide fibres reveals a transition from the central untextured region of ~13.5µm diameter (with a structure similar to a PAN-based CF) to a more highly aligned and crystalline outer region of ~35µm diameter (with a structure similar to a MPP-based CF). Thus the carbon monofilament core combines the two morphologies.

5.3.1. Experimental Setup – B16, DLS

The combined synchrotron imaging and scattering experiment (Fig. 5.17) was conducted on the Test Beamline B16 at Diamond Light Source, Harwell Science and Innovation Campus, UK (MT-9318). The bending magnet delivers X-ray flux with smooth spectral distribution in the range between 8keV and 25keV. The beam can be passed through a double crystal

monochromator, or above it at the fixed offset of 25mm in the white beam mode. Focusing in both modes can be achieved using a Kirkpatrick-Baez (KB) mirror pair. The mirrors are operated at a nominal 3mrad incidence angle and consist of a pair of elliptical pre-shaped silicon substrates coated with 40nm thick Pt layer. Prior to collecting scattering data in the line scanning or mapping modes, the relative position of the focused beam is determined with respect to the main beam in the focal plane XY that is located 55mm downstream from the edge of the KB mirror chamber.

A set of 200 μ m diameter tungsten cross-wires is mounted on the XY translation stage. This arrangement allows precise determination of the beam position (x_{KB}, y_{KB}) with respect to the main beam, as well as its size (Fig. 5.17 (b)). In front of the cross-wires, there is a 150 μ m diameter guard pinhole, made in tungsten plate, that lets through the focused beam, whilst blocking the weak remains of main direct beam and single reflections of the mirrors. Both the cross-wires and the pinhole are placed on magnetic kinematic mounts that allow them to be repeatedly removed and repositioned to better than micron accuracy. The position of the cross-wires along the z axis collinear with the main beam is marked to micron precision by a theodolite. It is used to place the specimen in the correct z position (the focal plane).

The sample was first positioned approximately using lasers that indicated coarsely the position of the collimated beam. With the main beam passing outside the KB mirror chamber, the two sets of collimation slits were placed wide open to expose the full field of view of the “X-ray eye”. Sample stage tilting around the x -axis and rotation around the y -axis were used to achieve best possible fibre alignment parallel to the beam. By progressive reduction of the beamsize (from 3.2×3.2 mm down to $10 \times 10\mu$ m), the core carbon monofilament was brought into alignment with the centre of the main beam. The procedure is continued to improve the alignment down to 1 μ m and better by using the PCO camera detector for imaging.

The unstrained values of the c lattice parameter values used for strain evaluation were $c_0^i=6.9980$ Å for the inner carbon region, and $c_0^o=6.8353$ Å for the outer carbon region. The

reference value of the a lattice parameter in the fcc SiC was taken to be $a_0=4.3982\text{\AA}$. Interplanar spacing values corresponding to the diffraction peaks 002 and 111 were used for strain evaluation in the carbon monofilament and SiC coating, respectively.

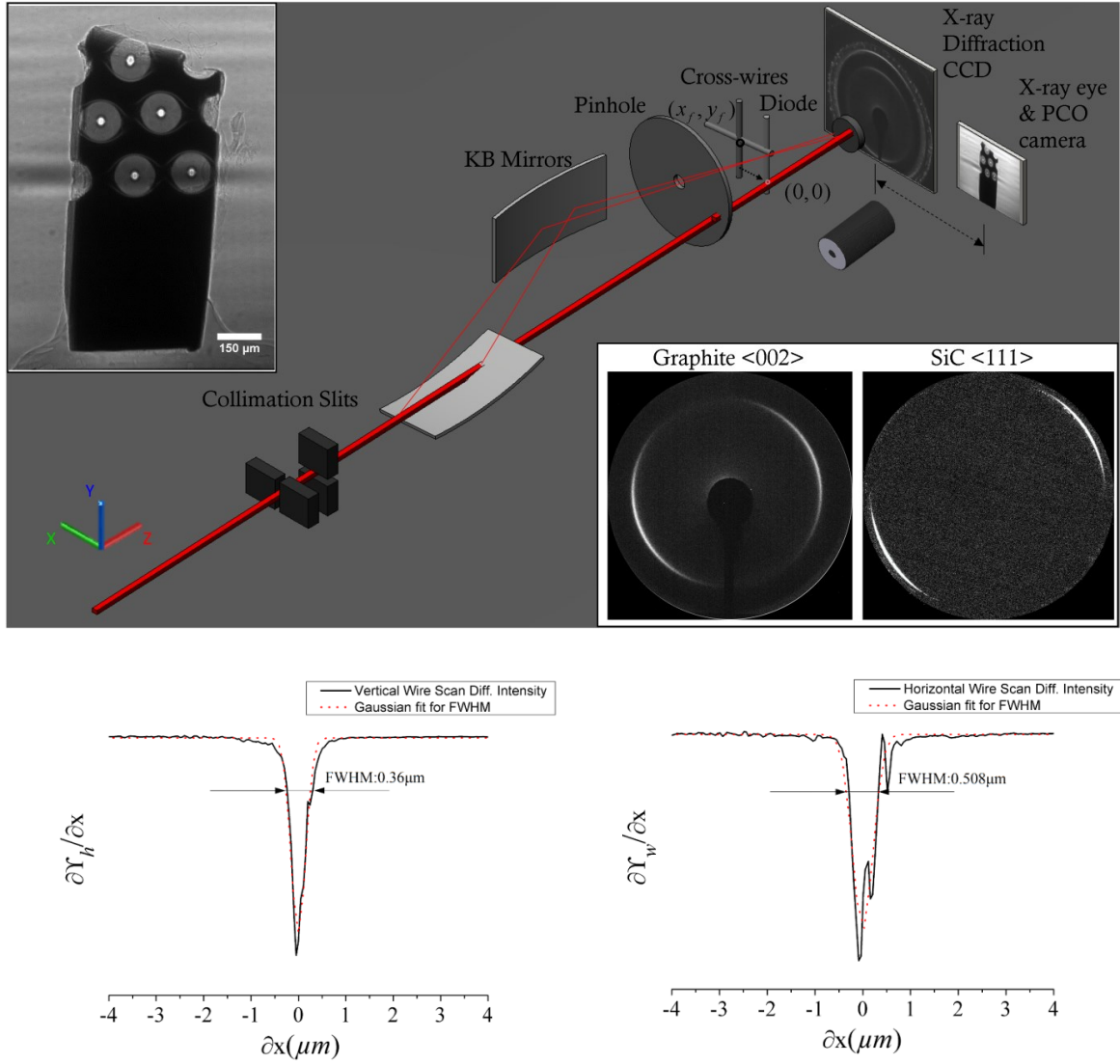


Figure 5.17: (a) Setup for simultaneous X-ray imaging and diffraction using synchrotron beam: beam collimation, focusing and alignment. Examples of diffraction patterns are shown as insets; (b) Beam size determination using the derivative of the wire scan [4].

The crystallite size was determined using the Scherrer formula [231],

$$L_c = \frac{(0.91)\lambda}{\beta \cos \theta} \quad (5.3)$$

where λ is the wavelength (nm), β is the Full Width at Half Maximum (FWHM), and θ is half of the scattering angle (radians).

5.3.2. Focused-Ion-Beam (FIB) – Digital-Image-Correlation (DIC) for microscopic strain analysis

Following careful polishing of the sample, down to 0.1 μm colloidal Silica, surface the micro-ring-core FIB and DIC technique was performed using a Tescan LYRA3 SEM-FIB. FIB milling of six $4 \times 4 \mu\text{m}^2$ islands (three in the SiC and three in the neighbouring carbon region) was then performed using a "ladder" like mask (Fig. 5.18 (a, b)). An island size of 4 μm was selected as a balance between strain spatial resolution and providing sufficient surface area to produce a reliable estimate of strain relief. A trench width of 1 μm and a milling current of 1 nA was chosen to reduce the impact of material re-deposition. In order to improve surface contrast and topology a beam voltage of 10 kV was used to image the sample and over 340 high resolution micrographs were captured. It was found that the z -contrast between the carbon in specimen surface and embedded colloidal silica nano-particles was sufficient to enable effective DIC of these micrographs. Several thousand markers were seeded onto these images and, following manual and automated outlier removal, a tracking script was used to determine the surface strain relief in each of the islands in the hoop and radial direction (as a function of image number). Least-squares fitting of known strain relief profiles [55] was then used to determine the average full strain relief in each island and to provide an estimate of confidence intervals on each measurement. Details of the combined FIB-DIC method are given in Chapter 2.

In order to provide calibration for the strain free lattice parameters in the CF the micro-ring core FIB and DIC technique was implemented at the interface between the central SiC and the neighbouring carbon region. This method relies on an incremental FIB milling process to fully stress relieve micro to nano-scale islands of material on the specimen surface [232]. During this procedure, high resolution micrographs of this region are collected which are later processed using DIC software to provide a direct measurement of the average complete radial and hoop strain relief experienced by the islands. Novel arrangement of these stress relieved regions in the form of a "ladder" has enabled, the production of spatially resolved maps of residual strain (Fig. 5.18 (c, d)).

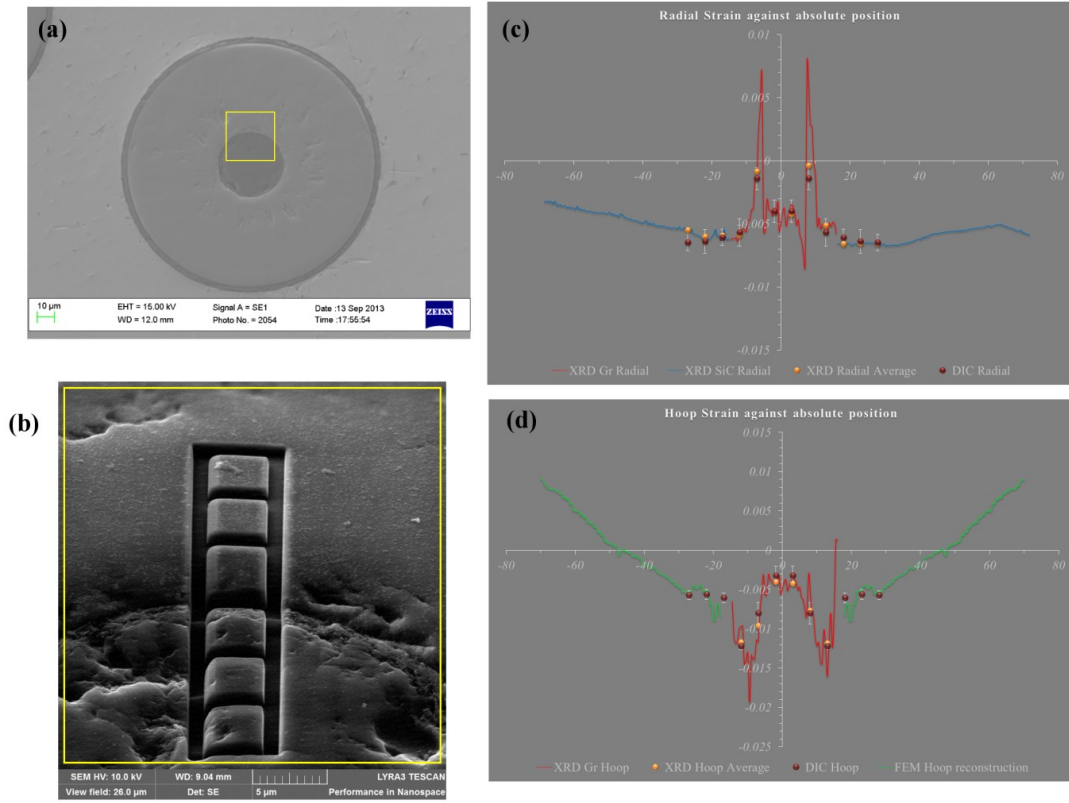


Figure 5.18: Focused Ion Beam milling - Digital Image Correlation and XRD measurement and FE modelling results: SEM image (a) indicating the locating of FIB analysis where "ladder" pattern (b) was milled at the interface of SiC and carbon in order to determine the radial and hoop residual strain components for calibration and validation of the X-ray diffraction results (c, d).

5.3.3. Inverse Eigenstrain Finite Element Analysis formulation

Consider an axisymmetric elastic fibre occupying the region $-x_r < x < x_r, -\infty < y < \infty$ and containing a distribution of radially varying eigenstrain $\varepsilon_{rr}^*(r) = \varepsilon_{\theta\theta}^*(r)$. The inverse eigenstrain modelling framework presented in [60] was used, with modifications explained here. An axisymmetric plain strain FE model was created in which the fibre consisted of three different regions with distinct properties. The material occupying the inner and outer carbon regions were assigned Young's modulus of 41GPa and Poisson's ratio ν of 0.4. The outer SiC part was assigned Young's modulus of 380GPa and Poisson's ratio ν of 0.2. Element type CAX8R was used for the simulation. The basis functions for inversion were obtained from the solution of the direct eigenstrain problem for each individual "step function" [233] corresponding to the peak eigenstrain values of $\varepsilon_{rr}^*(r) = \varepsilon_{\theta\theta}^*(r) = 10^{-3}$. The step functions were installed in the FE model as axisymmetric bands of local pseudo-thermal strain at radial position increments of $1\mu\text{m}$ between 0 and $70\mu\text{m}$.

The inverse eigenstrain reconstruction strategy used is thoroughly presented in Chapter 6. The unknown coefficients c_i of each basis function in the linear combination were sought by minimising the penalty function $\nabla_c J$ as formulated in Eq. 6.6.

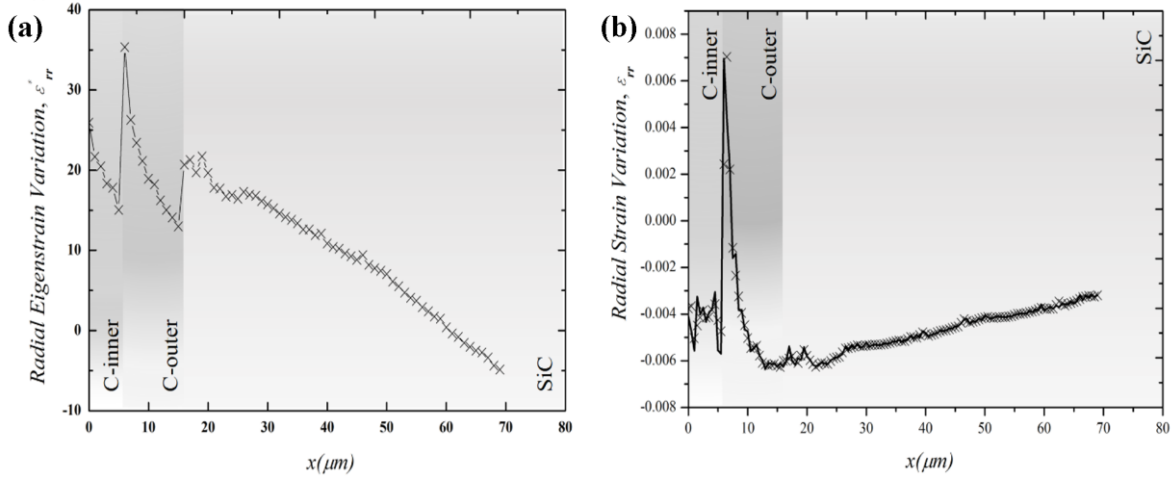


Figure 5.19: (a) The radial variation of the radial and hoop eigenstrain components, $\varepsilon_{rr}^* = \varepsilon_{\theta\theta}^*$, and (b) comparison between the reconstructed radial variation of the radial component of the residual elastic strain (continuous line) and the experimental data (markers).

The symmetric model inverse eigenstrain solution reveals the discontinuities amongst the different stages of manufacturing processes which have a severe impact on the residual strain distribution of the fibre. A maximum of $0.035 \varepsilon_{rr}^*(r)$, $r = 5 \mu\text{m}$ is observed at the transition between the non-textured CF core and the highly textured outer CF layer which are indicative of the different manufacturing routes and cause a maximum 0.7% of $\varepsilon_{rr}(r)$ residual strain within the carbon fibre outer layer. Furthermore, the residual elastic strain transition from the C-outer to the SiC fibre is continuous, however the eigenstrain distribution shows a secondary concentration of $\varepsilon_{rr}^*(r)$ at $r = 17.5 \mu\text{m}$ is observed associated to the deposition process of SiC at high temperatures which later results into SiC contraction at room temperature.

5.3.4. Results and discussion

The combination of the techniques described above not only provides result validation, but displays complementarity in terms of helping understand the morphology and stress state of the SiC fibre and the carbon monofilament in a joined-up, holistic fashion. Figure 5.20 illustrates the nanostructure of the carbon monofilament core in terms of the spatial distribution of the

crystalline particle size and shape, lattice orientation (Fig. 5.21) and the crystalline volume fraction [234].

Carbon fibres can be used in a wide variety of applications ranging from lithium ion batteries to polymer matrix composites. The complex nanostructure of the fibre (Fig. 5.20) suggests that deformation analysis must take into account the multi-scale load transfer mechanisms between the crystallites and the surrounding amorphous “matrix”, as well as the interaction between the unoriented central region and the surrounding highly textured material [235]. Furthermore, significant residual stresses are likely to be present within the system, having implications for its deformation response and the ultimate strength in tension and compression.

Measuring these residual stresses is not a trivial task, since they vary at sub-micron length scales. X-ray diffraction can be used to evaluate the lattice interplanar spacing. However, the conversion of the results into elastic strains and residual stresses remains a challenge due to the impossibility of using a strain-free powder reference, the lattice parameter changes due to chemistry variation, and the load-bearing contribution of the amorphous content. To overcome these challenges, the study reported in this chapter employed a combination of nano-focused synchrotron X-ray beams with FIB milling and DIC analysis for direct residual strain evaluation [55]. The FIB-DIC technique does not require strain-free reference samples [236] and copes equally well with crystalline and amorphous components.

Published reports on carbon fibres describe experimental setups whereby the probing X-ray beam is directed at the fibre perpendicularly to its axis [225, 237]. Due to the lack of gauge volume definition along the beam direction this produces diffraction patterns resulting from integrating the contributions from different radial positions within the fibre. High resolution mapping of the fibre structure and strain is impossible in this setup. Furthermore,

without the knowledge of d_0 variation the interpretation of the results is of limited value.

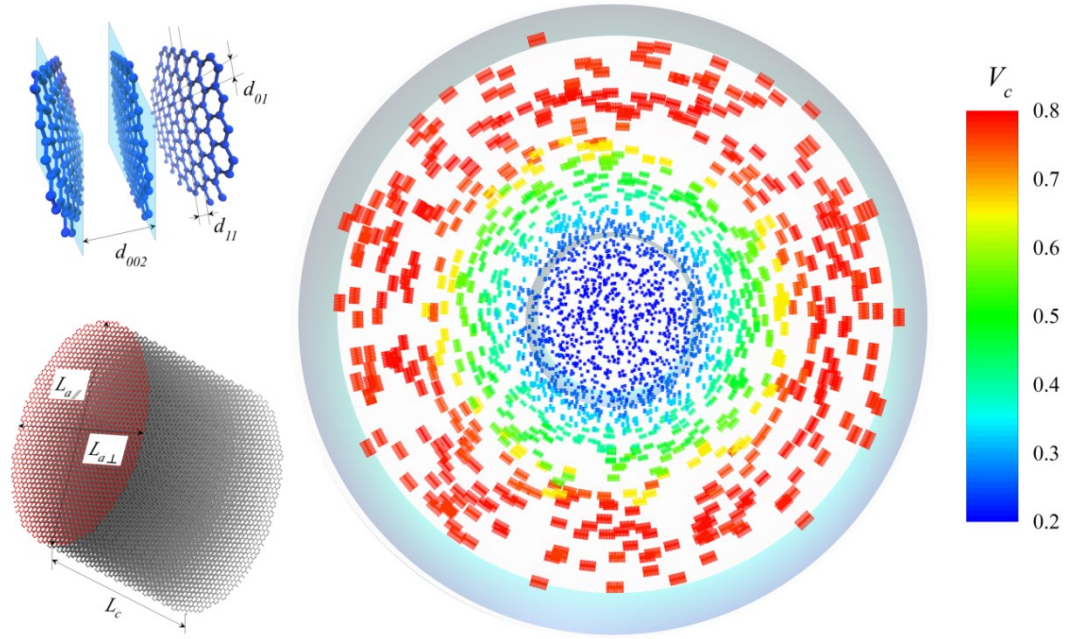


Figure 5.20: Nanostructure of the 35μm diameter carbon fibre: The crystallite content V_c is shown by the colour. The lattice spacing parameters d_{01} , d_{11} , d_{002} are indicated in the upper inset showing the structure of the smallest sized crystallite. The shapes in the main Figure illustrate the aspect ratio of the crystallites as viewed along the fibre axis. The crystallites have the dimensions L_c , $L_{a//}$, and $L_{a\perp}$, as shown in the lower inset. For illustration purposes, the size of the crystallites in the main Figure was exaggerated 1000x, and the number density shown is proportional to the local crystalline volume fraction divided by the average volume of individual crystallites.

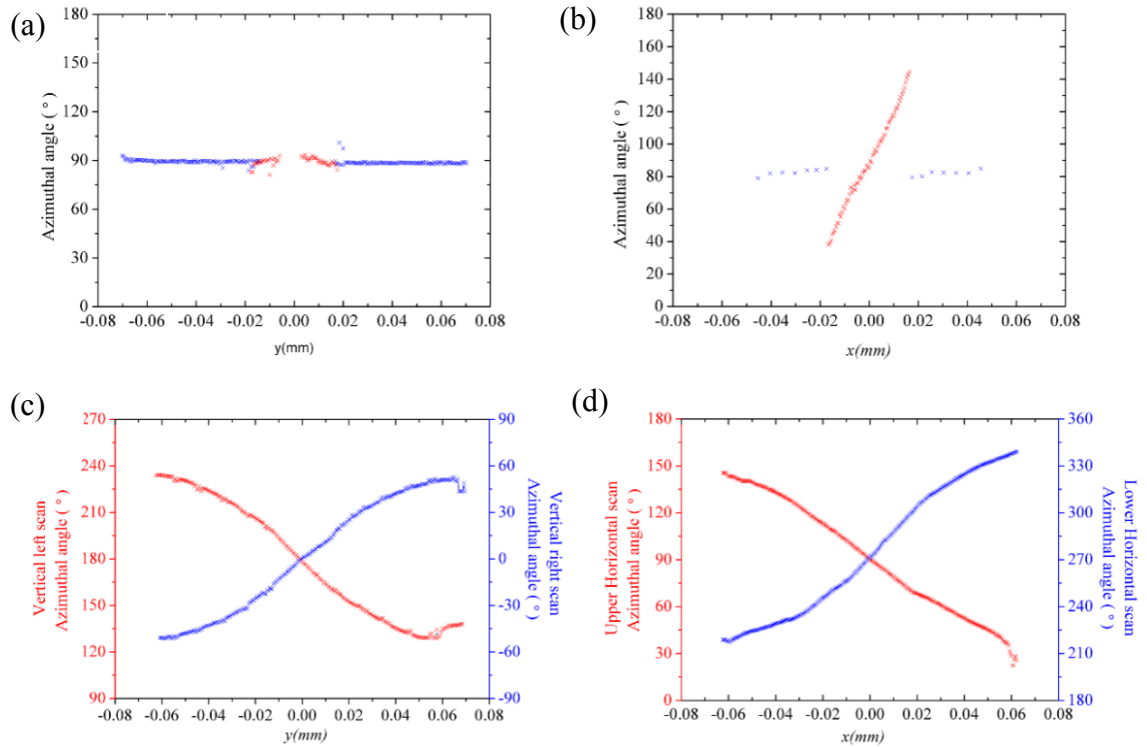


Figure 5.21: Line profiles of crystal preferred orientation in the carbon monofilament: (a) Vertical scan across the centre of the fibre with SiC shown in blue and CF in red; (b) horizontal scan 5μm away from the fibre centre, with SiC shown in blue and CF in red; (c) and (d) Vertical and horizontal scans on the SiC coating (scan positions are illustrated on Figure 5.24).

To overcome these difficulties we performed high resolution mapping of fibre cross-section, as illustrated in Figure 5.22 and 5.24. In order to achieve high resolution, careful alignment of the fibre axis with the incident X-ray beam was essential. Initial characterisation of the sample structure was carried out using X-ray tomography. It reveals the multi-layer fibre structure (Fig. 5.22 (a)) consisting of the C monofilament core, SiC fibre bulk, and outer C coating at the interface between SiC and titanium alloy matrix. Next the sample was placed on a high resolution, six degree-of-freedom tomography stage with the beam travelled along the carbon monofilament core. Using high resolution imaging detector and monitoring the integrated transmitted intensity, alignment to better than 100nm along the 300 μ m fibre extent was achieved between the beam direction and the fibre axis (see inset in Figure 5.17).

The SiC fibres with carbon monofilament cores embedded in the titanium alloy matrix are shown at different magnification in Figure 5.22 (a) that combines the results of SEM imaging with local tomographic rendering. It is important to note that the inner carbon region has similar electron density to the outer carbon layer in the monofilament, leading to very low contrast in electron and X-ray imaging modes alike (Fig. 5.23). However, X-ray diffraction provides clear discrimination that serves as the basis for the schematic illustration.

Following sample alignment the position on the sample of the X-ray beam focused by Kirkpatrick-Baez (KB) mirrors was determined to better than 500nm. The diffraction patterns were recorded and indexed for different regions on the sample (the inner and outer carbon regions, and SiC), and the integrated diffracted intensity scan was correlated with the sample radiographic image in order to determine the positions of the scanning lines. A grid was defined consisting of three vertical and three horizontal lines that were scanned at the step of 400nm vertically and 500nm horizontally. Different exposure times were carefully selected for different regions of the specimen to ensure optimal statistics (10s for the SiC, 40s for the carbon monofilament core). To obtain detailed mapping of the outer carbon layer and to avoid unnecessary data overlap, the middle horizontal line scan was offset by 5 μ m with respect to the fibre centre. This offset was chosen in view of the typical diameter of PAN-based carbon fibres,

in order to capture the detail of the preferred orientation and residual strain variation in the outer carbon region. Sampling positions, lattice parameter variation, crystalline volume fraction and the preferred orientation of the 002 plane normal are shown on Figure 5.22 (b).

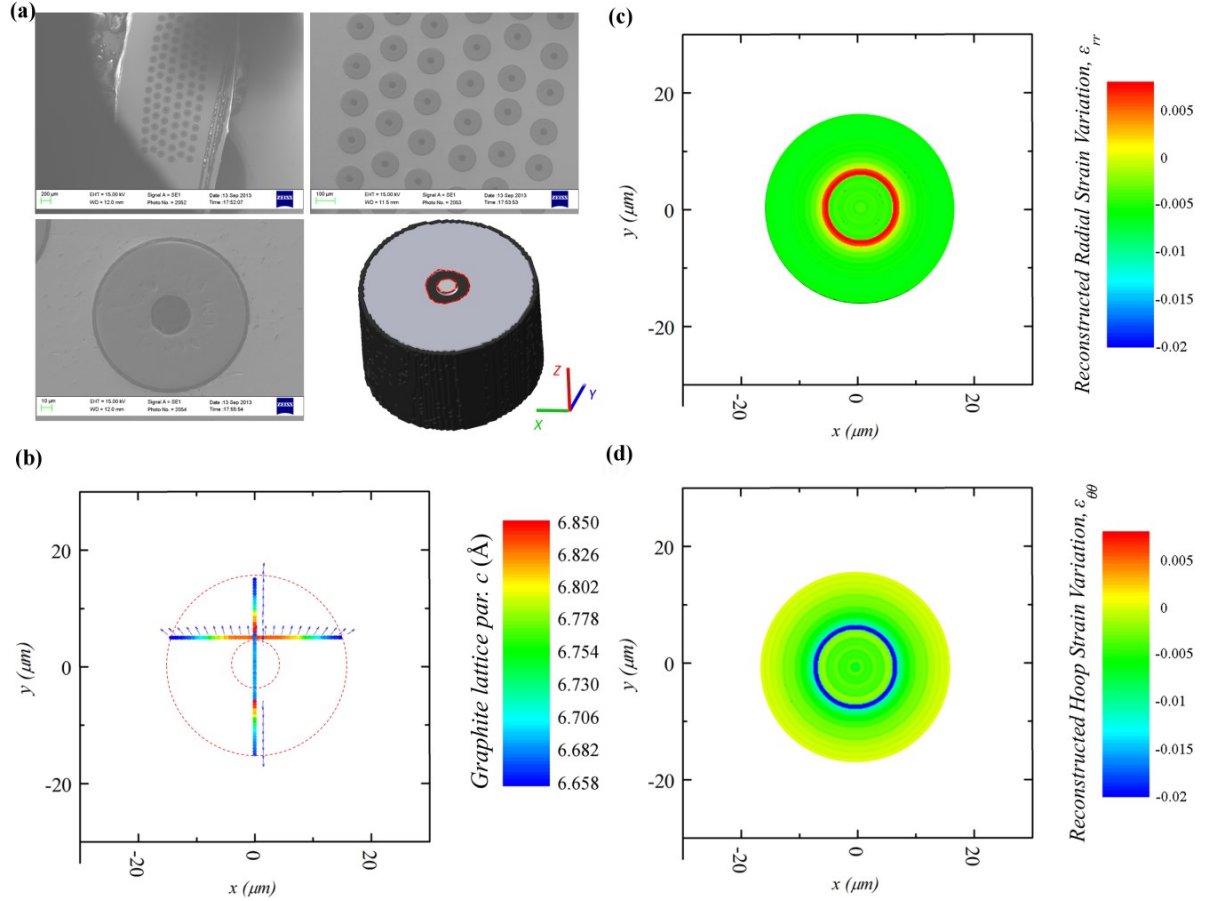


Figure 5.22: Residual elastic strain analysis in the carbon monofilament: SEM imaging and tomography reconstruction of the carbon core indicated in (a). The diffraction data collection points are indicated in (b). Figures (c) and (d) show eigenstrain reconstructions of the hoop $\theta\theta$ and radial rr elastic strains in the carbon fibre, respectively.

Residual stress evaluation requires converting the lattice parameter into elastic strain using the unstrained lattice parameter value, d_0 . The inner and outer regions in the carbon fibre are distinct and show a significant peak shift that exceeds the maximum strain-induced change that can be expected. This indicates that different strain-free lattice parameters (d_0^i and d_0^o) must be used for accurate strain calibration. Calibration was achieved by FIB nano-milling at the specimen surface to create a series (“ladder”) of strain-relieved islands arranged along the fibre radius [236]. DIC processing of the images collected before, during and after nano-milling revealed the residual elastic strain profiles at $4\mu\text{m}$ spatial resolution used to calibrate the X-ray diffraction measurements. The complete dataset for the residual elastic strain profiles along the

six horizontal and vertical scan lines was used as input into the inverse eigenstrain analysis procedure [60] described in the Methods section.

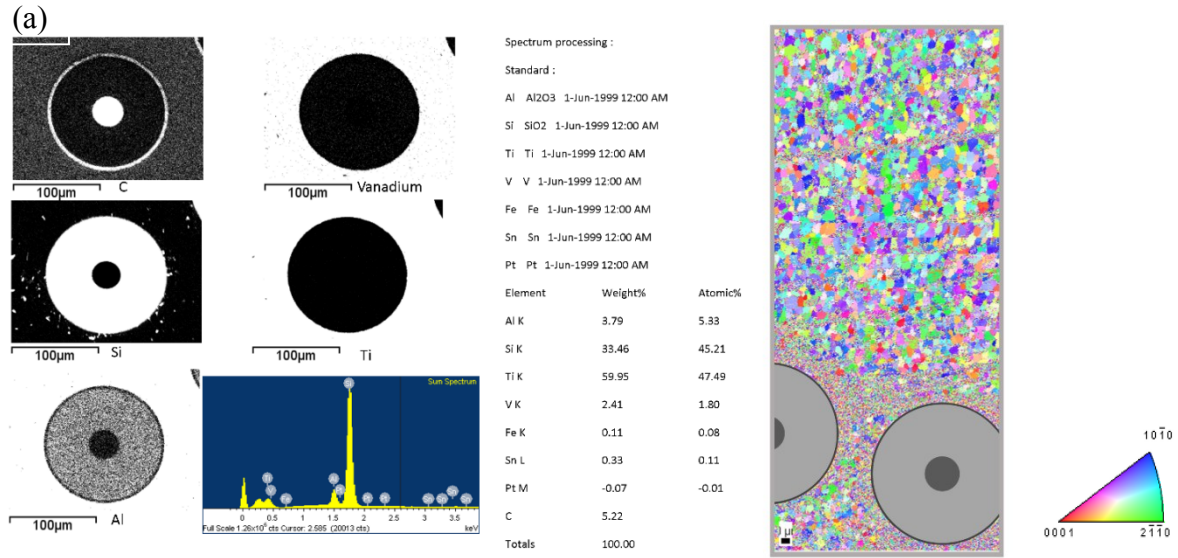


Figure 5.23: Multiple background characterisation data from SEM of SiC, carbon core reinforced Ti alloy composite for investigating material dispersion using (a) EDX detector and crystal size variation with (b) EBSD counting detector.

The reconstruction of the complete axisymmetric elastic strain state of the carbon fibre (radial ε_{rr} and hoop $\varepsilon_{\theta\theta}$ strain components) is shown in Figures 5.22 (c, d). The glassy core of the carbon monofilament finds itself under mildly compressive hydrostatic strain of $\sim 4 \times 10^{-3}$. At the transition from the glassy inner region to the more crystalline outer carbon layer, a sharp strain gradient is found close to the interface, with the compressive hoop strain in excess of 1% (equivalent to 0.75GPa compressive stress). The dual structure of the carbon monofilament results in a reduction in both the ultimate tensile strength ($UTS = 0.86\text{GPa}$) and Young's Modulus ($E = 41.5\text{GPa}$ [238]). This is due to the fact that the monofilament manufacturing route was optimised in terms of the surface properties suitable for subsequent addition of the outer SiC layer. The observed mild linear variation of the hoop and radial residual elastic strains is likely to be associated with the radial growth of the fibre during the carbonization process. Overall, the observations suggest that the source of significant compressive stress in the carbon monofilament is associated with the presence of the outer SiC layer.

The 135µm diameter SCS-6 SiC fibre was built around the carbon monofilament by CVD deposition. The growth of SiC grains proceeds in the radial direction, resulting in the highly

aligned morphology and texture (preferred orientation), with the [111] plane normals pointing outwards, as illustrated in the quiver plot in Figure 5.24 (b). Figure 5.24 (a) shows a tomographic rendering of the entire SCS-6 fibre (with the SiC layer shown in grey and carbon monofilament in black). It is worth noting the presence of a $\sim 3\mu\text{m}$ -thick outer layer that consists of the mixture of SiC and C that is deposited in order to improve the bonding to the alloy matrix during fibre incorporation into a composite structure [239, 240].

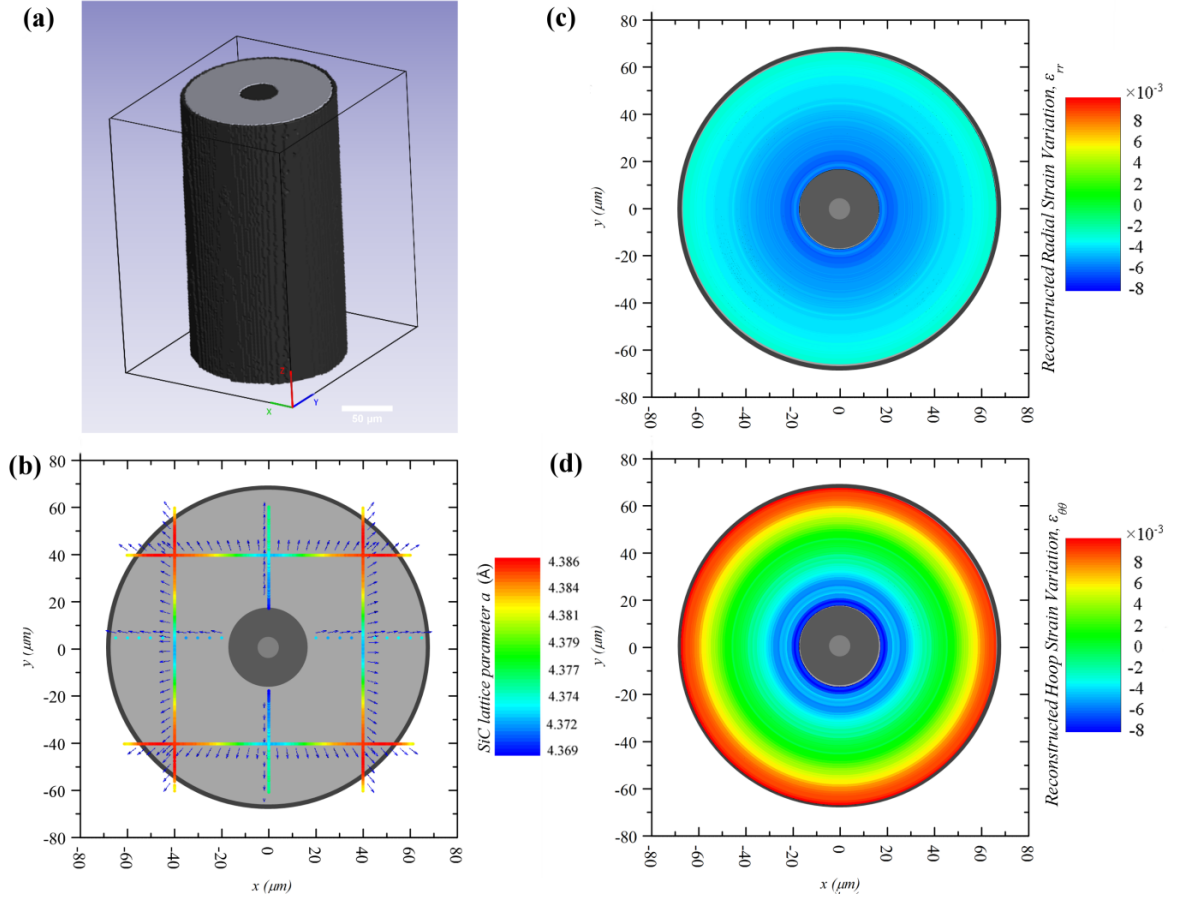


Figure 5.24: Residual elastic strain analysis in the SiC fibre: Tomographic reconstruction ($\sim 0.9\mu\text{m}$ resolution) showing carbon core (dark) and SiC outer layer (light grey). In (b) the inner light grey region represents the original PAN filament, with the outer layer representing carbon grown from pitch. Based on the diffraction data collected at locations shown in (b), figures (c) and (d) show maps of $(\theta\theta)$ and radial (rr) elastic strain components, respectively.

Figure 5.24 (b) reports the results of lattice parameter mapping in the SiC layer. The built-in thermal misfit strains (eigenstrains) arise in the course of the CVD process, with additional thermal mismatch stresses developing during cooling from the deposition temperature due to the difference in the linear coefficients of thermal expansion ($\alpha_c \sim 2 \times 10^{-6} \text{ K}^{-1}$, $\alpha_{SiC} \sim 4 \times 10^{-6} \text{ K}^{-1}$) [241]. The distribution of eigenstrains that acts as the source of residual stress was determined by the inverse formulation [232]. The complete reconstruction of the residual

strain distributions are shown in Figure 5.24 (c, d). It is apparent that the outer layers of SiC sustain significant tensile hoop strains that place the middle section of the fibre into compression (cf. the results for the carbon monofilament reported above). Residual elastic hoop strain (also calibrated in the SiC by FIB-DIC) increases approximately linearly towards the outer layers of the fibre, reaching the value of $\sim 5 \times 10^{-3}$ at the outer circumference of the SiC layer.

5.4. Conclusions and remarks

For the three individual case studies of “hybrid” experimental setups that have been developed in the study of three discrete scientific cases, the results show significant advances in the experimental methodology and acquired data quality. The modern measuring equipment, together with the flexibility of the test rigs allow for maximum efficiency of the beamtime usage and support the contingency of engineering projects which simulate *in operando* conditions within complex engineering assemblies or high spatial resolution probing nanoscopic distributions of strain and structural variation.

Conclusions can be deduced in the basis of separate scientific aims related to the individual cases showing the successful implementation and further combination of experimental and numerical techniques towards complete engineering characterisation of materials and components. The *in operando* feasibility study of internal combustion engine components is uses the most brilliant and high-penetration X-ray synchrotron beams and allows working on full scale components at **high spatial resolution (<20µm)** achieved through beam collimation. High flux affords very small counting times and thus time resolution, upon investigating fast processes, e.g. studying vibration effects at short exposure (<1µs), including signal accumulation by coupling system operation to data acquisition. Imaging with X-rays is a powerful tool that not only paves the way for applying diffraction techniques (alignment) but also gives quantitative submicron resolution results in terms of dimensions and tolerances of advanced technological components and processes.

In the case of thin films, X-ray micro-diffraction provides direct demonstration and validation of complete, substrate-independent strain relief of “islands”. The FIB milling technique can itself be used to generate fully stress relieved regions on the surface of engineering components or thin material sections (thin films, coatings, shot peened surfaces etc.) at submicron resolution levels. The measurement of those regions can provide the value of d_o that is crucial for residual strain and stress evaluation and not always available in the cases of bulk engineering samples, e.g. welds where d_o is crucial. Additionally, it was demonstrated that, when difference in absorption of FIB milled regions is not sufficient for locating those regions during X-ray imaging, 2-D X-ray diffraction mapping at a fixed 2θ angle allows the precise spatial determination of such regions on the sample. This is due to the minimum material removal from the surface of the sample which requires a systematic approach and combination of optical microscopy arrangements in the experimental hutch for approximate positioning of the sample and subsequent diffraction mapping.

The ability to obtain submicron size X-ray beams via optical focusing down to $<0.5\mu\text{m}$ and the use of high resolution X-ray imaging together with several wire scanning arrangements allowed a full 2-D investigation of residual strains and structure inside an SiC fibre-reinforced titanium alloy. The use of alternative FIB-DIC technique to cross correlate the experimental results increased the validity of the findings and allowed for safe conclusions to be drawn on the performance and mechanical characterization of such high-strength fibre reinforcements. Moreover, the FEM investigation using the eigenstrain concept allowed further understanding of the thermo-mechanical processing stages, as well as the fundamental consequences of the high thermal gradients applied during manufacturing.

Chapter 6

Neutron Diffraction Strain Tomography Time-of-Flight Technique

Complete knowledge of the residual stress distribution throughout the bulk of structural components requires point-wise evaluation of six independent components of the 3-D tensor of stresses or elastic strains. It has been demonstrated so far that a systematic non-destructive approach towards probing 3-D stress-strain states at sub-millimetre spatial resolution in the bulk of components, can be well pursued into the context of Neutron imaging and diffraction techniques.

A recently proposed novel approach utilises the well-defined change in the transmitted intensity of a neutron beam as a function of wavelength that is associated with the limiting condition for the satisfaction of Bragg's law as a function of scattering angle. As a logical next step, a tomographic acquisition strategy using multiple angle projections was employed for reconstructing 3-D bulk variation of the spatially resolved strain tensor components. This approach has an obvious correlation with the "rich" tomography techniques such as diffraction strain tomography (DST) discussed in Chapter 4. However, unlike in DST, neutron strain tomography, described here, was used to reconstruct the full in-plane strain tensor within an engineering sample.

Previously, the reconstruction of multi-component, tensorial quantities (such as strains) using neutrons has been reported for the case of axially symmetric strain variation [46], based on a single projection with the low spatial resolution defined by the 2.5mm pixel size. The novel approach of TOF neutron diffraction strain tomography (TOF-NDST) has been to the case of non-axisymmetric strain variation using multiple projections at ~40 times improved resolution.

6.1. Experimental setup for transmission Bragg-edge diffraction tomography, ENGIN-X, ISIS

The schematic representation of the neutron Bragg-edge transmission experimental setup is shown in Figure 6.1 (a). The measurements were collected on ENGIN-X stress spectrometer at ISIS pulsed neutron source, Oxford Harwell Campus, Didcot, UK. The beam size was set to 30mm (vertical) $\times$ 30mm (horizontal). A high resolution microchannel plates (MCP) neutron counting detector was used with the pixel size of $55 \times 55 \mu\text{m}^2$ [45]. The vertical cylindrical sample axis was perpendicular to the incident beam illumination (the y axis). The sample was mounted on a rotation stage aligned with the sample cylindrical axis and with the column of pixels near the middle of the detector field of view, in line with the common procedure for absorption tomography data collection. Duration of exposure required to obtain adequate signal statistics was in excess of 1h (72min), meaning that data collection could be undertaken only at 6 different sample rotation angles, namely, 0° , 22.5° , 45° , 90° , 135° , and 180° . Each exposure provided a 2-D map of 512×512 pixels, each containing a spectrum with 2032 TOF channels (Fig. 6.1 (a)). A flat field image was collected using the same exposure time to ensure consistent statistics for the normalisation of the collected spectra. For each rotation increment the data was re-binned (summed up) in the vertical direction to enhance the data quality, and to produce the equivalent 1-D “scans” of spectra as a function of the horizontal pixel position within the rows. This data reduction step was possible due to the plain strain nature of the deformation state within the specimen.

The specimen used, was a shrink-fitted “ring-and-plug” sample made out of two separate aluminium alloy pieces shown in Figure 2.7 (b), a hollow right circular cylinder referred to as the “ring”, and a solid elliptical cylinder (the “plug”). The ring had the inner and outer circle diameters of 7mm and 14mm, respectively. The ellipsoidal plug had the major axis of 7.03mm (highlighted with a bold black line in Figure 2.7 (b)), and the minor axis of 7.00mm. Prior to assembly of the ring-and-plug specimen, the outer hollow cylinder was heated above 300°C, causing the inner diameter to increase to 7.1mm and allowing the plug to be inserted freely. Upon subsequent cooling to room temperature, a permanently frozen-in residual stress state was created that was approximately uniform in the axial direction, and displayed non-axisymmetric (two-fold symmetric) variation within each plane perpendicular to the cylindrical axis. This corresponding residual elastic strain variation was the subject of the present study.

A MATLAB®-based routine was written to analyse the $[111]$ Bragg-edge in the collected transmission spectra using a three-parameter fitting function [43, 44]. By carrying out fitting for the range of horizontal positions of pixels on the detector, line profiles of the average lattice parameter were obtained, and converted to the average strain values. Due to the parallel configuration of the TOF experiment, the sample-to-detector distance calibration as described in Baimpas et al. [23] was not needed in this instance.

6.2. Direct problem: determination of residual elastic strain from given eigenstrain

Following the discussion on eigenstrain theory in Chapter 2. The general residual stress state in a solid is represented by the six components of the stress tensor (σ) at each point. Due to this inherent complexity it is practically impossible to recover each strain component by direct experimental onslaught, particularly given the spatial resolution required to probe a quantity that varies as a function of three spatial coordinates within the sample. Furthermore, it is important to note that the residual stress distribution must, by definition, be *self-equilibrating*. This requirement establishes a relationship between the gradients of different components of

the stress tensor ($\nabla \boldsymbol{\sigma} = 0$). The stresses must also satisfy *traction-free* boundary conditions ($\boldsymbol{\sigma} \cdot \mathbf{n} = \mathbf{0}$), where $\mathbf{n}$ denotes the surface normal.

The approach introduced by Mura [242] suggests that for a known non-uniform eigenstrain distribution $\varepsilon_{ij}^*(x')$, the elastic residual strain $e_{ij}(x)$ (and hence stress) in the body can be found using Eq. (2.27) incorporating C_{pqmn} which is the known tensor of elastic constants, and $G_{ipqj}(x-x')$ Green's function. Furthermore, total strain (ε) is given by the sum of the elastic (e) and the inelastic parts (ε^*), the latter denoting eigenstrain: $\varepsilon = \varepsilon^* + e$, or in the index notation, $\varepsilon_{ij} = \varepsilon_{ij}^* + e_{ij}$.

The fundamental principles that lay the foundation for building a realistic practical approach to the use of eigenstrain in residual stress modelling are summarised as follows.

(a) The compatibility requirement for overall strain is written as:

$$\nabla \times (\nabla \times \boldsymbol{\varepsilon}) = 0 \Rightarrow \frac{\partial^2 \varepsilon_{xx}}{\partial y^2} + \frac{\partial^2 \varepsilon_{yy}}{\partial x^2} - \frac{\partial^2 \varepsilon_{xy}}{\partial y \partial x} = 0 \quad (6.1)$$

(b) The presence of non-trivial eigenstrains acts as a source of incompatibility. Saint-Venant compatibility equations can be re-written by separating the elastic and inelastic strains as follows:

$$\frac{\partial^2 e_{xx}}{\partial y^2} + \frac{\partial^2 e_{yy}}{\partial x^2} - \frac{\partial^2 e_{xy}}{\partial y \partial x} = K \quad (6.2)$$

where:

$$K = \frac{\partial^2 \varepsilon_{xy}^*}{\partial y \partial x} - \frac{\partial^2 \varepsilon_{xx}^*}{\partial y^2} - \frac{\partial^2 \varepsilon_{yy}^*}{\partial x^2} \quad (6.2a)$$

In the above expression, the term K turns to zero in the case of uniform or linearly varying eigenstrain distributions, so that a body that contains such residual strain distributions is also free from residual elastic strain and residual stress.

(c) If the term K is non-zero, elastic strains arise that must satisfy the above equation, and simultaneously correspond (via Hooke's law) to the residual stress field that is self-equilibrating and traction-free:

$$\boldsymbol{\sigma} = \mathbf{C} : \mathbf{e}, \text{ or in index notation, } \sigma_{ij} = C_{ijkl} e_{kl} \quad (6.3)$$

(d) In fact, an arbitrary eigenstrain distribution can successfully be represented in the FE modelling environment by the introduction of non-uniform, anisotropic thermal expansion across the body. Indeed, it is clear that arbitrary forcing term K can be constructed in this way. This approach has been adopted in the modelling section below.

This sample chosen for the present study has a well-known non-axisymmetric eigenstrain distribution with well-defined principal axis caused by the misfit introduced during “ring-and-plug” assembly. The variation of eigenstrain can be assumed as a 1-D function of radial position. However the eigenstrain component ε_{11}^* and ε_{22}^* are different by the default geometry. Lastly, the almost fully axisymmetric geometry of the specimen provides an excellent basis for testing the capability of time-of-flight neutron diffraction strain tomography (TOF-DST) in application to non-axisymmetric cases.

In a FE interpretation the geometry imposed “misfits” are compensating for an ellipsoidal inner “plug”, inserted into a circular outer “ring”. This is readily accomplished by the application of uniform, anisotropic thermal expansion coefficient (CTE) in the inner “plug” region results in it deforming elastically to assume the actual ellipsoidal shape of the machined plug.

In this section, the implementation within the tomographic acquisition-reconstruction framework described below allowed to reconstruct 3-D variation of the multi-components strain tensor from a redundant set of lower order projection data. The inverse problem was formulated aimed at reconstructing the unknown distribution of eigenstrains that gave rise to the observed residual elastic strains measured in the average sense along the path of each transmitted beam.

6.3. Inverse eigenstrain analysis problem

As described in the previous section, an arbitrary eigenstrain distribution can be represented by pseudo-thermal strains incurring from radial variation of thermal expansion coefficients $\alpha_{ij}(x_r, y_r)$ under the application of a uniform temperature field ΔT [60, 243]. Hence, the relationship between eigenstrain and temperature is:

$$\varepsilon_{ij}^*(x_r, y_r) = \alpha_{ij}(x_r, y_r) \Delta T, \text{ where } i, j = 1, \dots, 3 \text{ and } x_r, y_r \in [0, R] \quad (6.4)$$

Now let the eigenstrain distribution $\varepsilon_{ij}^*(x_r, y_r)$ be the unknown tensor at the radial positions (x_r, y_r) and assume that a finite number of *residual elastic strain* component values $\varepsilon_{ij}(x_r, y_r)$. From the experiment let there be known a finite number of averaged across the sample residual strain component values $(t_q)_\theta$ at locations $(x_q, y_q)_\theta$, where $q=1 \dots, m$ refers to pixel variation at rotation increment θ . The equivalent numerical components $(T_q)_\theta$ derived by interpolation of the FEM nodes at the relative experimental defined positions (sample-projection on pixel). The result undergoes 2-D rotation (Fig. 6.1) as well as the equivalent strain transformation. The final $(T_q)_\theta$ can be deduced by averaging the ε'_{22} strain value across [22].

Therefore, in order to access the goodness of choice of $\varepsilon_{ij}^*(x, y)$ in terms of the matching to the experimental data, the following functional can be introduced:

$$J_\theta[\varepsilon_{i,j}^*(x, y)] = \sum_{q=1}^m w_q (T_q - t_q)^2 \Big|_\theta \quad (6.5)$$

where the use of weights w_q can influence the magnitude and significance of individual term contribution according to the data quality. The minimisation of the functional J_θ ensures that the best overall fit is obtained as the result of analysis [60]. The choice of sum-of-squares used to represent the mismatch between prediction and the experiment is non-negative, and has a unique minimum.

The problem of defining the eigenstrain distribution can now be reduced to identifying the function $\hat{\varepsilon}_{ij}^*(x, y)$ that delivers a minimum of J_θ . To do this the eigenstrain distribution $T_q = \varepsilon_{ij}^*(x, y) = \sum_{k=1}^N c_k s_k(x, y)$ can be discretised to a truncated series of base functions $s_k(x, y)$, $k=1 \dots N$ with unknown coefficients c_k . Consequently, the value of eigenstrain at the q -th measurement point at a specific rotation increment θ with coordinates (x_q, y_q) is given by the linear combination of the base function values $s_{kq}|_\theta = s_k(x_q, y_q)|_\theta$. The solution of sought as a combination of the c_k coefficients over the base functions. By substituting the truncated function series in J_θ and after differentiation:

$$\nabla_c J_\theta = 0 \Rightarrow \frac{\partial J_\theta}{\partial c_k} = 2w_q \left(\sum_{m=1}^N c_m \sum_{q=1}^m s_{mq} s_{kq} - \sum_{q=1}^m e_{kq} t_q \right) \Big|_\theta = 0 \quad (6.6)$$

By introducing the following matrix and vector notation: $\mathbf{E} = \{s_{mq}|_\theta\}$, $\mathbf{E}^T = \{s_{kq}|_\theta\}$, $\mathbf{t} = \{t_q|_\theta\}$ and

$\mathbf{c} = \{c_k\}$ Eq. (6.6) can be rewritten in matrix notation as:

$$\nabla_c J = 2(\mathbf{A}\mathbf{c} - \mathbf{b}) = 0, \text{ where } \mathbf{A} = \sum_{q=1}^m s_{mq} s_{kq} \Big|_\theta = \mathbf{E}\mathbf{E}^T \text{ and } \mathbf{b} = \sum_{j=1}^m s_{mq} t_q \Big|_\theta = \mathbf{E}\mathbf{t} \quad (6.6a)$$

where $\mathbf{t}$ and $\mathbf{E}$ are being filled in for the different rotation increments θ from 0° to 180° , according to what was mentioned previously:

$$\mathbf{t} = \begin{bmatrix} 0^\circ \begin{Bmatrix} t_1 \\ \vdots \\ t_{q=m} \end{Bmatrix} \\ 22.5^\circ \begin{Bmatrix} t_1 \\ \vdots \\ t_{q=m} \end{Bmatrix} \\ \vdots \\ 180^\circ \begin{Bmatrix} t_1 \\ \vdots \\ t_{q=m} \end{Bmatrix} \end{bmatrix} \quad \mathbf{E} = \begin{bmatrix} \alpha_{ij}(x_1, y_1) \begin{Bmatrix} \varepsilon'_{22, \varepsilon_{11}^*, 0^\circ} & \dots & \varepsilon'_{22, \varepsilon_{11}^*, 180^\circ} & 0 & \dots & 0 \\ \varepsilon'_{22, \varepsilon_{22}^*, 0^\circ} & \dots & \varepsilon'_{22, \varepsilon_{11}^*, 180^\circ} & 0 & \dots & 0 \end{Bmatrix} \\ \alpha_{ij}(x_2, y_2) \begin{Bmatrix} 0 & \dots & 0 & \varepsilon'_{22, \varepsilon_{11}^*, 0^\circ} & \dots & \varepsilon'_{22, \varepsilon_{11}^*, 180^\circ} \\ 0 & \dots & 0 & \varepsilon'_{22, \varepsilon_{22}^*, 0^\circ} & \dots & \varepsilon'_{22, \varepsilon_{22}^*, 180^\circ} \end{Bmatrix} \\ \vdots \\ \alpha_{ij}(x_r, y_r) \begin{Bmatrix} 0 & \dots & 0 & 0 & \dots & 0 & \varepsilon'_{22, \varepsilon_{11}^*, 0^\circ} & \dots & \varepsilon'_{22, \varepsilon_{11}^*, 180^\circ} \\ 0 & \dots & 0 & 0 & \dots & 0 & \varepsilon'_{22, \varepsilon_{22}^*, 0^\circ} & \dots & \varepsilon'_{22, \varepsilon_{22}^*, 180^\circ} \end{Bmatrix} \end{bmatrix}$$

And the solution of the inverse problem has thus been reduced to the solution of a linear system of equations:

$$\mathbf{A}\mathbf{c} = \mathbf{b} \quad (6.7)$$

in order to determine the unknown vector coefficients $\mathbf{c} = \{c_k\}$.

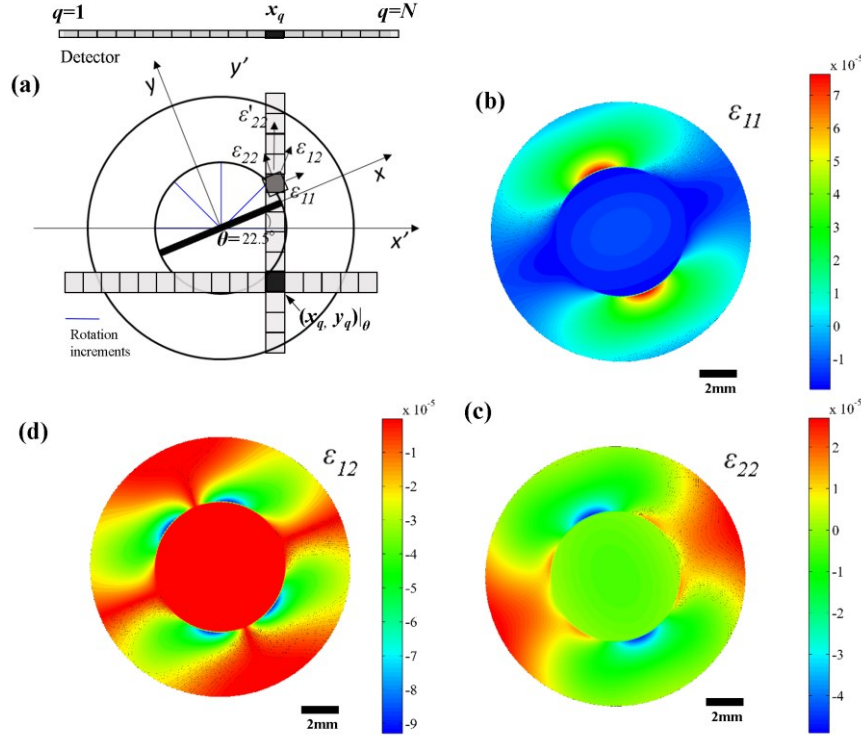


Figure 6.1: (a) An illustration of the analysis framework for the 22.5° rotation increment. The FEM calculated applied strain variation is subject to rotational strain transformation shown in (b) ϵ_{11} , (c) ϵ_{22} and (d) ϵ_{12} . Next, the strain component along the beam (ϵ'_{22}) is averaged over the beam path through the sample to simulate the experimentally measured average strain projection data deduced from the data collected by each detector pixel.

6.4. Modelling step-wise constant eigenstrain representation

The model hereinafter referred to as the “*shrink-fit*” model was created first. The “*shrink-fit*” solution effectively arises from the simulation of the process of putting together the elliptical plug-circular ring sample. The geometric misfit between the plug and ring results in embedding inelastic strains (eigenstrains) in an effectively axisymmetric geometry. This gives rise to the elastic fields that most closely represent those that are expected to be found in the sample studies. The “*shrink-fit*” model provides a point of reference for subsequent inverse eigenstrain reconstructions, assessment of accuracy, etc.

The FE plane strain model was implemented using ABAQUS™ software. The model represents a quarter of the overall “ring” and “plug” shape and the presence of the remaining

material is implied by symmetry. The mesh was created using the SWEEP meshing function to obtain a uniform grid of Integration Points (IPs), with two axes of symmetry prescribed that allowed the reduction of the model to one quadrant. The model illustrated in Figure 6.2 (a) used element type CPE8R (8-node biquadratic plane strain quadrilaterals with reduced integration) and 0.02 mm seeding step.

A fine distribution of step-wise constant radial variation basis functions that prescribe anisotropic in-plane eigenstrains, namely, the components $\varepsilon_{11}^*(x_r, y_r)$ and $\varepsilon_{22}^*(x_r, y_r)$ was used. The shear component of eigenstrain $\varepsilon_{12}^*(x_r, y_r)$ was considered to be zero. The set of eigenstrain basis functions used in the simulation consisted of step functions given by the following:

$$\varepsilon_{11}^*(x_r, y_r) = \begin{cases} C, & \text{for } R \leq r \leq R+h \\ 0, & \text{for } 0 < r < R \text{ and } R+h < r < R_{\max} \end{cases} \quad (6.8)$$

where $r = \sqrt{x_r^2 + y_r^2}$ is the radial position, and R is the lower value of the domain of the basis function. The radial discretisation step h could be set to values [0.1, 0.5, 1, 3.5] to consider the effect of the fineness of discretisation. The outer value of the eigenstrain domain was set to $R_{\max}=7$ mm, and the constant value C for the purposes of elastic strain calculation was set to 10^{-3} . The linearity of the FE model implementation meant that it was sufficient to perform the calculations only for the 0.1 mm discretisation step, since the solutions corresponding to the larger discretisation steps and other values of eigenstrain can be obtained by superposition and scaling. Figure 3(a) illustrates the prescription within the FE model via pseudo-thermal strain of the particular basis function that corresponds to the constant eigenstrain component $\varepsilon_{11}^*(x_r, y_r) = C = 10^{-3}$ imposed over the radial position range $R \leq r \leq R+h$, from $R = 3.5$ mm to $R+h = 3.6$ mm ($h = 0.1$). For the purposes of illustration the line profile of the function is shown as inset. The same procedure was followed for $\varepsilon_{22}^*(x_r, y_r)$ separately.

It is important to note that the choice of discretisation step is an important matter of judgement that remains at the disposal of the modeller. A model using coarse discretisation (i.e. constrained approach) may fail to capture the details of strain variation within the sample, but offers an inversion that is both fast and robust. Despite the additional computational effort involved in running the model using fine discretisation (i.e. non-constrained approach), higher resolution reconstruction may be obtained. An example of step-wise constant imposed radial variation base function $\varepsilon_{11}^*(x_r, y_r) = 0.001$ for $R \leq r \leq R + h$, where $R = 3.5\text{mm}$ and $h = 0.1\text{mm}$ is shown in Figure 6.2 (a). To facilitate comprehension a line profile of the function is shown as inset. The same procedure was followed for $\varepsilon_{22}^*(x_r, y_r)$ separately.

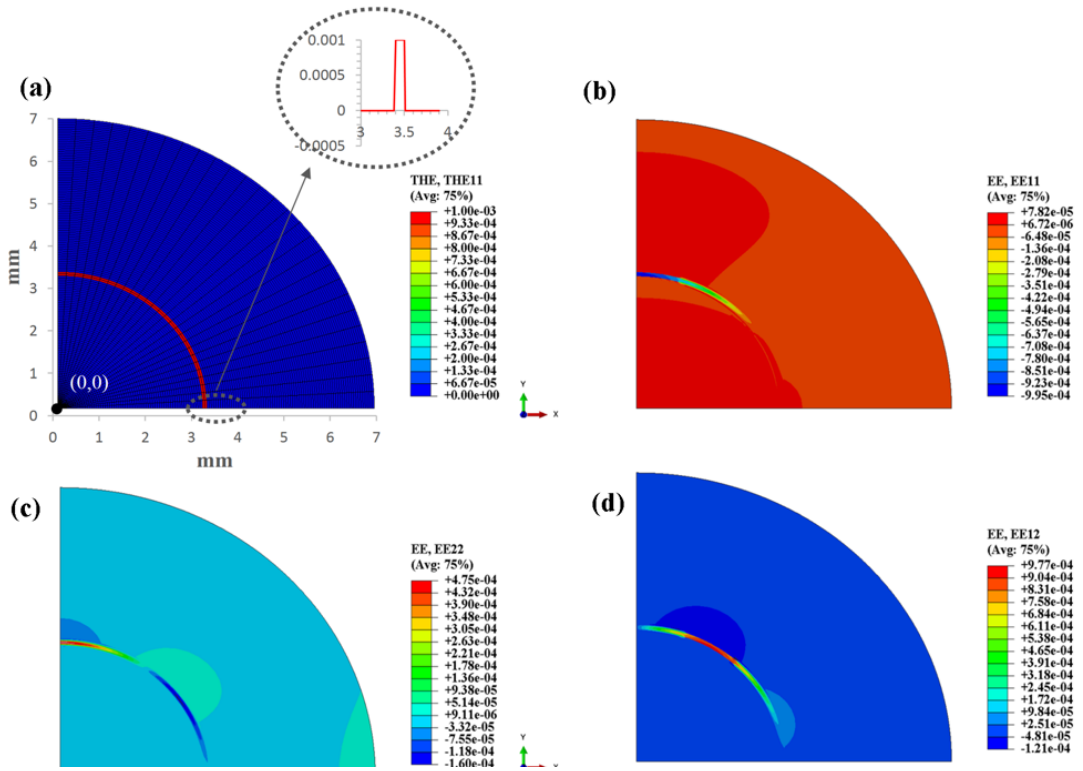


Figure 6.2: (a) Paradigm of step-wise constant imposed radial variation basefunction $\varepsilon_{11}^*(x_r, y_r) = 0.001$ for $R \leq r \leq R + h$, where $R = 3.5\text{mm}$ and $h = 0.1\text{mm}$. Resulting FE model calculated elastic in-plane strain components (b) $\varepsilon_{11}(x, y)$, (c) $\varepsilon_{22}(x, y)$ and (d) $\varepsilon_{12}(x, y)$.

The output of the overall elastic strain response to the eigentrain imposition, namely components $\varepsilon_{11}(x, y)$, $\varepsilon_{22}(x, y)$ and $\varepsilon_{12}(x, y)$ is illustrated in Figure 6.2 (b, d). The FE calculation output (average at node) was interpolated to match the experimental points (pixel

distribution) using `meshgrid` MATLAB[®] function and mirroring values on x and y axes to retrieve full sample shape around $(0, 0)$ which represents the sample axis of rotation, illustrated in Figure 6.2 (a). An image-based rotation approach, shown in Figure 6.1 (a), was used in parallel with strain component transformation calculation along the beam direction (ε'_{22}) (i.e. simulated strain component registered by the detector) by using ε_{11} , ε_{22} and ε_{12} at the respective experimental rotation increment θ , according to:

$$\varepsilon'_{22}(x, y) = \varepsilon_{11}(x, y) \sin^2 \theta + \varepsilon_{22}(x, y) \cos^2 \theta - 2\varepsilon_{12}(x, y) \sin \theta \cos \theta \quad (6.9)$$

where $x, y \in [0, R_{\max}]$.

6.5. Optimising reconstruction using the constrained model approach

In order to explore the abilities and limitations of the inverse eigenstrain method, forward and inverse simulations using “synthetic” experimental data were first performed to verify the robustness of the reconstruction approach. To this end, a *forward simulation* was performed (the “shrink-fit model”) to provide synthetic experimental data, for which the results were expressed in the form of angular projections at 0° , 22.5° , 45° , 90° , 135° and 180° . The data provided the input for inverse analysis and direct comparison between the solutions obtained following different strategies (Fig. 6.3 (a)). The residual elastic strain distribution used as the synthetic data was the solution for the strain caused by the insertion of the ellipsoidal inner “plug” with the major semi-axis of 3.515 mm and minor semi-axis of 3.5 mm that was inserted into the “ring” with the inner radius of 3.5mm radius (see details on Figure 2.7). Therefore, the misfit strain (eigenstrain) was specified as $\varepsilon^*_{11} = (3.515 - 3.5)/3.5 = 0.00428$. The following distribution of eigenstrain ε^*_{11} was introduced into the direct eigenstrain basis function model:

$$\varepsilon^*_{11}(x_r, y_r) = \begin{cases} 4.28 \times 10^{-3}, & \text{for } 0 < r \leq 3.5 \\ 0, & \text{for } 3.5 < r < 7 \end{cases} \quad (6.10)$$

and $\varepsilon^*_{22}(x_r, y_r) = 0, 0 < r < 7$.

As the final step of the forward calculation, the $\varepsilon'_{22}(x_r, y_r)$ component for the region $0 < r < 7\text{mm}$ was computed for the given experimental geometry and at all six rotation increments. The results are shown in Figure 4 (a) by the markers.

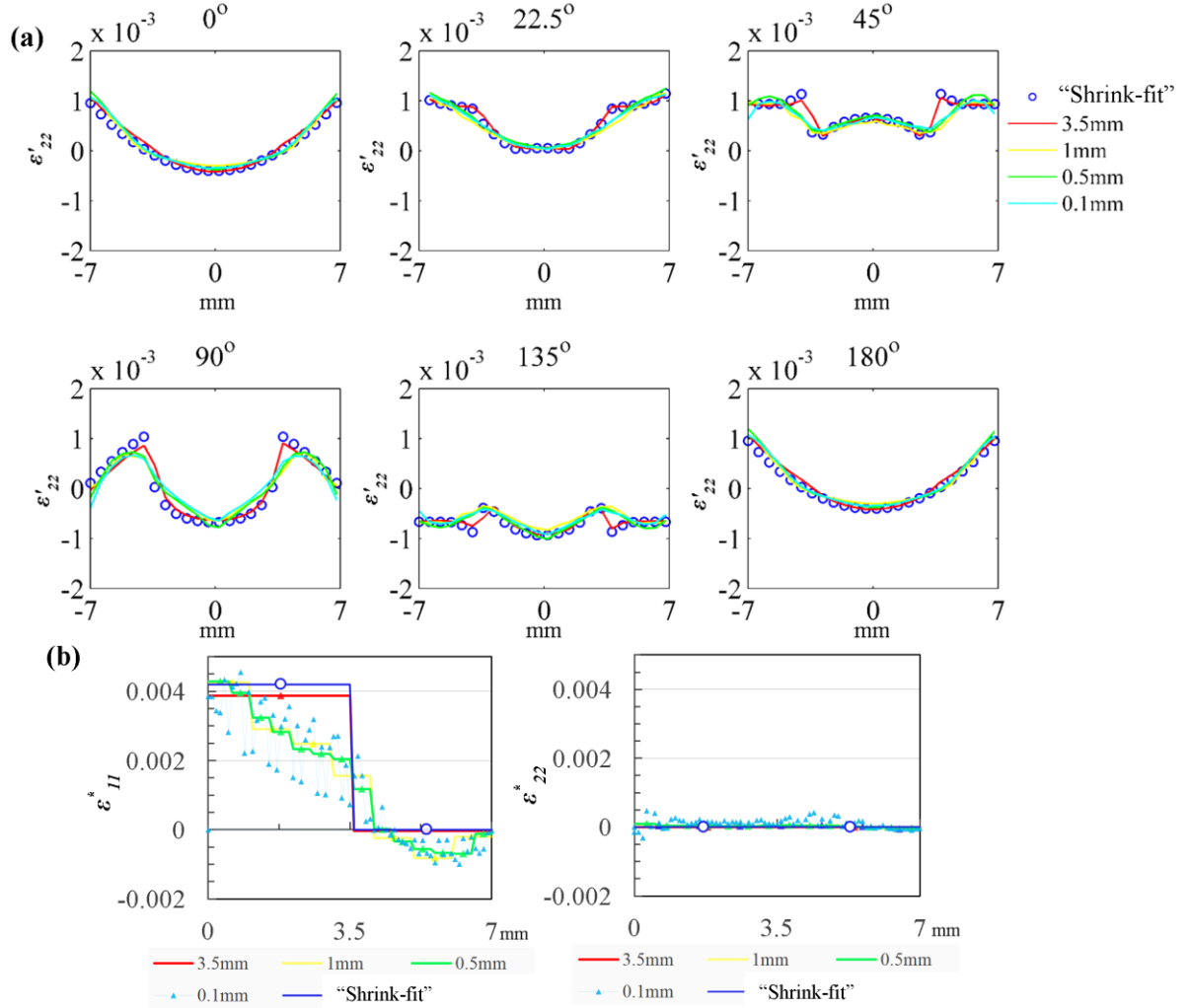


Figure 6.3: (a) Projections of the simulated experimental data and of the strain reconstructions the respectively for 3.5mm, 1mm, 0.5mm and 0.1mm discretisation segmentation, and (b) the reconstructed eigenstrain variation for the different discretisation approaches.

The inverse approach described in Section 6.3, was used to explore the reconstruction quality for the underlying eigenstrain variation and the resulting elastic strain distribution depending on the different discretisation strategies employed according to the number of discretisation steps (h). Figure 6.3 shows that for the specific condition the most stable and accurate solution emerges by imposing eigentrain at $h = 3.5\text{mm}$ increments, which results in $\varepsilon^*_{11}(x_r, y_r) = 3.95 \times 10^{-3}$ for $0 < r \leq 3.5$ and $\varepsilon^*_{11}(x_r, y_r) = -3.78 \times 10^{-4}$ for $3.5 < r < 7$. The ε^*_{22}

values were found to be $<0.3 \times 10^{-4}$ for $0 < r < 7$. The eigenstrain result is also graphically represented on Figure 6.3 (b).

Results obtained for other discretisation steps are presented in Figure 6.3 (a) and (b) where finer segmentation strategies lead to a progressive deterioration of calculated strains. The error of the calculated eigenstrains also increases to $\sim 13\%$ for 1mm, $\sim 14\%$ for 0.5mm and $\sim 17\%$ for 0.1mm increments. By introducing further unknowns (i.e. more eigenstrain values) into the system of equations for the given amount of projections, the steep variation at $r = 3.5\text{mm}$ fails to be represented accurately.

The above analysis serves the purpose of providing some indication that the use of any prior knowledge of the problem solution can be instrumental in constraining the solution to obtain accurate results even in cases when a limited number of projections is available. Using *a priori* knowledge of the magnitude and gradient of the inelastic strain variation, good quality reconstruction could be obtained even using synthetic data for a small number of projection angles. In the context of neutron beam experimentation, the potential to acquire projections at multiple rotation angles with a small step (i.e. $2\text{-}5^\circ$) is limited by the acquisition time (of the order of $\sim 1\text{h}$ required for each projection to reach good signal-to-noise ratio). Furthermore, experimental data inevitably contain noise that affects the quality of the reconstruction - significant level of noise may be present in X-ray and Neutron diffraction measurements. The origins are associated with the limited flux (particularly for neutrons), the scattering characteristics of the examined materials (e.g. graininess, or small sampling volume used), and the inherent noise present in the detector. These considerations must be taken into account in order to determine the correct exposure time in order to collect data of suitable quality. Data quality is particularly crucial for strain measurements, since small noise-induced Bragg-edge shifts correspond to significant strain variations.

The increased noise level in the tomographic acquisition measurements leads to reconstruction aberrations, especially when conventional back-projection techniques are used.

Since the present formulation uses an approach similar to Algebraic Reconstruction Techniques (ART), it deals efficiently both with the low number of angular projection data available, and also with the effects of noise in the data. In the ART version used here, the solution is not obtained from inverse Fourier analysis, but rather is extracted through the least squares approximation which “penalises” the mismatch in pixel values and converges to least disagreeable solution from the pre-defined functional space determined by the basis chosen.

To evaluate the robustness of convergence and stability of the inverse problem solution, the effect of adding noise to the simulated data was examined. Using 1mm eigenstrain discretisation step in the previously described formulation, random noise was added to the simulated experimental data. The noise level was set at 10%, 20% and 50% of the maximum absolute value of strain, and reconstruction performed. The resulting profiles of ε'_{22} is shown in Figure 6.4 (a). The results of this approach are shown in Figure 6.4 (b) and (c). They clearly demonstrate the robustness and stability of the method that appears to be resistant to noise. Even for the 50% added noise, the resulting reconstructions of eigenstrain components ε^*_{11} and ε^*_{22} remain minimally affected.

6.6. Results and discussion

In this section the analysis of the collected spectra is presented and the results of the applied TOF-NDST method are discussed. In Figure 6.5 a direct comparison between the inverse eigenstrain FE simulation and the collected experimental data at all rotation increments is illustrated. In order to achieve average error of strain values in the order 10^{-5} , careful assessment of the fitting results resulted in binning the data across by 10x (horizontal pixel resolution) reducing the spatial resolution of the experimental measurements to 0.55mm. The experimental data interpretation and fitting was carried using MATLAB[®] based algorithm using least squares refinement method.

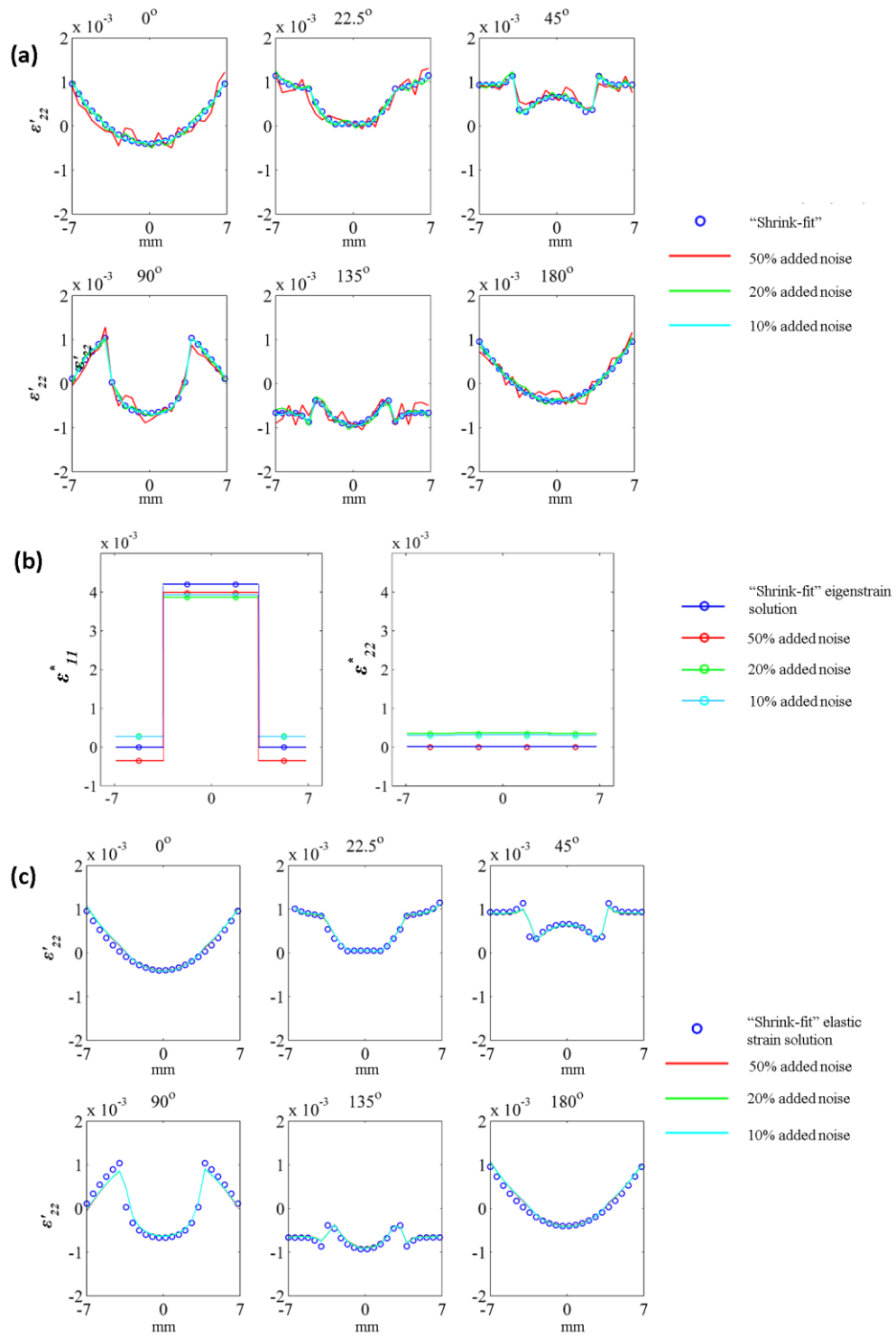


Figure 6.4: (a) Projection of simulated experimental data ε'_{22} with added noise at 10%, 20% and 50%, (b) comparison of the reconstructed ε^*_{22} and ε^*_{11} eigenstrain components and (c) the resulting reconstruction of ε'_{22} .

A direct relationship between the collected spectra and the simulated solution and the fitted ε'_{22} results on projections 0° , 22.5° , 45° and 180° is shown in Figure 6.5. The 90° and 135° projections appear to have reduced statistics, therefore the fitting appears to be less accurate.

The fluctuation of the side measurements at projection 0° is related to the decrease in size of the illuminated volume on the cylindrical sample. However, this fitting related uncertainty was not registered on the 22.5° , 45° and 180° projections.

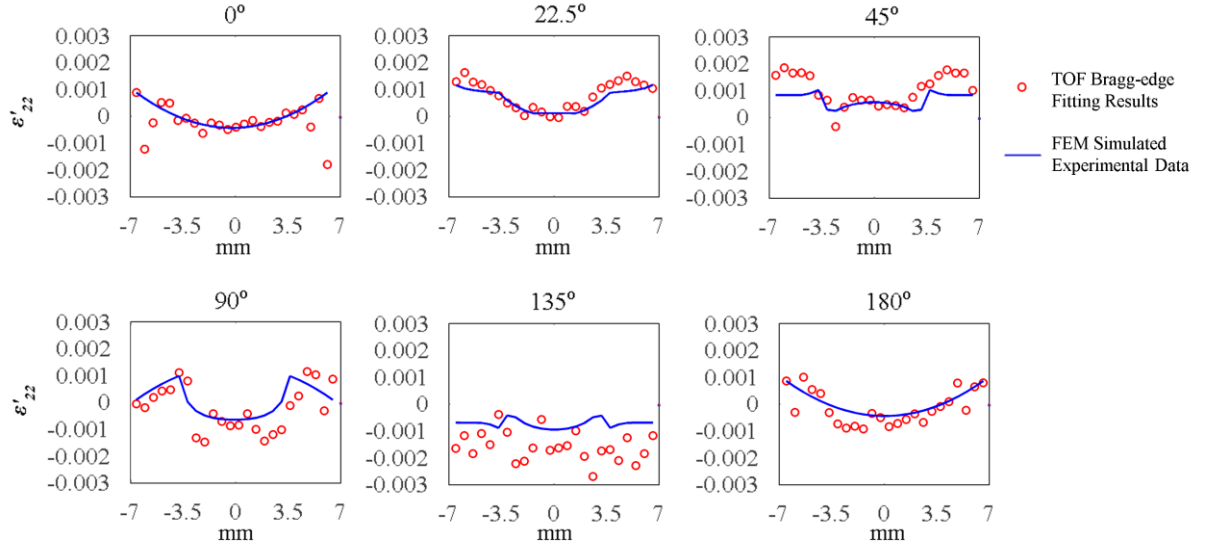


Figure 6.5: TOF Neutron Bragg-edge fitting experimental data plotted against the simulated projection strain data.

A direct comparison of the reconstructed eigenstrain components ε_{11}^* and ε_{22}^* at two discretisation segmentation steps, namely for 3.5mm and 1mm is presented in Figure 6.6 alongside the eigenstrain variation expected from the “ring-and-plug” sample. The fully constrained 3.5mm discretisation strategy shows remarkable agreement in terms of the reconstructed eigenstrain magnitude showing a robust convergence shown in Figure 6.6. The non-constrained $h = 1\text{mm}$ segmentation step contains a characteristic transition region close to $r = 3.5\text{mm}$ due to the intermediate $\varepsilon_{11}^* = 2.071 \times 10^{-3}$ value calculated for the increment $3 < r < 4\text{mm}$. It is also important to highlight the slight fluctuation of ε_{22}^* component values in the inner circle part ($r \leq 3.5\text{mm}$) which manifests the convergence ability of the proposed approach. The values recorded for both segmentation approaches for the ε_{22}^* component were determined of the order of $<10^{-4}$ of the maximum expected ε_{22}^* value in the sample.

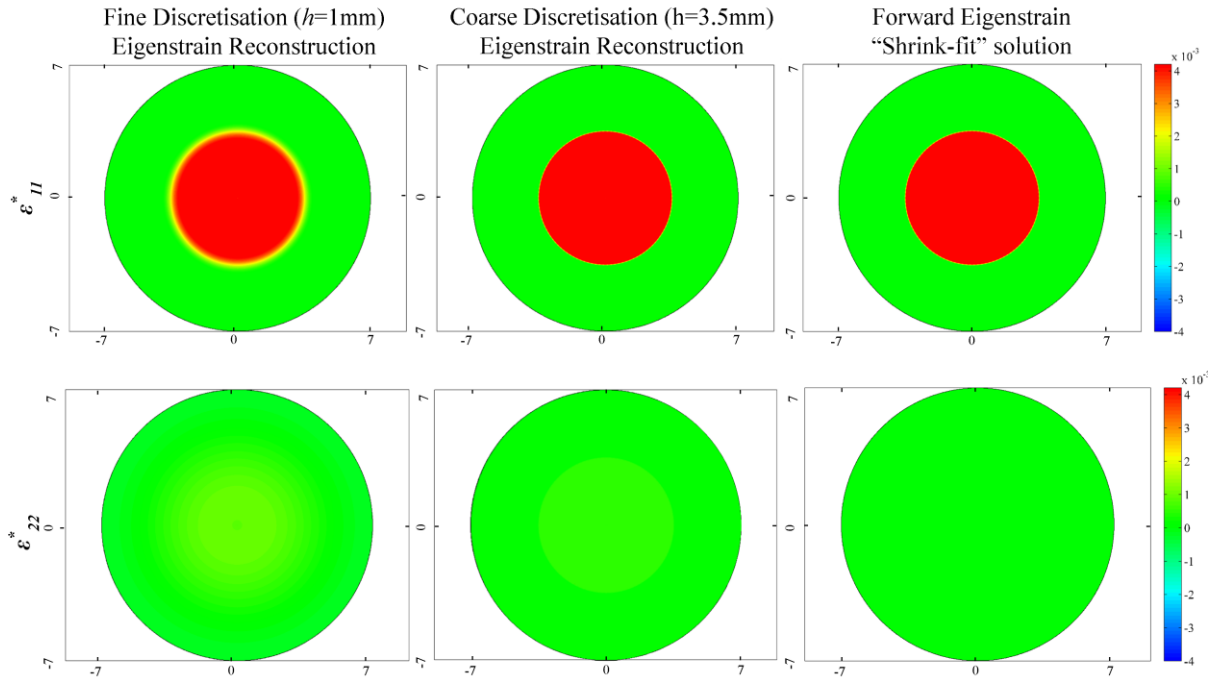


Figure 6.6: TOF-NDST reconstruction of eigenstrain for 3.5mm and 1mm increments comparison with FEM simulated applied strain variation on the shrink-fit 'ring' and 'plug' sample.

Following the eigenstrain component reconstruction for the ring and plug sample, non-axisymmetric elastic strain variation of the sample can be deduced, by substituting the calculated eigenstrain components into the FE model. The spatial resolution of the 2-D elastic strain variation reconstruction is determined by the amount of radial discretisation segmentation steps for eigenstrain calculation. In this case the resolution of the constrained solutions is governed by the 10x pixel binning value that was used during fitting.

The resulting reconstructions for the in-plane residual elastic strain components ε_{11} , ε_{22} and ε_{12} are presented in Figure 6.7. The scale bars used are individual to each of the strain components but consistent across all the different reconstruction approached. The 3.5mm discretisation approach is in very good agreement with the simulated elastic variation within the "shrink-fit" sample, both in terms of strain contour and magnitude. The 1mm discretisation segmentation does not capture the sharp strain gradient change in the near $r = 3.5\text{mm}$ region. The absolute strain magnitude is also smaller with respect to the simulated reconstruction.

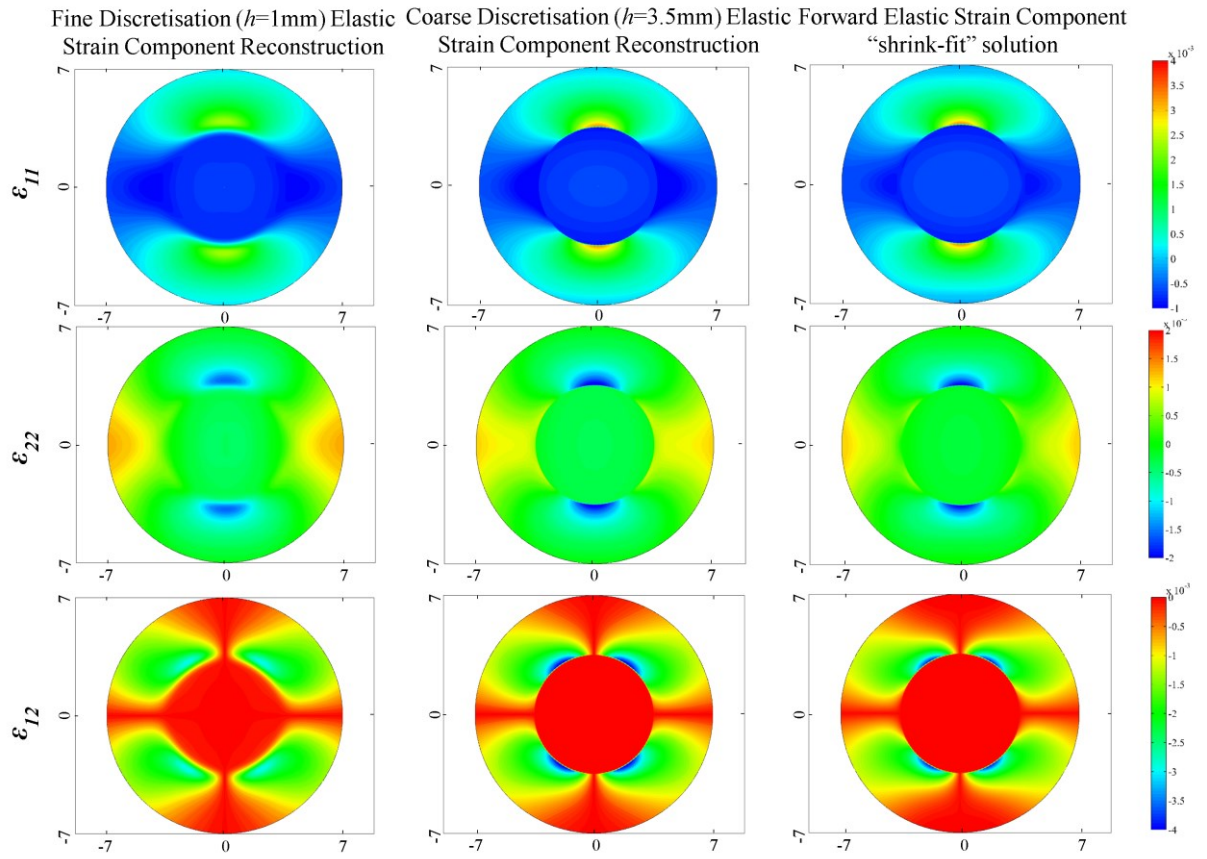


Figure 6.7: TOF-NDST reconstruction of in-plane residual elastic strain components ε_{11} , ε_{22} and ε_{12} for 3.5mm and 1mm discretisation steps compared with FEM simulated applied strain variation on the "shrink-fit" sample.

The consequences of the “misfit” strains defined by the "ring and plug" geometry are represented by the two-fold symmetry of the ε_{11} and ε_{22} components and the four-fold symmetry of the ε_{12} component, shown in Figure 6.7. The numerical error associated with the 1mm discretisation segmentation strategy is <10% which is not considered sufficient for strain determination purposes. Whereas, the 3.5mm discretization segmentation leads to <2% numerical error. The error value reflects the accumulation of aberrations as a result of the noise in the collected projection data and the low amount of projections that were used for the inverse calculation.

6.7. Conclusions

In the present investigation the first application of TOF-NDST for the reconstruction of non-symmetric strain distributions is reported. Six angular projection measurements of the strain component in along the beam direction were used for the purposes of the reconstruction via a

FE-based inverse eigenstrain formulation. The combination of state-of-art high resolution neutron TOF area detector and the tomographic acquisition allowed the formulation of an inverse problem allowing the reconstruction of full 2-D in-plane strain state within the specimen.

For the non-axisymmetric ring-and-plug sample, the solution for the strains has been constructed based on the radial discretisation of piece-wise constant eigenstrain variation. The FE environment of the system formulation ensures that strain compatibility and traction-free boundary conditions are strictly enforced in the solution. *A priori* knowledge of the magnitude and gradient of pre-existing inelastic strain variation are significant factors influencing the experimental acquisition strategy and allowing a more targeted set up in terms of the number of angular projections, and the data binning strategy. For example, increasing the number of collected projections and decreasing the pixel binning parameters, whilst ensuring adequate statistics for lattice parameter determination, directly influences the spatial resolution for recovering strain variation at finer length scales. In the case of neutron beams long exposure times that will allow for better statistics and an increase of the total number of collected projections may not be feasible depending of the specimen size and its scattering properties. Overall, it is important for the purposes of efficient data acquisition and modelling to adopt a constrained simulation strategy according to the specimen loading conditions or processing history.

Chapter 7

Conclusions and overview

In the course of this thesis several applications of combined X-ray imaging and diffraction techniques have been investigated, providing novel approaches for structural and mechanical analysis of engineering components. A variety of modes were employed in terms of the spatial resolution (length-scale), time resolution (frequency), and the nature of the physical quantity being interrogated. The diversity of the techniques analysed overall reflects only some of the immense versatility of X-ray synchrotron-based instruments that exploit the potentials of high-brilliance, hard X-ray probes in the engineering science field.

Mechanical engineering is a result-focused applied science that has been traditionally based upon empirical observations and approximations related to the material response. Robust knowledge of the intricate failure mechanisms related to the material structure at the micro and nano-scopic level can significantly enhance the existing technology by introducing lightweight, low-cost, efficient and reliable engineering structures. A great variety of engineering samples have been investigated non-destructively yielding valuable information regarding the mechanical behaviour aspects of the examined specimens, and a useful insight into micro-scale structure-property relationships. The studies carried out can be categorised into materials applications and component testing contexts, whereas different experimental approaches can primarily be classified according to the mode of interaction between beam and matter that was used.

In the study of large *in situ* deformation of materials, parallel beam X-ray computed tomography and digital volume correlation were used to measure the 3-D full-field displacements. *In situ* tomographic acquisition during compression experimental setup was developed in TOMCAT beamline (SLS). In the first set of experiments an in-house designed and built 1kN tomography compatible compression rig was used in the study of co-spayed (Al-12%Si)-12%Ti metal-metal composite. The reconstructed data were analysed using commercial LaVision Davis[®] DVC software package. In order to achieve the best match with the experiment, realistic boundary conditions were applied to a continuum FE model, and results were compared with the observation. At the homogenised, continuum level, satisfactory agreement was achieved between the DVC results and FE modelling predictions. It was also shown that the analysis of microscopic displacement fields around individual inclusions was possible on the basis of multi-scale DVC analysis. The demonstration of determining local strain concentrations by CT-DVC opens up the prospects of systematic application of this approach to non-destructive analysis of inhomogeneous internal deformation within the bulk of samples of microstructured materials.

For the second implementation of the method, an *in situ* tomographic experimental setup was used in I12 beamline (DLS). A polygranular Gilsocarbon graphite sample was examined during quasi-static fracture. An in-house MATLAB[®] algorithm was used for the synchrotron experimental data reconstruction, filtering, artefact removal, etc. The 3-D crack opening displacements were mapped using digital volume correlation, identifying the damage and fracture process zone with the size of approximately 1.5mm, which was found to be consistent with the fracture properties of this material. A 3-D cohesive zone modelling approach was used to simulate successfully the experimentally observed crack opening displacement (COD) and fracture process zone. The effect of out-of-plane constraint on the fracture resistance was deduced, and was proposed to be a general characteristic of quasi-brittle materials. Hence the conclusion was drawn that a more accurate damage and fracture model would require a cohesive law with constraint-dependent parameters.

Next, the developments of novel X-ray “rich” imaging non-destructive experimental techniques and data interpretation were presented. Namely, Diffraction Strain Tomography (DST) and Laue Orientation Tomography (LOT) were conceived, developed and implemented in various materials and components studies. The principle of DST and LOT proposed (an instance of broader “rich” tomography methodology) makes use of the reconstruction tomography technique widely employed in imaging applications. Redundant data sets are collected, with each location within the object multiply sampled by incident X-ray beams travelling through the specimen at different angles. In both cases, "pencil" beam translation-rotation acquisition is used, where a micron sized collimated beam is rastering across the specimen, in steps specified by the size of the probe. The procedure is repeated at different rotation angles from 0° to 180° in 2° to 3° increments. The strain value computed using the overall lattice spacing, during DST acquisition, represents the average strain value within the sampling volume, provided the calculation is based on the correct sample–detector distance for the centre of each gauge volume. This hypothesis was tested and validated using numerical simulations. Further insight into the origins of geometrically imposed pseudo-strains due to the sample detector distance variation at different scanning positions were taken into consideration. The data were then subjected to a mathematical inversion procedure that allows the reconstruction of the internal distribution of a particular component of strain.

In the first application of the DST method an experimental diffraction setup in I12 (DLS) was used allowing for simultaneous absorption tomography data to be acquired. A Ni-base superalloy turbine blade fabricated by Direct Laser Deposition (DLD) was investigated in terms of residual elastic strain distribution and preferred in the bulk due to the processing history, as well as dimensional accuracy as a result of the sintering fabrication manufacturing technique. The subject of the present study was a turbine blade manufactured from a C263 nickel-base superalloy using the additive procedure of Direct Laser Deposition (DLD). The key advance reported in the investigation concerned the utilisation of tomographic projections and subsequently of absorption intensity reconstructions for fine-tuning of the position of the

specimen with respect to the diffraction detector. The outcome 2-D cross-sectional mapping of the out-of-plane residual elastic strain component was found to be in reasonable agreement with the contour method for residual out-of-plane stress determination at the same location. A recently proposed eigenstrain modelling approach offering the ability to recover the 3-D stress component variation from the contour data will allow direct validation and error assessment between the two methods.

In the second section a multi-scale analysis of human dental tissue, combining two non-destructive synchrotron-based experimental techniques, wide and small X-ray scattering (WAXS/SAXS) was attempted (I22 beamline, DLS). Quantitative spatial distribution maps were obtained for various parameters in the human dental tissues, such as mean HAp crystallite thickness by the interpretation of SAXS patterns, and lattice parameter distribution across the DEJ by DST-WAXS analysis. A multi-phase DST approach was implemented resulting in the separate reconstruction of the dentine and enamel lattice parameter distributions. The complex nature of dental tissue means that several sources of lattice parameter variation are present, including mechanical strain, local chemical composition variation, and other aspects of inherent structural evolution. A combined FIB-DIC approach was used in addition to the diffraction experiments to assess and correlate mechanical strain contribution to the lattice parameter variation. The output result of the FIB milling was the in-plane strain components (ϵ_{11} , ϵ_{22}) on a line of equally spaced points across the sample surface. In comparison with DST results for ϵ_{33} out-of-plane strain component in dentine, similar variation trends were identified towards the dentine-enamel junction. The DST results in the enamel show a large out-of-plane residual elastic strain variation up to 5%, whereas the FIB-DIC indicates a close to uniform in-plane strain variation across the same line. It is proposed that FE-based analysis be adopted incorporating the mechanical state information together with multi-scale structure-related information. This approach will be able to correlate the data collected via different experimental routes, thus giving more definitive results.

In the third section of the Chapter, DST was used to investigate the applied bending strains in sintered polycrystalline multi-unit fixed prosthetic dentures. The diffraction measurements were carried in ID15 beamline (ESRF). A dental coping (bridge) sample mounted on a PMMA base was tested under application of 300 N applied bending load simulating mastication loading. To assist in carrying out strain analysis of bridge deformation under simulated mastication loading, a solid CAD model was constructed. The input used for model assembly was derived from the results of conventional absorption contrast X-ray tomographic reconstruction. Comparative results showed good correlation demonstrating the viability and potential value of DST for the analysis of internal stresses in all-ceramic zirconia frameworks. The method opens the way to (i) experimental investigation of the effects of FPD support arrangements on stress distributions, and hence strength; (ii) validation of numerical models; and (iii) experimental analysis of the effects of heat treatment, application of porcelain veneer, fatigue, corrosion, etc.

Lastly, the latest developments on Laue Orientation Tomography were presented. The first LOT experimental setup was implemented in I12 beamline (DLS). Grain specific information were obtained using a high-energy transmission Laue technique, across the specimen at 2° rotation increments. An initial indexation of the total number of grains (i.e. distinct orientations) in the bulk at 0° allows for a Laue pattern "prediction" algorithm to determine the grain specific orientation at subsequent rotation increments. A Laue spot generation algorithm is then used to predict the resulting Laue spot position on the detector, using fcc crystal lattice information, energy bandwidth and the geometry layout of the experiment. Ellipsoidal masks are produced on the predicted spot locations and the grain specific intensity across the specimen is "harvested" for each grain at different angles. An improved algorithmic approach is proposed resulting in improved grain reconstruction within a polycrystalline annealed Ni sample allowing for better grain intensity reconstructions to be achieved. Furthermore, using a Rodriguez vector formulation reconstruction of intragranular deformation is achieved. The grain shape reconstruction, using grain-specific diffracted intensity, was validated using optical imaging in

the periphery of the sample and identifying the grain boundaries visible on the surface. A second experiment series has recently been undertaken in ID15 (ESRF) beamline on annealed Ni specimens pre-characterised by EBSD on the top and bottom surfaces. The preliminary analysis of the data shows good correlation and the future aim is to cross-correlate the diffraction data intragranular mis-orientation reconstruction with the obtained high-resolution EBSD maps.

The usefulness of the "hybrid" imaging and diffraction *in situ* acquisition is exploited in three state-of-art experimental setups presented in the hierarchical order of increasing spatial resolution. The **first case study** describes the details and implementation of an *in operando* mechanical analysis experimental setup. The project investigated the feasibility of using synchrotron X-ray beams for time-resolved *in situ* imaging and diffraction of the interior components of an internal combustion engine during its operation. The experiments were carried out on beamline I12 (JEEP) at Diamond Light Source, U.K. A series of time-resolved imaging results from various engine parts were presented and the dynamic internal strain within the moving con-rod was evaluated with the accuracy of $\sim 50 \times 10^{-6}$ at a spatial resolution of $100 \times 100 \mu\text{m}^2$. Further development of the ideas outlined in the study is envisaged in terms of establishing close coupling between modelling of the engine operation (precise geometric representation of component shape; modelling the thermo- and fluid dynamics of gas flow through the engine; the incorporation of inertia effects in deformation) and experimental model validation.

The **second case study** addressed the challenge of identifying and mapping the strain in a Focused Ion Beam (FIB) milled region ($75 \times 75 \mu\text{m}^2$) at the surface of a nanocrystalline metallic coating of sub-micron thickness. A Transmission X-ray diffraction setup was used for data collection at DIFFABS beamline (Synchrotron Soleil, France). A 150nm-thick multi-layered W-Cu nano-composite thin film on polyimide (Kapton[®]) substrate was studied. The samples were loaded incrementally using a compact uniaxial loading device, and micro-beam diffraction data at a $30 \times 30 \mu\text{m}^2$ spatial resolution were collected on and away from the reference array. It was shown experimentally that the "island" array remained approximately strain free

throughout the experiment, providing an on-board d_o lattice spacing reference. The changing lattice spacing d in the coating was also monitored away from the array, to deduce the elastic strain evolution during deformation and associate to the macroscopic applied strain from the rig.

In the **third case study** an ultra-high resolution nano-probe was used to evaluate the residual strain profile within unidirectional SiC fibres with carbon monofilament core used in aerospace engine casing. A transmission X-ray diffraction setup, and optical focusing down to a $0.3 \times 0.5 \mu\text{m}^2$ spatial resolution was used for data collection on beamline B16 at Diamond Light Source, U.K. By combining synchrotron-based imaging and nano-focused X-ray beam scattering with Focused Ion Beam stress evaluation, detailed maps of structure and strain inside this material were presented. Eigenstrain modelling was used to reconstruct the full residual strain state within the fibre cross-section. The joined-up experimental and theoretical approach allowed extracting information about fibre structure down to the nanoscale, developing insight into its processing history, and revealing the existence of significant residual strains that have a strong effect on the performance of CFs in service.

In the last Chapter of the first application of the Time-of-Flight Neutron Diffraction Strain Tomography (TOF-NDST) for the determination of a non-axisymmetric elastic strain distribution within a shrink-fit ‘ring-and-plug’ Al-alloy sample is discussed. The experimental setup was set in ENGIN-X (ISIS) using tomographic data acquisition principle. Neutron TOF spectra were collected on a micro-channel plate neutron area detector with the pixel size of $55 \mu\text{m}$. Eigenstrain (ϵ^*) - FE framework was used for the solution of the direct eigenstrain problem, that is, the calculation of residual elastic strain distribution within a right circular cylinder due to a known distribution of eigenstrain. The convergence of the method is analysed by carrying out numerical experiments and considering the effects of the number of rotation increments, as well as the data quality, uncertainty and noise.

The experimental observations made in this thesis serve the purpose of non-destructive characterisation of deformation in engineering materials. The tomographic acquisition and computer reconstruction together with DVC allows obtaining 3-D full-field displacement information. The application of DVC is straightforward through the use of commercial software and relies on the quality and the spatial resolution of the acquired tomographic data.

Diffraction strain tomography (DST) is another “rich” tomography technique using tomographic acquisition principles of diffraction data to reconstruct the 2-D variation of the out-of-plane strain component within the bulk. The resolution depends on the effective collimated beam size, as well as on the number of rotation increments. DST is proposed in order to overcome the limitation of elongated gauge volume at small diffraction angles characteristic of high-energy X-ray beams.

A novel means of obtaining the accurate knowledge of grain shape and orientation within the bulk of polycrystalline aggregates is proposed through LOT. The Laue high-energy transmission configuration has been used successfully to recover the diffracted intensity related to this grain at different sample rotation increments. A tomographic reconstruction of multi grains within a 2-D cross section of a polycrystalline material can be achieved. In addition a Rodriguez vector (RV) formulation, valid for small relative intragranular misorientation ($<0.1^\circ$), has been used to reconstruct the relative misorientation within individual grains. The potential fields of application of engineering measurements using *in situ* and *in operando* engineering setups at state-of-the-art synchrotron-based experimental beamlines has been demonstrated for different length scales. The ability to use nano-focused X-ray beams in order to map the evolution of mechanical strains and structure within high-performance miniaturised components has also been investigated. In addition, the world first application of time-resolved stress mapping within ICE con-rods at 2000rpm paves the way to a series of non-destructive experimental investigations in “real” life engineering assemblies operating at high-frequencies ($>100\text{Hz}$).

Finally, the TOF-NDST has been developed in order to demonstrate the ability to handle data of increased dimensionality, i.e. for the reconstruction of unknown quantities of tensorial nature, such as multi-component strains. With the advantage of increased flux available in synchrotron X-ray diffraction facilities, larger number of projection angles can be covered (typically 2° or 3° increments are used) for 180° overall coverage, which will increase the robustness and accuracy of the reconstruction method. Therefore, the proposed implementation can be used to reconstruct non-axisymmetric 3-D elastic strain variation using X-ray diffraction strain tomography (DST) acquisition. The author believes that the presented inverse eigenstrain approach sets the foundation for further development of the technique into a general tool for strain imaging via neutron and X-ray diffraction.

References

- [1] Willmott P. An Introduction to Synchrotron Radiation: John Wiley & Sons, Ltd; 2011.
- [2] Berger M, Hubbell J, Seltzer S, Coursey J, Zucker D. XCOM: Photon Cross Section Database 1.2. 1999.
- [3] J. Als-Nielsen. CR Phusique. 2008;9:479-86.
- [4] Baimpas N, Lunt A, Dolbnya I, Korsunsky A. Nano-scale mapping of lattice strain and orientation inside carbon core SiC fibres by synchrotron X-ray diffraction. Carbon. 2014;In press.
- [5] Hung YC, Withers PJ. Fibre bridging during high temperature fatigue crack growth in Ti/SiC composites. Acta Materialia. 2012;60:958-71.
- [6] McLaren P1. <http://www.autoexpress.co.uk/mclaren/p1/63871/mclaren-p1-pictures/page/5/0>. 2013.
- [7] Deurbergue A, Oberlin A. TEM study of some recent high modulus pan-based carbon fibers. Carbon. 1992;30:981-7.
- [8] Fortin F, Yoon SH, Korai Y, Mochida I. Structure of round-shaped methylnaphthalene-derived mesophase pitch-based carbon fibres prepared by spinning through a Y-shaped die hole. J Mater Sci. 1995;30:4567-83.
- [9] Fitzer E. Pan-based carbon fibers—present state and trend of the technology from the viewpoint of possibilities and limits to influence and to control the fiber properties by the process parameters. Carbon. 1989;27:621-45.
- [10] Ju Li ZS, and Evan Ma , Guest Editors. Elastic strain engineering for unprecedented materials properties MRS Bulletin. 2014;39:108-14.
- [11] Boresi A., Schmidt R., Advanced Mechanics of Materials: Jon Wiley & sons; 2000.
- [12] Mase T., Mase G. Continuous Mechanics for Engineers: CRC press; 1999.
- [13] Ashby MF. Philosophical Magazine 1970;21:399 – 424
- [14] Withers PJ, Bhadeshia H. Residual stress part 1 and 2 - Measurement techniques Materials Science and Technology. 2001;17:355-65.
- [15] Webster PW. Neutron and Synchrotron X-ray Strain Scanning. Strain. 2001;37.
- [16] Allen A, et al., . . Measurement of internal stress within bulk materials using neutron diffraction. NDT International. 1981;14:249-54.
- [17] Hammond Ch. The Basics of Crystallography and Diffraction: Oxford Science Publications; 2002.
- [18] Compton A. PhysRev. 1923;21:483-502.
- [19] Kak AC, Slaney M. Principles of computerized tomographic imaging. Biology Society: IEEE Press; 1988.
- [20] Baimpas N, Drakopoulos, M., Connolley, T., Song, X., Pandazaras, C., Korsunsky, A.M.,. A feasibility study of dynamic stress analysis inside a running internal combustion engine using synchrotron X-ray beams. J of synch rad. 2013;20:316-23.
- [21] Korsunsky A.M., Vorster, W.J.J. , Zhang, S.Y. , Dini, D., Latham, D.,. The principle of strain reconstruction tomography: determination of quench strain distribution from diffraction measurements. Acta Materialia. 2006;54:2101–8.

- [22] Korsunsky A.M., Baimpas N, Song X, Belnoue J, Hofmann F, Abbey B, et al. Strain tomography of polycrystalline zirconia dental prostheses by synchrotron X-ray diffraction. *Acta Materialia*. 2011;59:2501-13.
- [23] Baimpas N, Xie M, Reinhard C, Korsunsky AM. The application of geometry corrections for Diffraction Strain Tomography (DST) analysis of a Ni-base superalloy blade. *Powder Diffraction*. 2013;28:pp. 436-47.
- [24] F. Hofmann BA, L. Connor, N. Baimpas, X. Song, S. Keegan, A. M. Korsunsky. Imaging of grain-level orientation and strain in thicker metallic polycrystals by high energy transmission micro-beam Laue (HETL) diffraction techniques. *International Journal of Materials Research*. 2011:P. 192-9.
- [25] ESRF. Coherent Image Formation, Absorption and Phase-Contrast Tomography. 2011.
- [26] Cloetens P. Phase Contrast Imaging - Coherent Beams. School on X-ray imaging Techniques ESRF2007.
- [27] L. Salvo CP, Maire E., Zabler S., Blandin J. J., Buffière J. Y., Ludwig W., Boller E., Bellet D., Josserond C., . X-ray micro-tomography an attractive characterisation technique in materials science. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*. 2003;200:273-86.
- [28] Donath T, Pfeiffer F, Bunk O, Groot W, Bednarzik M, Grünzweig C, et al. Phase-contrast imaging and tomography at 60 keV using a conventional x-ray tube source. *Review of Scientific Instruments*. 2009;80:053701.
- [29] Stampanoni M, Mokso R, Marone F, Vila-Comamala J, Gorelick S, Trtik P, et al. Phase-contrast tomography at the nanoscale using hard x rays. *Physical Review B*. 2010;81:140105.
- [30] Snigirev A, Snigireva I, Kohn V, Kuznetsov S, Schelokov I. On the possibilities of x-ray phase contrast microimaging by coherent high-energy synchrotron radiation. *Rev Sci Instr*. 1995;66:5486-92.
- [31] P. Cloetens MP-Se, J.-Y. Buffière, G. Peix, J. Baruchel, F. Peyrin, M. Schlenker, J. Observation of microstructure and damage in materials by phase sensitive radiography and tomography *J Appl Phys*. 1997;81.
- [32] Warren BE. X-ray Diffraction: Addison-Wesley Pub. Co.; 1969.
- [33] Steuwer A, Santisteban, J.R., Turski, M., Withers, P.J., Buslaps, T. High-resolution strain mapping in bulk samples using full-profile analysis of energy dispersive synchrotron X-ray diffraction data. *Nuclear Instruments and Methods in Physics Research B*. 2005;238:200-4.
- [34] Korsunsky AM, Song, X., Hofmann F, Abbey B., Xie, M., Connoley, T., Reinhard, C., Atwood, R.C., Connor, L., Drakopoulos, M.,. Polycrystal deformation analysis by high energy synchrotron X-ray diffraction on the I12 JEEP beamline at Diamond Light Source. *Mat Let*. 2010;64:1724-7.
- [35] Rowles M.R. On the calculation of the gauge volume size for energy-dispersive X-ray diffraction. Erratum. *Journal of Synchrotron Radiation*. 2011;18:p.938-41.
- [36] Hofmann F. Probing the Deformation of Ductile Polycrystals by Synchrotron X-ray Micro-diffraction.: University of Oxford; 2011.
- [37] Hofmann F, Song, X., Abbey, B., Jun, T-S., Korsunsky, A.M.,. High Energy Transmission Laue (HETL) micro-beam X-ray diffraction: a probe for intra-granular lattice orientation and elastic strain in thicker samples. *Journal of Synchrotron Radiation* 2012;19:307-18.
- [38] Larson BC, Tamura, N., Chung, J. S., Ice, G. E., Budai, J. D., Tischler, J. Z., et al. Materials Research Society Symposium - Proceedings 2000;590:47–252.
- [39] Barabash RI, Ice, G. E., Pang, J. W. L., . Materials Science and Engineering A. 2005;400:125–31.
- [40] Budai JD, Yang, W., Tamura, N., Chung, J.-S., Tischler, J. Z., Larson, B. C., Ice, G. E., Park, C., and Norton, D. P. . *Nat Mater*. 2003;2:487–92.
- [41] Poulsen H.F., Nielsen S.F., Lauridsen E.M., Schmidt S., Suter R.M., Lienert U., et al. Three-dimensional maps of grain boundaries and the stress state of individual grains in polycrystals and powders. *Journal of Applied Crystallography*. 2001;34:p.751-6
- [42] King AH, M.; Ludwig, W.; Reischig, P.; Lauridsen Mejdal, Erik; Marrow, T.; Buffière, J.Y. Non-destructive analysis of micro texture and grain boundary character from X-ray diffraction contrast tomography. . *Nuclear Instruments & Methods in Physics Research Section B: Beam Interactions with Materials and Atom*. 2010;268:291-6.
- [43] Steuwer A, Withers PJ, Santisteban JR, Edwards L, Bruno G, Fitzpatrick ME, et al. Bragg Edge Determination for Accurate Lattice Parameter and Elastic Strain Measurement. *Physica Status Solidi (A), Applied Research*. 2001;185:pp.221-30

- [44] Santisteban JR, Edwards L, Steuwer A, Withers PJ. Time-of-flight neutron transmission diffraction. *Journal of Applied Crystallography*. 2001;34:289-97.
- [45] Tremsin AS, McPhate JB, Steuwer A, Kockelmann W, M Paradowska A, Kelleher JF, et al. High-Resolution Strain Mapping Through Time-of-Flight Neutron Transmission Diffraction with a Microchannel Plate Neutron Counting Detector. *Strain*. 2012;48:296-305.
- [46] Abbey B, Zhang SY, Vorster W, Korsunsky AM. Reconstruction of axisymmetric strain distributions via neutron strain tomography. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*. 2012;270:28-35.
- [47] Tebedge N, Alpsten, G., Tall, L.,. Residual-stress measurement by the sectioning method. , 1973. 13(2): p. Experimental Mechanics. 1973;13:88-96.
- [48] Bueckner HF. The propagation of cracks and the energy of elastic deformation. *Trans ASME* 1958;80:1225-30.
- [49] Prime MB. CROSS-SECTIONAL MAPPING OF RESIDUAL STRESSES BY MEASURING THE SURFACE CONTOUR AFTER A CUT. *Journal of Engineering Materials and Technology*. 2001;Volume 123 P.162-8.
- [50] Prime M.B., Hughes, D.J., Webster, P.J.,. Proceedings of 2001 SEM annual conference on experimental and applied mechanics,Portland. 2001:608–11.
- [51] Kartal M., Liljedahl, CDM., Gungor, S., Edwards, L., Fitzpatrick, ME.,. Determination of the profile of the complete residual stress tensor in a VPPA weld using the multi-axial contour method. *Acta Materialia*. 2008;56:4417-28.
- [52] Group VM. Measurement of Residual Stresses by the Hole-Drilling Strain Gage Method. R Tech Note TN-503. 2001.
- [53] Wu Z. H, B., . Study of residual stress distribution by a combined method of moiré interferometry and incremental hole drilling. *J Appl Mech*. 1998;65:Part I:837-43, Part II:44-50.
- [54] Nelson DV. Residual-stress determination through combined use of holographic interferometry and blindhole drilling. *Exp Mech*. 1986;26:371–8.
- [55] Korsunsky AM, Sebastiani M, Bemporad E. Focused ion beam ring drilling for residual stress evaluation. *Materials Letters*. 2009;63:1961-3.
- [56] Winiarski B, et al.,. Mapping Residual Stress Distributions at the Micron Scale in Amorphous Materials. *Metallurgical and Materials Transactions a-Physical Metallurgy and Materials Science*. 2010;41A:1743-51.
- [57] Sebastiani M., Eberl, C., Bemporad, E., Pharr, G.M., . Depth-resolved residual stress analysis of thin coatings by a new FIB-DIC method. *Mat Sc and Eng A* 2011;528:7901-8.
- [58] Lunt A.J.G., Korsunsky, A.M.,. Intragranular Residual Stress Evaluation Using the Semi-Destructive FIBDIC Ring-Core Drilling Method. *Advanced Materials Research*. 2014;In press.
- [59] Mura T., *Micromechanics of Defects in Solids*, Martinus Nijhof, The Hague 1982.
- [60] Korsunsky AM, Regino GM, Nowell D. Variational eigenstrain analysis of residual stresses in a welded plate. *International Journal of Solids and Structures*. 2007;44:4574-91.
- [61] Korsunsky AM. Variational eigenstrain analysis of synchrotron diffraction measurements of residual elastic strain in a bent titanium alloy bar
Journal of Mechanics of Materials and Structures. 2006;1:pp. 101–19.
- [62] Song X, Korsunsky AM. Fully Two-Dimensional Discrete Inverse Eigenstrain Analysis of Residual Stresses in a Railway Rail Head. *Journal of Applied Mechanics*. 2011;78:031019-.
- [63] Bay B, Smith, T., Fyhrie, D., Saad, M. Digital volume correlation: Three-dimensional strain mapping using X-ray tomography. *Experimental Mechanics*. 1999;39:217-26.
- [64] Forsberg F, C. R. Siviour. 3D deformation and strain analysis in compacted sugar using x-ray microtomography and digital volume correlation. *Measurement Science and Technology*. 2009;20:095703.
- [65] Bing P, Dafang W, Zhaoyang W. Internal displacement and strain measurement using digital volume correlation: a least-squares framework. *Measurement Science and Technology*. 2012;23:045002.
- [66] Hierarchical Data Format, version 5. The HDF Group; 1997-2014.
- [67] Münch B. TP, Marone F., Stampanoni M. Stripe and ring artifact removal with combined wavelet—Fourier filtering. *Optics Express*. 2009;17:pp. 8567-91

- [68] Mostafavi m, Baimpas, N., Tarleton, E., Atwood, R.C., McDonald, S.A., Korsunsky, A.M., Marrow, T.J.,. Three-dimensional crack observation, quantification and simulation in a quasi-brittle material *Acta Materialia*. 2013;61:6276-89.
- [69] Quinta D, Mummery, PM, Withers, PJ. Full-field strain mapping by optical correlation of micrographs acquired during deformation. *Journal of Microscopy*. 2005;218:9-21.
- [70] Mostafavi M, T.J. Marrow. "Observation and quantification of three-dimensional crack propagation in poly-granular graphite". *Engineering Fracture Mechanics*, Under Review. 2012.
- [71] Liu L, Morgan, E.F., bone Aapodvciqdasit. Accuracy and precision of digital volume correlation in quantifying displacements and strains in trabecular bone. *Journal of Biomechanics*. 2007;pp. 3516-20.
- [72] Gates M. High Performance Digital Volume Correlation. University of Illinois, Illinois164. 2011.
- [73] Scarano F. Tomographic PIV: principles and practice. *Measurement Science and Technology*. 2013;24:012001.
- [74] Madi K, Tozzi, G, Zhang, QH, Tong, J, Cossey, A, Au, A,Hollis, D, Hild, F. Computation of full-field displacements in a scaffold implant using digital volume correlation and finite element analysis. *Medical Engineering & Physics*. 2013;35:1298-312.
- [75] Roux S, Hild, F., Viot, P., Bernard, D. Three-dimensional image correlation from X-ray computed tomography of solid foam. *Composites Part A: Applied Science and Manufacturing*. 2008;39:1253-65.
- [76] Mostafavi M, TJ. M. Fatigue and Fracture of Engineering Materials and Structures. 2012;35:695.
- [77] Nakhodchi S, Smith DJ, PEJ F. *Journal of Material Science*. 2012;In press.
- [78] Becker T, Tait R. Fracture and slow crack characteristics of nuclear grade graphite using the double torsion beam technique. 12th International Conference on Fracture. Ottawa, Canada.2009.
- [79] Droillard C, Lamon J. *Journal of American Ceramic Society*. 1996;79:849.
- [80] Hambli R. *Med Biol Eng Comput*. 2013;51:219.
- [81] Berto F, Lazzarin P, Ayatollahi M. *Carbon*. 2012;50:1942.
- [82] Berto F, Lazzarin P, Marangon C. *Mater Des*. 2012;41:421.
- [83] Hillerborg A, Modeer M, PE. P. *Cem Concr Res*. 1976;6:773.
- [84] Rosa AL, Yu RC, Ruiz G, Saucedo L, JLAO. S. *Engineering Fracture Mechanics*. 2012;82:195.
- [85] Song SH, Paulino GH, W. B. *Engineering Fracture Mechanics*. 2006;73:2829.
- [86] Ho FH. *Graphite design handbook*. General Atomics. 1988;San Diego, CA.
- [87] Ayatollahi M, Torabi A. *Carbon*. 2010;48:2255.
- [88] Karihaloo BL. *Fracture Mechanics and Structural Concrete*. Longman Scientific and Technical Publishers. 1995;New York.
- [89] Frantik P, Vesely V, Z. K. *Adv Eng Softw*. 2013;In Press.
- [90] Ouagne P, Neighbour GB, B. M. *JOURNAL OF PHYSICS D: APPLIED PHYSICS* 2002;35:927.
- [91] Mostafavi M, Smith DJ, MJ. P. *Fatigue Fract Eng Mater Struct*. 2010;33:724.
- [92] Barenblatt G. The mathematical theory of equilibrium cracks in brittle fracture. In: Dryden HL, von Kármán T, Kuerti G, van den Dungen FH, L. H, editors. *Advances in Applied Mechanics* New York: Academic Press; 1962.
- [93] Dugdale D. *J Mech Phys Solids*. 1960;8:100.
- [94] Mostafavi M, McDonald SA, Mummery PM, TJ. M. *Eng Fract Mech*. 2013;In Press.
- [95] Tarleton E, Charalambides MN, C. L. *Comput Mater Sci*. 2012;64:183.
- [96] Ingraffea AR, V. S. Numericak modelling of discrete crack propagation in reinforced concrete and plain concrete. In: Shi G, A. DT, editors. *Fracture Mechanics of Concrete*. Dordrecht: Martinus Nihhoff Publishers; 1985. p. 171.
- [97] Ruiz G, Ortiz M, A. P. *Int J Numer Methods Eng*. 2000;48:963.
- [98] Yu RC, G. R. *Int J Fract*. 2006;141:357.
- [99] Karihaloo BL, Abdalla HM, QZ. X. *Cem Concr Res*. 2006;36:171.
- [100] Choupani N. *Eng Fract Mech* 2008;75:4363.
- [101] Harper PW, SR. H. 2008;75.
- [102] Chandra N, Li H, Shet C, Ghonem H. *Int J Solids Struct*. 2002;39:2827.
- [103] Jones AN, Hall GN, Joyce MR, Hodgkin AD, Wen K, Marrow TJ, et al. *J Mater Sci Technol*. 2008;381:152.
- [104] Tsang DKL, BJ. M. *J Nucl Mater*. 2006;350:208.
- [105] DaVis. User's Manual. Gottingen: LaVision GmbH; 2012.
- [106] Ouchterlony F. *Int J Rock Mech Mining Sci*. 1988;25:71.

- [107] Mostafavi M, TJ. M. Engineering Fracture Mechanics. 2011;78:1756.
- [108] Mostafavi M, McDonald SA, Çetinel H, Mummery PM, TJ. M. Carbon. 2013;59:325.
- [109] Camanho P, Dávila C, De Mora M. J Compos Mater. 2003;37:1415.
- [110] Siegmund T, W. B. Int J Fract. 1999;99:97.
- [111] Hodgkins AD, Marrow TJ, Mummery PM, Marsden BJ, ASL. F. J Mater Sci Technol. 2006;22:1045.
- [112] Cornec A, Scheider I, Schwalbe K. Eng Fract Mech. 2003;70:1963.
- [113] ABAQUS. User's Manual. Version 6.10. Providence, Rhode Island: Dassault Systèmes Simulia Corp.; 2010.
- [114] Joyce M, Marrow, TJ, Mummery, PM, Marsden, BJ. Eng Fract Mech 2008;75:3633.
- [115] Henshell RD, KG. S. Int. J. Numer. Methods Eng. . 1975;9:495.
- [116] Tracey DM. Nucl Eng Des 1974;26:282.
- [117] Wilson WK. Some crack tip finite elements for plane elasticity. In: Corten HT, Gallagher JP, editors. Stress Analysis and Growth of Cracks. Proceedings of the 1971 National Symposium on Fracture Mechanics: Part I Philadelphia: ASTM STP 513. 1971.
- [118] Barsoum R. Int J Numer Methods Eng 1976;10:25.
- [119] Stern M, Becker, EB, Dunhama, RS. . . Int J Fract. 1976;12:359.
- [120] Sinclair GB. . Int J Fract 1985;28:3.
- [121] Hodgkins AD, Marrow TJ, Wootton MR, Moskovic R, PEJ. F. Mater Sci Technol. 2010;26:899.
- [122] Alan F. Bower. Applied Mechanics of Solids: CRC press; 2009.
- [123] Becker T, Marrow T, Tait R. J Nucl Mater. 2011;414:32.
- [124] Berre C, Fok, SL, Marsden, BJ, Mummery, PM, Marrow, TJ, Neighbour, GB. J Nucl Mater. 2008;380:46.
- [125] Kim J, Gao G, Srivatsan TS. Int J Solids Struct 2003;40:7357.
- [126] R6. Assessment of the Integrity of Structures Containing Defects: British Energy. 2001.
- [127] Ortiz M., Computational Micromechanics. Comput Micromechanics. 1996;18:pp. 321-38.
- [128] Xu X-P., Needleman A., Numerical simulations of fast crack growth in brittle solids. . Int J of Solids and Struct. 1994;42:pp. 1394-434.
- [129] Yang B., Mall S., Chandar K.R., A cohesive zone for fatigue crack growth in quasibrittle materials. Int J of Solids and Struct. 2001;38:pp. 3927-44.
- [130] Leclerc H, Périé, J.N., Roux, S., Hild, F. Voxel-Scale Digital Volume Correlation. Experimental Mechanics. 2011;51:pp. 479-90(12).
- [131] Kelly AJ, Mi J, Sinha GV, Krug P, Crosa F, Audebert F, et al. An Al-Si-Ti hierarchical metal-metal composite manufactured by co-spray forming. Journal of Materials Processing Technology. 2011;211:2045-9.
- [132] Baimpas N, Mostafavi, M., Abbey, B., Hofmann, F., Mokso, R., Mi, J., Grant, P., Marrow, TJ., Korsunsky, AM., Multi-scale in-situ deformation analysis of (Al-12%Si)-12%Ti metal-metal composite using Digital Volume Correlation (DVC) of synchrotron X-ray tomography datasets. JSA. 2014;In Press.
- [133] Marrow T, Mostafavi, M, McDonald, SA, Mummery, PM. . Observation and quantification of three-dimensional crack propagation in poly-granular graphite. ECF19 (19th European Conference on Fracture). 2012.
- [134] Christen D, Levchuk A, Schori S, Schneider P, Boyd S, Müller R. Deformable image registration and 3D strain mapping for the quantitative assessment of cortical bone microdamage. Journal of the Mechanical Behavior of Biomedical Materials. 2012;8:184-93.
- [135] Limodin N, Réthoré, J., Buffière, J.-Y., Hild, F., Roux, S., Ludwig, W., Rannou, J., Gravouil, A. Influence of closure on the 3D propagation of fatigue cracks in a nodular cast iron investigated by X-ray tomography and 3D volume correlation. Acta Materialia. 2010;58 (8):pp. 2957-67.
- [136] Forsberg F, Mooser, R., Arnold, M., Hack, E., Wyss, P. 3D micro-scale deformations of wood in bending: Synchrotron radiation μ CT data analyzed with digital volume correlation. Journal of Structural Biology. 2008:pp. 255-62.
- [137] Bay B, Smith T, Fyhrie D, Saad M. Digital volume correlation: Three-dimensional strain mapping using X-ray tomography. Experimental Mechanics. 1999;39:217-26.
- [138] Stampanoni M, Groso, A, Isenegger, A, Mikuljan, G, Chen, Q, Bertrand, A, Henein, S, Betemps, R, Frommherz, U, Böhler, P, et al. Trends in synchrotron-based tomographic imaging: the SLS experience. Proceedings of SPIE. 2006;6318:63180M--M-14.
- [139] J. Pl. Light Alloys - Metallurgy of the Light Metals. 1996.

- [140] Ramberg W, Osgood, WR. . Description of stress-strain curves by three parameters. Technical Note No 902, National Advisory Committee For Aeronautics, Washington DC. 1943.
- [141] Dave S, Song, X., Hofmann, F., Dragnevski, K., Korsunsky, A. M. Digital image correlation and finite element analysis of inter- and intra-granular deformation. *Procedia Engineering*. 2009;1:197-200.
- [142] Daymond MR, Bourke, MAM., VonDreele, R.B., Clausen, B., Lorentzen, T., . Use of Rietveld refinement for elastic macrostrain determination and for evaluation of plastic strain history from diffraction spectra. . *J Appl Phys* 1997;82:1554–62.
- [143] Korsunsky AM, Wells KE, Withers PJ. Mapping two-dimensional state of strain using synchrotron x-ray diffraction. *Scripta Materialia*. 1998;39:pp. 1705-12.
- [144] Fitch AN. The high resolution powder diffraction beam line at ESRF. *J Res NIST* 2004;109:133–42.
- [145] Korsunsky A.M., Collins S.P., Owen R.A., Daymond M.R, Achtioui S., K.E. J. Fast residual stress mapping using energy-dispersive synchrotron X-ray diffraction on station 16.3 at the SRS. *Journal of Synchrotron Radiation*. 2002;9:p.77-81.
- [146] Wanner A, Dunand, D.C. . Synchrotron X-ray study of bulk lattice strains in externally loaded Cu–Mo composites. . *Metall Mater Trans A* 2000;31:2949–62.
- [147] Dini D, Song, X., Zhang, S.Y., Korsunsky, AM.,. Residual strain analysis in polycrystalline aggregates using diffraction measurement and finite element modelling. *J Strain Anal Eng Des*. 2009;44:55–70.
- [148] Korsunsky AM, Jarnes, K.E., Daymond, M.R., . Intergranular stresses in polycrystalline fatigue: diffraction measurement and self-consistent modeling. *Eng Fract Mech*. 2004;71:805–12.
- [149] Salimon AI, Korsunsky, A.M., Ivanov, A.N.,. The character of dislocation structure evolution in nanocrystalline FCC Ni–Co alloys prepared by high-energy mechanical milling. *Mater Sci Eng A – Struct Mater Prop Microstruct Process*. 1999;271:196–205.
- [150] Croft M, Shukla, V., Jisrawi, N.M., Zhong, Z., Sadangi, R.K., Holtz, R.L., et al. Mapping and load response of overload strain fields: synchrotron X-ray measurements. *Int J Fatigue*. 2009;31:1669–77.
- [151] Ludwig W, King, A., Reischig, P., Herbig, M., Lauridsen, E.M., Schmidt S., et al. . New opportunities for 3D materials science of polycrystalline materials at the micrometre length scale by combined use of X-ray diffraction and X-ray imaging. *Mater Sci Eng A – Struct Mater Prop Microstruct Process*. 2009;524:69–76.
- [152] Tunna L, Barclay, P., Cernik, R.J., Khor, K.H., O’Neill, W., Seller, P., . The manufacture of a very high precision X-ray collimator array for rapid tomographic energy dispersive diffraction imaging (TEDDI). . *Measur Sci Technol* 2006;17:1767–75.
- [153] Tamura N., MacDowell A.A., Spolenak R., Valek B.C., Bravman J.C., Brown W.L., et al. Scanning X-ray microdiffraction with submicrometer white beam for strain/stress and orientation mapping in thin films. *Journal of Synchrotron Radiation*. 2003;10:p.137-43
- [154] Gupta V, Agnew, SR.,. Indexation and misorientation analysis of low quality Laue diffraction patterns. *J Appl Crystallogr*. 2009;42:116-24.
- [155] Bale HA. Interface residual stresses in dental zirconia using Laue micro-diffraction. Lawrence Berkeley National Laboratory LBNL.paper LBNL-1750E.
- [156] R.J. Moat AJP, L. Li, P.J. Withers, M. Preuss. Crystallographic texture and microstructure of pulsed diode laser-deposited Waspaloy. *Acta Materialia* 2009;57:p.1220-9.
- [157] Toyserkani E, Khajepour, A., and Corbin, S. *Laser Cladding*. CRC, LLC, Boca Raton, FL. 2000;Chap. 2.
- [158] Xie M, Baimpas, N., Reinhard, C., Korsunsky AM. Texture analysis in cubic phase polycrystals by single exposure synchrotron X-ray diffraction. *Journal of Applied Physics*. 2013;114:163502.
- [159] Korsunsky A, Xie, M, Baimpas, N, Song, X. X-ray Texture Analysis and Imaging of Engineering Materials at Oxford HEX-lab *Proceedings of the International MultiConference of Engineers and Computer Scientists* 2012. Hong Kong 2012.
- [160] De Sant'anna G, Dos Santos, EAP., Soares, LES., Santo, AMD., Martin, AA., Duarte, DA., et al. Dental Enamel Irradiated with Infrared Diode Laser and Photoabsorbing Cream: Part 1-FT-Raman Study. *Photomed Laser Surg*. 2009;27:499-507.
- [161] Vennat E, Bogicevic, C., Fleureau, JM., Degrange, M. Demineralized dentin 3D porosity and pore size distribution using mercury porosimetry. *Dent Mater*. 2009;25:729-35.
- [162] Johansen E, Parks, HF. . Electron Microscopic Observations on the 3-Dimensional Morphology of Apatite Crystallites of Human Dentine and Bone. *J Biophys Biochem Cy*. 1960;7:743.

- [163] Voegel J, Frank, RM. . Ultrastructural-Study of Apatite Crystal Dissolution in Human Dentin and Bone. *J Biol Buccale* 1977;5:181-94.
- [164] Gao H, Ji, BH., Jager, IL., Arzt, E., Fratzl, P. . Materials become insensitive to flaws at nanoscale: Lessons from nature. *P Natl Acad Sci USA*. 2003;100:5597-600.
- [165] White S, Miklus, VG., Chang, PP., et al. Controlled failure mechanisms toughen the dentino-enamel junction zone. *J Prosthet Dent*. 2005;94:330-5.
- [166] Young M, Almer, JD., Daymond, MR., Haeffner, DR., Dunand, DC. Load partitioning between ferrite and cementite during elasto-plastic deformation of an ultrahigh-carbon steel. *Acta Materialia*. 2007;55:1999-2011.
- [167] Mueller R, Rossoll, A., Weber, L., Bourke, MAM., Dunand, DC., Mortensen, A. Tensile flow stress of ceramic particle-reinforced metal in the presence of particle cracking. *Acta Materialia*. 2008;56:4402–16.
- [168] Young M, DeFouw, J., Almer, JD., Dunand, DC. Load partitioning during compressive loading of a Mg/MgB₂ composite. *Acta Mater*. 2007;55:3467–78.
- [169] Fratzl P, Schreiber, S., Klaushofer, K. Bone mineralization as studied by small-angle X-ray scattering. *Connect Tissue Res*. 1996;35:9-16.
- [170] Deymier-Black A, Yuan, F., Singhal, A., Almer, JD., Brinson, LC., Dunand, DC. Evolution of load transfer between hydroxyapatite and collagen during creep deformation of bone. *Acta Biomater*. 2012;8:253-61.
- [171] Singhal A, Almer, JD., Dunand, DC. Variability in the nanoscale deformation of hydroxyapatite during compressive loading in bovine bone. *Acta Biomater*. 2012;8:2747-58.
- [172] Almer J, Stock, SR. Internal strains and stresses measured in cortical bone via high-energy X-ray diffraction. *J Struct Biol*. 2005;152:14-27.
- [173] Almer J, Stock, SR. Micromechanical response of mineral and collagen phases in bone. *J Struct Biol*. 2007;157:365-70.
- [174] Deymier-Black A, Almer, JD., Stock, SR., Haeffner, DR., Dunand, DC. Synchrotron X-ray diffraction study of load partitioning during elastic deformation of bovine dentin. *Acta Biomater*. 2010;6:2172-80.
- [175] Almer J, Stock, SR. High energy X-ray scattering quantification of in situ-loading-related strain gradients spanning the dentinoenamel junction (DEJ) in bovine tooth specimens. *J Biomech*. 2010;43:2294-300.
- [176] Schroer C, Kuhlmann, M., Roth, SV., Gehrke, R., Stribeck, N., Almendarez-Camarillo, A., Lengeler, B. Mapping the local nanostructure inside a specimen by tomographic small-angle x-ray scattering. *Appl Phys Lett* 2006;88:164102.
- [177] Schindelin J, Arganeda-Carreras, I., Frise, E., Kaynig, V., Longair, M., Pietzsch, T., Preibisch, S., Rueden, C., Saalfeld, S., Schmid, B., et al. Fiji: an open-source platform for biological-image analysis. *Nature Methods*. 2012;9:676-82.
- [178] Sui T, Sandholzer, MA., Le Bourhis, E., Baimpas, N., Landini, G., Korsunsky, AM. Structure-mechanical function relations at nano-scale in heat-affected human dental tissue. *J Mech Behav Biomed Mater*. 2014;32:113-24.
- [179] Rinnerthaler S, Roschger, P., Jakob, HF., Nader, A., Klaushofer, K., Fratzl, P. Scanning small angle X-ray scattering analysis of human bone sections. *Calcif Tissue Int* 1999;64:422-9.
- [180] Tesch W, Vandenbos, T., Roschgr, P., Fratzl-Zelman, N., Klaushofer, K., Beertsen, W., Fratzl, P. Orientation of mineral crystallites and mineral density during skeletal development in mice deficient in tissue nonspecific alkaline phosphatase. *J Bone Miner Res*. 2003;18:117-25.
- [181] Sui T, Sandholzer, MA., Baimpas, N., Dolbnya, IP., Walmsley, A., Lumley, PJ., Landini, G., Korsunsky, AM. Multiscale modelling and diffraction-based characterization of elastic behaviour of human dentine. *Acta Biomater*. 2013;9:7937-47.
- [182] Kelly JR, Nishimura, I., Campbell, S.D.,. Ceramics in dentistry: historical roots and current perspective. *J Prosth Dent* 1996;75:18–32.
- [183] Pittayachawan P, McDonald, A., Young, A., Knowles, J.C.,. Flexural strength, fatigue life, and stress-induced phase transformation study of Y-TZP dental ceramic. *J Biomed Mater Res*. 2008;88B.
- [184] Fischer H, Weber, M., Marx, R.,. Lifetime prediction of all-ceramic bridges by computational methods. *J Dent Res*. 2003;82:238–42.
- [185] Dittmer MP, Kohorst, P., Borchers, L., Stiesch-Scholz, M.,. Finite element analysis of a four-unit all-ceramic fixed partial denture. *Acta Biomater*. 2009;5:1349-55.

- [186] Studart AR, Filser, F., Kochera, P., Luthy, H., Gauckler, L.J.,. Cyclic fatigue in water of veneer-framework composites for all-ceramic dental bridges. . *Dent Mater.* 2007;23:177-85.
- [187] Schmitter M, Mussotter, K., Rammelsberg, P., Stober, T., Ohlmann, B., Gabbert, O., . Clinical performance of extended zirconia frameworks for fixed dental prostheses: two-year results. *J Oral Rehab.* 2009;36:610–5.
- [188] Daniels JE, Drakopoulos, M., . High-energy X-ray diffraction using the Pixium 4700 flat-panel detector. . *J Synch Rad.* 2009;16:463–8.
- [189] Fewster P. *Critical Reviews in Solid State and Materials Sciences.* 1997;22:69-110.
- [190] Jakobsen B, Poulsen, HF., Lienert, U., Almer, J., Shastri, SD., Sorensen, HO., Gundlach, C., Pantleon, W. *Science.* 2006;312:889-92.
- [191] Déchamps M, Baribier, F, and Marrouche, A. Grain-boundaries: Criteria of specialness and deviation from CSL misorientation. *Metallurgica. Acta Biomater.* 1987;35:101-7.
- [192] West S, Schmidt, S, Sørensen, HO, Winther, G, Poulsen, HF, Margulies, L, Gundlach, C, Juul Jensen, D. . *Scripta Materialia.* 2009;61:875-8.
- [193] Shenoy P.S., Fatemi A. Dynamic analysis of loads and stresses in connecting rods. *Proceedings of the Institution of Mechanical Engineers. Part C: Journal of Mechanical Engineering Science.* 2006;220:p.615-24
- [194] Shenoy P.S. Dynamic load analysis and optimization of connecting rod. PhD thesis. 2004;Univ. of Toledo
- [195] Manner E. Continuous Acquisition of Measuring Data up to an Enviromental Temperature of 180°C (Temperature, Deformation, Pressure between Piston Rings). http://www.sensortelemetrie.de/fileadmin/user_upload/Downloads/Vertriebsunterlagen/Motormess technik_enpdf.
- [196] CompactRIO. Developing an Engine Connecting Rod Fatigue Test System Based on Compact RIO.
- [197] Ishida S., Hori Y., Kinoshita T., T. I. Development of Technique to Measure Stress on Connecting Rod during Firing Operation. *SAE Technical Paper* 951797. 1995.
- [198] Buffiere J-Y, Maire E, Cloetens P, Lormand G, Fougères R. Characterization of internal damage in a MMC, using X-ray synchrotron phase contrast microtomography. *Acta Materialia.* 1999;47:1613-25
- [199] Sharma H., Huizenga R.M., S.E. O. *Journal of Applied Crystallography.* 2012;45:p.693-704.
- [200] Daymond M, Young M, Almer J, Dunand D. Strain and texture evolution during mechanical loading of a crack tip in martensitic shape-memory NiTi. *Acta Materialia.* 2007;55:3929-42
- [201] Efstathiou C, Boyce D, Park J, Lienert U, Dawson P, Miller M. *Acta Materialia.* 2010;58:5806-19.
- [202] Thomas O., Loubens A., Gergaud P., P. L. *Applied Surface Science.* 2006;253:p.182-7.
- [203] San Martin D., Jimenez-Melero E., Duffy J.A., Honkimaki V., van der Zwaag S., N.H. vD. *Journal of Applied Crystallography.* 2012;45:p.748-57.
- [204] Stampanoni M., Marone F., Mikuljan G., Jefimovs K., Trtik P., Vila-Comamala J., et al. Broadband X-ray full field microscopy at a superbend. 9th International Conference on X-ray Microscopy. *Journal Physics Conf.* 2009.
- [205] Rack A., Garcia-Moreno F., Baumbach T., J. B. Synchrotron-based radioscopy employing spatio-temporal micro-resolution for studying fast phenomena in liquid metal foams. *Journal of Synchrotron Radiation.* 2009;16:p.423-34.
- [206] High Speed Camera v73.
- [207] Plech A., Kurbitz S., Berg K.J., Graener H., Berg G., Gresillon S., et al. Time-resolved X-ray diffraction on laser-excited metal nanoparticles. *Europhysics Letters.* 2003;61:p.762-8
- [208] Sutton MA, Wolters WJ, Peters WH, Ranson WF, McNeill SR. Determination of displacements using an improved digital correlation method. *Image and Vision Computing.* 1983;1:133-9.
- [209] Noyan IC, Huang TC, York BR. Residual stress/strain analysis in thin films by X-ray diffraction. *Critical Reviews in Solid State and Materials Sciences.* 1995;20:2, 125-177.
- [210] Thompson CV, Carel R. *Journal of the Mechanics and Physics of Solids.* 1996;44:657.
- [211] Guisbiers G, Overschelde OV, M W. Nanoparticulate origin of intrinsic residual stress in thin films. *Acta Materialia.* 2007;55: 3541
- [212] Sebastian KV. Properties of Sintered and Infiltrated Tungsten-Copper Electrical Contact Material. *The International Journal of Powder Metallurgy Powder Technology.* 1981;17:297-303.

- [213] Lee YJ, Lee BH, Kim GS, Kim DG, Kim DS, Y. D. Kim. Evaluation of conductivity in W–Cu composites through the estimation of topological microstructures. *Materials Letters*. 2006;60:2000-3.
- [214] Yoo S, Krupashankara MS, Sudarshan TS, R. J. Dowding. Ultrahigh pressure consolidation (UHPC) of W-Cu composites. *Materials Science and Technology*. 1998;14 no.2:170-4.
- [215] Luedtke A. Thermal Management Materials for High-Performance Applications. *Advanced Engineering Materials*. 2004;6:142-4.
- [216] Ilić DJ, Fiscina J, Oliver CG, Ilić N, Mücklich F. Self Formed Cu-W Functionally Graded Material Produced Via Powder Segregation. *Advanced Engineering Materials*. 2007;9:542-7.
- [217] Girault B, Geandier G, Eyidi D, Renault P-O, E. Le Bourhis, Goudeau P. Size effects on the mechanical behavior of nanometric W/Cu multilayers. *Materials Research Society Symposium Proceedings*. 2008;1086.
- [218] Mordike BL, Kaczmar J, Kielbinski M, Kainer KU. Powder powder mixtures decreased with increase in milling time. *Powder Metallurgy*. 1991;23:91.
- [219] Kim YD, Oh NL, Oh ST, I. H. Moon. Thermal conductivity of W–Cu composites at various temperatures. *Materials Letters*. 2001;51:420-4.
- [220] Withers P, Preuss, M, Steuwer, A, Pang, JWL. *J Appl Cryst*. 2007;40:891-904.
- [221] Chung J, Ice G. Automated indexing for texture and strain measurement with broad-bandpass x-ray microbeams. *Journal of Applied Physics*. 1999;86:5249-55.
- [222] Moore MG, Evans WP. *SAE Transact* 1958;6:340–5.
- [223] Song X, Yeap KB, Zhu J, Belnoue J, Sebastiani M, Bemporad E, et al. Residual stress measurement in thin films using the semidestructive ring-core drilling method using Focused Ion Beam. *Procedia Engineering*. 2011;10:2190 – 5.
- [224] Girault B, Eyidi D, Chauveau T, Babonneau D, Renault P-O, Bourhis EL, et al. Copper coverage effect on tungsten crystallites texture development in W/Cu nanocomposite thin films. *Journal of Applied Physics*. 2011;109:014305.
- [225] Loidl D, Peterlik H, Müller M, Riekel C, Paris O. Elastic moduli of nanocrystallites in carbon fibers measured by in-situ X-ray microbeam diffraction. *Carbon*. 2003;41:563-70.
- [226] Tanaka F, Okabe T, Okuda H, Ise M, Kinloch IA, Mori T, et al. The effect of nanostructure upon the deformation micromechanics of carbon fibres. *Carbon*. 2013;52:372-8.
- [227] Chung D. *Carbon Fiber Composites*: Butterworth-Heinemann; 1994.
- [228] Matsumoto T. Mesophase pitch and its carbon fibers. *Pure & Appl Chem*. 1985;57.
- [229] Edie D. *Fibers and composites*. New York: Taylor & Francis; 2003.
- [230] Edie D. The effect of processing on the structure and properties of carbon fibers. *Carbon*. 1998;36:345-62.
- [231] Klug HP, Alexander LE. *X-Ray Diffraction Procedures: For Polycrystalline and Amorphous Materials*. 2nd Edition ed: Wiley; 1974.
- [232] Song X, Yeap KB, Zhu J, Belnoue J, Sebastiani M, Bemporad E, et al. Residual stress measurement in thin films at sub-micron scale using Focused Ion Beam milling and imaging. *Thin Solid Films*. 2012;520:2073-6.
- [233] Xu Song, Solène Chardonnet, Giancarlo Savini, Shu Yan Zhang, Willem J.J. Vorster, Korsunsky AM. Experimental/Modelling Study of Residual Stress in Al/SiCp Bent Bars by Synchrotron XRD and Slitting Eigenstrain Methods. *Materials Science Forum*. 2008;277:pp. 571-2.
- [234] Takaku A, Shioya M. X-ray measurements and the structure of polyacrylonitrile- and pitch-based carbon fibres. *J Mater Sci*. 1990;25:4873-9.
- [235] Paris O, Loidl D, Peterlik H. Texture of PAN- and pitch-based carbon fibers. *Carbon*. 2002;40:551-5.
- [236] Baimpas N, Le Bourhis E, Eve S, Thiaudière D, Hardie C, Korsunsky A. Stress evaluation in thin films: Micro-focus synchrotron X-ray diffraction combined with focused ion beam patterning for do evaluation. *Thin Solid Films*. 2013;549:245-50.
- [237] Davies R, Riekel C, Koziol K, Vilatela J, Windle A. Structural studies on carbon nanotube fibres by synchrotron radiation microdiffraction and microfluorescence. *Journal of Applied Crystallography*. 2009;42:1122-8.
- [238] SPECIALTY MATERIALS I.

- [239] Warriar SG, Rangaswamy P, Bourke MAM, Krishnamurthy S. Assessment of the fiber/matrix interface bond strength in SiC/Ti-6Al-4V composites. *Materials Science and Engineering: A*. 1999;259:220-7.
- [240] Hutson A, John R, Jira J. Effect of temperature on fiber/matrix interface sliding stress in SCS-6/Timetal®21S. *Scripta Materialia*. 1999;40:529-35.
- [241] Ying Long, Athar Javed, Zhao-ke Chen, Xiang Xiong, Xiao P. Deposition Rate, Texture, and Mechanical Properties of SiC Coatings Produced by Chemical Vapor Deposition at Different Temperatures. *International Journal of Applied Ceramic Technology* 2013;10.
- [242] Mura T. *Micromechanics of Defects in Solids*. Dekker, Dordrecht, The Netherlands. 1987.
- [243] Luckhoo HT, Jun TS, Korsunsky AM. Inverse eigenstrain analysis of residual stresses in friction stir welds. *Procedia Engineering*. 2009;1:213-6.