# Dislocation Loop Evolution During In-situ Ion Irradiation of Model FeCrAl Alloys

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**Abstract:** Model FeCrAl alloys of Fe-10%Cr-5%Al, Fe-12%Cr-4.5%Al, Fe-15%Cr-4%Al, and Fe-18%Cr-3%Al (in wt %) were irradiated with 1 MeV $Kr^{++}$ ions in-situ with transmission electron microscopy to a dose of 2.5 displacements per atom (dpa) at 320°C. In all cases, the microstructural damage consisted of dislocation loops with ½⟨111⟩ and ⟨100⟩ Burgers vectors. The proportion of ½⟨111⟩ dislocation loops varied from ~50% in the Fe-10%Cr-5%Al model alloy and the Fe-18Cr%-3%Al model alloy to a peak of ~80% in the model Fe-15%Cr-4.5%Al alloy. The dislocation loop volume density increased with dose for all alloys and showed signs of approaching an upper limit. The total loop populations at 2.5 dpa had a slight (and possibly insignificant) decline as the chromium content was increased from 10 to 15 wt %, but the Fe-18%Cr-3%Al alloy had a dislocation loop population ~50% smaller than the other model alloys. The largest dislocation loops in each alloy had image sizes of close to 20 nm in the micrographs, and the median diameters for all alloys ranged from 6 to 8 nm. Nature analysis by the inside-outside method indicated most dislocation loops were interstitial type.

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## 1. Introduction

Iron-chromium-aluminium (FeCrAl) alloys are an emerging class of Fe-based, high-Cr ferritic alloys for use in nuclear power generation. In particular, FeCrAl alloys are promising candidates as a fuel cladding in commercial light water reactors (LWRs) that can exhibit enhanced accident tolerance compared with the zirconium-based alloys in use today [1,2]. The perceived accident tolerance of FeCrAl alloys is predicated on FeCrAl alloys exhibiting oxidation kinetics more than 100× slower than those of zirconium-based alloys in high-temperature steam environments (>1000°C) [2,3]. The slower oxidation kinetics lead to lower rates of heat and hydrogen production in the reactor core under accident scenarios, resulting in additional coping time for active mitigation strategies [2]. Although FeCrAl alloys' oxidation resistance lends itself to high performance during specific accident scenarios, such as loss-of-coolant accidents, FeCrAl alloys could be susceptible to in-core degradation during normal operation. Degradation could be the result of the harsh environment of commercial LWRs, including elevated pressures/stresses, high-temperature water (288–320°C, nominally), and neutron radiation. The combination of elevated temperature and neutron radiation is a concern, as it may lead to significant radiation-induced hardening and embrittlement effects. Significant embrittlement could ultimately limit the lifetime of FeCrAl alloys in LWR cladding applications.

Initial work on neutron-irradiated model FeCrAl alloys in a materials test reactor investigated the magnitude of radiation-induced hardening and embrittlement [4]. These studies, through combined ex-situ structure-property evaluations, have determined the hardening and loss of ductility in this alloy class to be in part due to precipitation and dislocation loop formation. The precipitation and dislocation loop formation can be directly linked to the radiation damage processes, including radiation-enhanced diffusion and defect agglomeration in these alloys.

The development and eventual optimization of FeCrAl alloys for nuclear applications would require detailed insight into these fundamental radiation damage processes and any subsequent defect production and evolution. Unfortunately, materials test reactor irradiations do not lend themselves to detailed studies of these fundamental processes owing to their limited capability for active control of experimental parameters. For example, the same region of interest cannot be studied under increasing dose or irradiation temperature; and it is well known that microstructural variables such as grain size, prior existence of dislocations, or other local defect sinks can locally alter the precipitation and dislocation loop formation in metals. Furthermore, in-reactor temperature control is inherently difficult; this was especially the case in previous studies of FeCrAl alloys [4] in which temperature control was passively determined by the sample holder–capsule diametrical gap. Finally, radiation dose and dose rate can vary greatly depending on the axial and/or radial location within the material test reactor core.

Ion irradiations have proven to show significantly better control of experimental variables, compared with materials test reactor irradiations, including irradiation dose, dose rate, and temperature. Additionally, in-situ ion irradiation performed within a transmission electron microscope (TEM) on thin foils enables the determination of dynamical processes within a single region of interest, eliminating (or characterising) grain-to-grain variations, among many other variables typical of ex-situ studies. This results in the capability to design experiments in which parameters can be selected and closely controlled, with only one varying during an experiment (e.g., total dose); an ideal situation for examining the fundamental aspects of radiation damage in a specific material system, including defect nucleation and growth. Additionally, the correlation of single-parameter (or at least reduced-parameter) in-situ ion irradiations with modern modelling and simulation techniques provides an opportunity to extend the value of such experiments and modelling.

Preliminary work has already been performed on correlative studies of fundamental radiation damage aspects in Fe and FeCr alloys; for example, the impact of microstructure [5], chemistry [6–12], irradiation temperature [13,14] and dose [8,9] on defect formation. In particular, the dislocation loop production and evolution kinetics have been evaluated. It has been shown that dislocation loops formed in Fe and FeCr during irradiation have Burgers vectors of ⟨100⟩ or ½⟨111⟩, in contrast to other body-centred-cubic (BCC) metals such as molybdenum, tungsten, and vanadium, which contain almost exclusively ½⟨111⟩ dislocation loops. Understanding the mechanisms behind the formation of ⟨100⟩ dislocation loops is important, as they are a more resistant barrier to dislocation slip than ½⟨111⟩ dislocation loops [4,15] and therefore have a greater potential for hardening. It has been shown experimentally [13] that the fraction of ⟨100⟩ dislocation loops is higher at higher temperatures; Dudarev et al. [14] showed theoretically how the stability of ⟨100⟩ and ½⟨111⟩ dislocation loops depends greatly on temperature, which helps to explain experimental observations of a decreasing fractional population of ½⟨111⟩ loops with increasing temperature in Fe [13]. Monte Carlo–based simulations by Xu et al. [16] of coalescing ½⟨111⟩ dislocation loops have shown that new ⟨100⟩ dislocation loops can emerge from such an interaction. Such a formation process would be heavily dependent on the frequency of loop coalescence, which in turn is highly dependent on dose, dose rate, temperature, composition, and impurity content. From a combination of thin foil [8] and bulk-material irradiations [7,17–19] of FeCr, it has been shown that the proportion of ⟨100⟩ dislocation loops after irradiation declines with increasing Cr content (corresponding to decreasing ½<111> loop mobility [16]).

This paper discusses an investigation of the details of radiation-induced dislocation loop formation and evolution in model FeCrAl alloys by using ion irradiation coupled with in-situ and ex-situ TEM. This work provides a fundamental insight into how composition affects nucleation and growth of dislocation loops in these alloys, which are primary contributors to radiation-induced hardening. Ultimately, design of a cladding alloy requires tailoring composition and microstructure to balance performance with respect to radiation hardening/embrittlement (both from loops and precipitates), oxidation/corrosion, high temperature mechanical properties, component fabricability, and neutronics. As such, this study directly informs design with regards to one of the many performance metrics for a candidate alloy. However, it should be noted, neutron irradiation studies [20] have shown α' phases to be the dominant hardening mechanism in these alloys, and hence although this study provides insight on the loop formation, it does not provide a full picture of the features contributing to hardening in the alloys.

Experimental conditions were specifically tailored to provide insight into the role of composition in dislocation loop formation and evaluation within the low-dose regime, thereby serving as a guide toward compositional control for radiation tolerance in FeCrAl alloys. A set of well-annealed model FeCrAl alloys with a yttrium addition, that reside near the kinetic boundary for passivation in 1200°C steam environments [1], was used. The absence of significant dislocation densities, internal defect sinks, and minor alloying elements, such as molybdenum and silicon, reduced the complexities in this study. A site-specific focused ion beam (FIB) lift-out technique was used to produce large thin areas of relatively uniform thickness (on the order of 25–100 $\mu m^2$) orientated with a ⟨100⟩ pole aligned normal to the foil surface. These samples were irradiated at the Intermediate Voltage Electron Microscopy (IVEM)–Tandem Facility (Argonne National Laboratory) with 1 MeV $Kr^{++}$ ions at 320°C up to damage doses of 2.5 displacements per atom (dpa). The temperature was selected to represent commercial reactor conditions for fuel cladding in LWRs. Although a temperature shift such as the one proposed by Mansur [21] could have been used to correlate between irradiations of different dose rates, it was not used here because of the absence of a strictly established dose-temperature shift for dislocation loop formation in FeCrAl alloys. The end dose of 2.5 dpa represents early-life doses expected for a typical commercial fuel cladding and was found to reside well within the coarsening regime of the dislocation loop evolution.

## 2. Experiment

Four model FeCrAl alloys with nominal compositions of Fe-10%Cr-5%Al, Fe-12%Cr-4.5%Al, Fe-15%Cr-4%Al, and Fe-18%Cr-3%Al (in wt %) with designations of F1C5AY, B125Y, B154Y-2, and B183Y-2 in [4,22–25] were studied. The full compositions, including impurity contents, are provided in Table 1. The alloys span a wide composition range of model FeCrAl alloys shown to be protective in high-temperature steam at up to 1200°C [1] and for which a wealth of base materials properties and radiation performance data exist [4,22–25]. The base feedstock for each alloy was fabricated by arc-melting and drop casting to prepare bar-shape ingots with a size of 13 × 25 × 125 mm. The as-cast ingots were homogenized at 1200°C for 2 hours in an argon gas atmosphere and then water-quenched. The ingots were hot-rolled at 700°C with a 10% thickness reduction per pass, for a total thickness reduction of more than 90%, and then annealed at 700°C for 1 h, followed by air cooling. Cold-rolling introduced a significant density of line dislocations ($6.3\pm1.0 \times 10^{13}$ m$^{-2}$ to $1.5\pm0.7 \times 10^{13}$ m$^{-2}$) [4]. The cold-rolling process was initially applied to flatten the sheet product and simulate the as-drawn condition of commercially produced FeCrAl cladding tube.

A high density of line dislocations was deemed undesirable for this study, as these could interfere with the formation and evolution of radiation-induced dislocation loops; hence, bulk samples underwent a final annealing stage for 30 minutes at 700°C. This annealing stage resulted in relatively defect-free grains with a line dislocation density so low that a statistically significant counting of them could not be completed. All alloys had grain sizes on the order of 20–50 µm. Therefore, it should be noted, in comparing this study with those already in the literature, that the as-received microstructures of the alloys in Table 1 differed considerably in dislocation density and grain size from those in references [4,21,23], although the chemistry remained identical. A very low density of Y-Fe intermetallic compounds have been observed in these model alloys (when Y is present). In some cases, these compounds could contain small amounts of dissolved Al, but this dispersion of larger precipitates does not greatly affect the bulk composition of the given model alloy [20]. The irradiated samples in this paper did not contain any such precipitates.

The as-annealed bulk specimens were prepared into lamellae for TEM investigation using site-specific FIB guided by electronic backscatter diffraction (EBSD) imaging.

This preparation technique was used to ensure electron-transparent samples could be produced with the sample orientated near a ⟨100⟩ pole, thereby reducing the tilting needed in the TEM for dislocation loop analysis. Preparation by jet electropolishing was undesirable due to the difficulty associated with imaging magnetic specimens by TEM. EBSD and FIB preparation were completed on a FEI Versa 3D Dual Beam scanning electron microscope (SEM)-FIB at the Low Activation Materials Development and Analysis (LAMDA) facility at Oak Ridge National Laboratory [26]. EBSD used an Oxford Instruments EBSD system at an accelerating voltage of 20 kV and a working distance between 12 and 17 mm. Data acquisition was not optimized for high resolution, as only the relative grain orientation of the large-grained material was needed.

FIB lamellae on the order of 20 × 2 × 10 μm were mounted onto a TEM support grid on one side, and two window-like areas ~3.5 × 7μm were thinned to electron transparency. The mounting and thinning geometries were selected to produce a structurally robust sample that would limit sample deformation at elevated temperatures during in-situ ion irradiation. All samples were cleaned with a final polishing using a $Ga^+$ beam at 2 kV to minimize FIB-induced radiation damage. Electron-transparent sections of the lamellae were between 50 and 80 nm thick. Micrographs of each alloy prior to irradiation are shown in Figure 1. The straight lines running diagonally in Figure 1b and more clearly in Figure 1c are FIB induced artefacts that could not be avoided here.

The foils were irradiated with 1 MeV $Kr^{++}$ ions in-situ with TEM at the IVEM-Tandem Facility at Argonne National Laboratory [27]. Each alloy was irradiated at

320°C up to a dose of 2.5 dpa (fluence of $1.5\times10^{14}$ ions $cm^{-2}$) as calibrated by SRIM [28] using the "quick calculation mode" as recommended by Stoller et al [29] and using a displacement energy of 40 eV [30]. The ion flux was maintained near $5\times10^{11}$ ions $cm^{-2}s^{-1}$ ($8.3\times10^{-4}$ dpa $s^{-1}$). In-situ TEM observations were performed using a Hitachi H-9000NAR TEM operating at 200 kV. The incident ion beam was 30° from the microscope axis and the lamella normal was oriented typically ~5° from the electron beam. In-situ videos were recorded with the foil tilted to a **g**=011 strong two-beam condition (with the deviation parameter, $s_g$, close to zero) using a Gatan 622 video camera operating at 15 fps. More extensive imaging was carried out at intermediate dose steps of 0.5, 1.5, and 2.5 dpa by tilting also to **g**=$01\bar{1}$, 020, and 002. Immediate dose step images were recorded using a Gatan Orius SC 1000 CCD. The Burgers vectors of dislocation loops were determined by **g·b** analysis of micrographs taken at 1.5 and 2.5 dpa. The dislocation loops present at 0.5 dpa were very small, weak in contrast, and few in number and were not analysed. The Burgers vectors of dislocation loops at 0.75, 1.0, and 2.0 dpa were determined by correlating loops with those in micrographs taken at 1.5 and 2.5 dpa. Additional ex-situ TEM analysis was conducted at the LAMDA facility using a JEOL 2100F TEM with a field emission gun operating at 200 kV, and at the University of Oxford using a JEOL 2100 TEM with a $LaB_6$ source, also operating at 200 kV.

Dislocation loop volume number densities were calculated using measurements of foil thickness using convergent beam electron diffraction (CBED) [31] and areal number densities measured from strong two-beam micrographs taken using **g**=011 and $01\bar{1}$. The areas analysed were of relatively uniform thickness, as indicated by energy-filtered

TEM thickness maps of the foils, which allow the region of interest to be treated as "cuboids".

Defects were counted by a combination of manual loop-by-loop counting and the automated counting package available in ImageJ, based on recommendations made in [32] and [33]. The same ~300×450 nm region was imaged at each dose, which at 2.5 dpa would contain 200–400 dislocation loops. Error bars were calculated by counting features that were difficult to designate as dislocation loops. These were counted as "maybes," and half of this count was added to the total. The error was then taken as half the number of "maybe" loops. This follows previous practice in [32] and [17].

The dislocation loops were designated as having ⟨100⟩ or ½⟨111⟩ type Burgers vectors based on $\mathbf{g}\cdot\mathbf{b}$ analysis at $\mathbf{g}$=011 and $\mathbf{g}=01\bar{1}$ in strong two-beam conditions. It was assumed that dislocation loops satisfying the $\mathbf{g}\cdot\mathbf{b}$=0 invisibility criterion would be entirely absent or too weak to be visible in the micrographs. Individual dislocation loops found to be present in both $\mathbf{g}$=011 and $01\bar{1}$ were designated ⟨100⟩, whereas loops unique to each image were designated as ½⟨111⟩ loops. It was reasoned that if all variants of ⟨100⟩ and ½⟨111⟩ dislocation loops are equally likely, the ⟨100⟩ loop population visible in $\mathbf{g}$=011 or $01\bar{1}$ is $(2x/3)$, where $x$ is the total ⟨100⟩ dislocation loop population. Similarly, the ½⟨111⟩ dislocation loop population visible in $\mathbf{g}$=011 or $01\bar{1}$ is $(y/2)$, where $y$ is the total ½⟨111⟩ dislocation loop population. The values of $x$ and $y$ were then derived from the counts of ⟨100⟩ and ½⟨111⟩ dislocation loops in micrographs taken at $\mathbf{g}$=011 and $01\bar{1}$.

## 3. Results

The FIB lift-outs all had a grain orientation with a ⟨100⟩ pole close to the surface normal, and so all analysis was conducted close to this pole. Figure 2 shows the in-situ irradiated microstructures of each alloy at different doses. All micrographs in this figure were recorded under strong two-beam conditions using a diffraction vector **g**=011. In all cases, the microstructure comprised small dislocation loops with few, if any, line dislocations. In all cases, dislocation loops were larger and more numerous with increasing dose. The dislocation loops appeared to be distributed randomly in the foil with no loop strings or finger loops observed.

### *3.1. Dislocation loop volume number density and morphology*

Figure 3a presents the dislocation loop volume number densities of ⟨100⟩ and ½⟨111⟩ dislocation loops in each alloy as a function of dose and Cr content, and Figure 3b indicates the percentage of ½⟨111⟩ dislocation loops as a function of composition and dose.

The results show that

- With increasing dose, the total dislocation loop number density increased. This trend was weaker at higher doses, where the loop populations showed signs of approaching an upper limit, indicating a saturation dose might apply.
- For a given dose (dpa), the total dislocation loop volume number density decreased with increasing Cr content. This trend was quite pronounced in comparing the 10% Cr with the 18% Cr alloys.
- The balance between dislocation loop types varied with Cr content. From 10 to 15 wt % Cr content, the ⟨100⟩ dislocation loop population decreased but the

½⟨111⟩ dislocation loop population showed little change. From 15 to 18 wt % Cr content, the ½⟨111⟩ dislocation loop population decreased significantly, whereas the ⟨100⟩ dislocation loop population decreased only slightly. Figure 3b shows the percentage of ½⟨111⟩ dislocation loops present is largest in Fe-15%Cr-4%Al. The possible causes of this finding are considered in the discussion.

- There was no clear trend with increasing dose for the proportion of ½⟨111⟩ and ⟨100⟩ dislocation loops.

*3.2. Dislocation loop dynamics*

The in-situ TEM allowed investigation into possible dynamic dislocation loop effects; results are qualitatively summarised in Table 2. Dislocation loops first became visible at around 2–3 nm in size between 0.1 and 0.2 dpa in all alloys except the Fe-18%Cr-3%Al model alloy, in which some loops were visible from 0 dpa, and had probably nucleated during FIB preparation. An example of dislocation loop nucleation induced by the ion beam is shown in Figure 4, in which the maximum grayvalue from the region of interest as a function of time is plotted, alongside successive 1/15 s video frames before and after nucleation. The dislocation loops appeared very stable in position throughout irradiation. Dislocation loop hopping was not observed, unlike in simpler Fe-Cr alloys or pure Fe, in which hopping has been seen to occur [17,18]; however, Yao et al. [17] reported seeing a decline in the frequency of dislocation loop hopping in FeCr as the Cr composition increased up to 18 wt % Cr.

Some dislocation loops were seen to jump ~1 nm only once in an <011> direction, unlike the back-and-forth hopping motion over several nanometres that has previously been reported in FeCr [10]. The jumps were often accompanied by a change in image size, which could indicate the absorption of a large cluster or smaller invisible dislocation loop, or the rotation of the habit plane. Dislocation loop loss to the foil surface was also seen (Figure 5); theoretical modeling of dislocation loops in Fe suggests such losses occur when dislocation loops glide to within 5 nm of the foil surface [12]. As these foils were oriented along [100], the dislocation loops lost are likely to have Burgers vectors of ½⟨111⟩, as the ⟨100⟩ loops either do not have a glide path in the direction of the foil surface (**b** = [010] and [001]) or are not present in the micrographs owing to the invisibility criterion (**b** = [100]).

Dislocation loop dynamics were investigated further by quantifying the number of loops seen to nucleate for each increment in dose, and the number of loops lost for each increment. Dislocation loop loss could be either by loss to the free surface or via coalescence. The observed nucleation and loss events are plotted in Figure 6 for dislocation loops observed in a **g**=011 condition.

The results show that

- The nucleation rate of dislocation loops increased between 0.75 and 1.0 dpa for all materials. Although number densities were not recorded below 0.75 dpa, the observation of an increased nucleation rate at that level implies that below ~1.0 dpa, dislocation loops in the alloys are still in a nucleation-dominant regime.
- Above 1.0 dpa, the nucleation rate decreased for all materials, although the nucleation rate within Fe-12%Cr-4.5%Al increased again between 1.5 and 2.0 dpa. This is unusual and is most likely an experimental error due to poor image

contrast. A decrease in dislocation loop nucleation rate indicates that in this dose range, loop growth is favoured over loop nucleation.

- Dislocation loop loss rate increased for all alloys with dose and was small in comparison with the nucleation rate. Given that the foil surface had a normal pointing along a $\langle 100 \rangle$ zone axis, the visible dislocation loops being lost to the foil surface were most likely to be $\frac{1}{2}\langle 111 \rangle$, since they have a component of glide in the direction of the foil surface.

*3.3. Dislocation loop sizes*

Quantitative analysis of dislocation loop sizes was performed using images taken using strong two-beam conditions at $\mathbf{g}$=011 and $01\bar{1}$. Using the same method for counting described in [15], the dislocation loop sizes were calculated by measuring the major diameter of an elliptical envelope around each loop image. The measured dislocation loop size depended on the threshold value used to produce the elliptical selections. The error was estimated by changing the threshold value by plus or minus the standard deviation of the background intensity, and measuring the resultant change in dislocation loop size. This method indicated an individual dislocation loop image size error of around ±1.5 nm. Dislocation loops less than 4 nm in diameter were measured manually and were much more difficult to size accurately because of their typically weak contrast against the background fluctuations. In strong two-beam conditions, inside-outside contrast should not be significant; but the dislocation loop image will appear larger than the true size of the dislocation loop [34]. Weak beam imaging was attempted to estimate this bias, but it was made difficult by a large population of surface oxides; therefore, the data presented merely refer to the projected loop image sizes. Figure 7a shows kernel density estimations (KDEs) of the dislocation loop sizes in each alloy and at each dose, with box plots overlaid indicating the median and 5th, 25th, 75th, and 95th percentiles. The KDE was calculated in Python using SciPy [35] which uses the "Scotts" rule bandwidth estimator to calculate the probability density function of the distribution. The median is preferred to the mean as a measurement of size, as it is unaffected by outlying data points, and as the distribution is not symmetrical. Figure 7b shows the ratio of median ⟨100⟩ size to median ½⟨111⟩ size.

The results show that

- Dislocation loop size generally increased with increasing dose. The dislocation loop image size distributions for each alloy flattened out over a wider range with increasing dose as the loops grew and the population increased. The median image size generally grew with increasing dose.
- Dislocation loops in Fe-10%Cr-5%Al coarsened more quickly than in the other alloys; but growth rapidly declined above 1.0 dpa, as indicated by the small changes in the 5th, 25th, 50th, 75th, and 95th percentiles from 1.0 to 2.5 dpa. Dislocation loops in the other alloys showed continued growth up to the final dose of 2.5 dpa.
- Above 1.0 dpa, the difference between the median sizes of ⟨100⟩ and ½⟨111⟩ dislocation loops increased, implying preferential growth of ⟨100⟩ loops. There is no clear compositional effect represented in the data for this case.

The number of point defects within each dislocation loop, N(d), was estimated by assuming the major diameter, *d*, of every dislocation loop is equal to the diameter of a circular dislocation loop with either **b**=⟨100⟩ or ½⟨111⟩. Hence,

$N_{\{100\}}(\mathrm{d}) = \pi(d/2)^2(1/a_0{}^2)$ and $N_{\{111\}}(\mathrm{d}) = \pi(\mathrm{d}/2)^2(1/\sqrt{3}a_0{}^2)$,

where $a_0$ is the lattice parameter for iron; and the dislocation loops are treated as lying purely on {100} or {111} planes (though this is not necessarily the case). The observed dislocation loops were then grouped into 1<N<100, 101<N<200, etc. Figure 8 shows the distribution of the dislocation loop population as a function of dislocation loop defect content. The histograms are normalised to the total dislocation loop density so that the area under the plotted data is equal to the dislocation loop volume number density. Figure 6 makes for a useful comparison with similar plots produced from modelling defect evolution, in [36] for example; but it should be stressed that, because of the imaging conditions, the results presented here are likely to overestimate the true values of N(d). For ½⟨111⟩ loops retaining shear components, the estimation of N(d) is below the true value because the packing density of atoms on a {111} plane is smaller than the packing density for atoms lying closer to a {110} nucleation plane.

The results show that

- For all alloys besides Fe-10%Cr-5%Al, the frequency of small dislocation loops (<100 atoms) seemed to decrease with increasing dose. This supports the implications of Figure 5, which suggests that the nucleation rate decreases with increasing dose, and so the frequency of small dislocation loops should decrease with dose. This trend was not clear for Fe-10%Cr-5%Al, which contained similar densities of dislocation loops with <200 atoms for each dose.
- Dislocation loops containing >200 atoms generally decreased in frequency as their sizes increased.

*3.4. Dislocation loop nature*

The interstitial/vacancy nature of dislocation loops can be determined from their inside-outside contrast behaviour [34]. Inside-outside contrast analysis was performed after irradiation on the Fe-18Cr-6Al alloy by imaging under kinematical two-beam conditions with $s_g$>0. Nature analysis of other alloys was attempted, but the high dislocation loop density made the correlation of loops among all imaging conditions impossible. Weak beam imaging was attempted but did not produce usable images owing to overwhelming background noise from the charge-coupled device camera and surface oxide contaminants. In kinematical bright field conditions, the technique is less reliable for dislocation loops <30 nm [34]. Images of the same region were taken at **g** = ±020, ±002, ±011, $\pm 01\bar{1}$, ±121, ±112, $\pm 1\bar{1}2$, and $1\bar{3}2$, using the [100], $[20\bar{1}]$ and $[3\bar{1}\bar{1}]$ poles, with the direction of the electron beam designated as [100]. Care was taken to ensure that the deviation parameter was kept positive, the indexing of the diffraction pattern was consistent, and the image and diffraction pattern were properly aligned.

Inside-outside contrast analysis was conducted on ½⟨111⟩ dislocation loops with the assumption that they could be treated as pure edge dislocation loops. Inside-outside contrast for ⟨100⟩ loops was taken from images at **g**=±112, where the loops were tilted ~18$^{o}$ from edge-on. Interstitials are much more mobile than vacancies [37], and so interstitial dislocation loops typically grow faster than vacancy dislocation loops. Since the analysis here was confined to the largest loops visible, there was an inherent and possibly overwhelming bias in the analysis toward interstitial type dislocation loops. Figure 9 shows an example of the analysis of ⟨100⟩ dislocation loops. Analysis of the ½⟨111⟩ loops can be found in the supplementary material.

Of the 26 dislocation loops analysed, inside-outside contrast indicated that

- Thirteen ⟨100⟩ dislocation loops were interstitial-type.
- Four ½⟨111⟩ dislocation loops were interstitial-type and four ½⟨111⟩ dislocation loops were vacancy-type.
- Five dislocation loops could not be conclusively analysed.

Vacancy dislocation loops are rarely observed in ferritic systems because of the low mobility of vacancies and the preference for vacancies to congregate as voids [12]. The observation of vacancy ½⟨111⟩ dislocation loops is therefore unusual. However, it remains possible that these loops were not pure edge, but had shear components with habit planes close to or on {110} nucleation planes. In this case, interstitial dislocation loops can show vacancy-like contrast [34]. Fresnel contrast revealed void-like structures <5 nm in diameter distributed randomly throughout the foils for all alloys; however, it could not be verified that these were not merely oxides on the sample surface, and so they were not analysed.

## 4. Discussion

### *4.1. The role of irradiation dose*

Within all the FeCrAl alloys for doses up to 1.0 dpa, dislocation loop number densities increased with dose at an increasing rate, and new loops continued to nucleate up to the final dose of 2.5 dpa. However, at doses above 1.0 dpa, the nucleation rate generally decreased as the dose increased. These observations are consistent with previous irradiation experiments on Fe and FeCr [8], and even on other BCC metals such as tungsten [33]. They can be explained by considering the competing processes of dislocation loop nucleation and annihilation. At low doses, the dislocation loop density, and hence the sink density, is low. This means that defect clusters produced after displacement cascades are more likely to nucleate as dislocation loops than to annihilate with an already existing defect sink or cluster of opposite nature. At higher doses, many small defect clusters are already present in the foil from previous cascades, and so new clusters are likely to coalesce with other clusters. Larger interstitial clusters are more stable as dislocation loops [38], rather than other configurations such as crowdions. Therefore, the dislocation loop nucleation rate increases with increasing dose.

When the dislocation loop population reaches some threshold, defect clusters generated by irradiation will more likely be close to already nucleated dislocation loops. Thus, clusters will be more likely to coalesce with such loops than with other defect clusters, and hence the dislocation loop nucleation rate will decrease. This marks a transition from a nucleation-dominant regime to a coarsening-dominant regime. Figure 10 illustrates this concept for nucleation and coarsening-dominant scenarios. As dislocation loops grow, each will become an increasingly larger sink for defects, so dislocation loop nucleation rate should continue to drop until saturation is reached.

As the dose increased, so did the sizes of dislocation loops in all alloys. This was to be expected based on the observations of nucleation rate. As the dislocation loop density increases, new defect clusters are increasingly more likely to coalesce with an already existing defect, such as a dislocation loop or cluster. Existing defects may either shrink or grow depending on whether a coalescing defect is similar or opposite in nature.

As dislocation loops grow, so does the volume encompassed in projection of their glide path, as illustrated by Figure 11. They will therefore intersect a larger proportion of smaller defects as they glide through the foil, and their motion will be restricted as a result. That means dislocation loop growth will be dominated by the movement of small and highly mobile interstitial dislocation loops and clusters, rather than by the merging of pairs of larger dislocation loops. The tendency toward saturation of the dislocation loop population [39], and the low observed mobility of dislocation loops, support the argument that loop coalescence with small dislocation loops and defect clusters is the dominant dislocation loop growth mechanism, as opposed to growth by cascade overlap. Owing to the higher mobility of interstitials, this process will also favour interstitial dislocation loop growth and inhibit vacancy dislocation loop growth. Most dislocation loops identified were of the interstitial type, and even those identified as vacancy-type may simply be interstitial-type dislocation loops with large shear components. This result seems to be in general agreement with other findings in FeCr [18]. While some small vacancy dislocation loops may have formed from vacancy-rich regions, the most likely fate of vacancies is to form voids, which are the most stable vacancy defect in BCC Fe [12].

The absence of dislocation loop hopping in the in-situ videos in all alloys indicates that dislocation loop movement is being supressed by pinning points throughout the matrix. This could be due to the high Cr content, the addition of Al, an unobserved quantity of small (<2 nm) voids and vacancy clusters, or a combination of all of these.

It has been shown here that ⟨100⟩ dislocation loops coarsen faster than ½⟨111⟩ loops. In a study of BCC Fe, Xu et al. [16] showed via the kinetic Monte Carlo method that two small ½⟨111⟩ dislocation loops can coalesce to form one larger ⟨100⟩ dislocation loop or one larger ½⟨111⟩ dislocation loop. A larger average size of ⟨100⟩ dislocation loops is therefore an indication that growth by absorption of small clusters is more prominent than growth by loop-loop coalescence. That would mean the population is biased toward the ½⟨111⟩ type, since coalescence is rare; but the loops that do coalesce to form ⟨100⟩ dislocation loops would then have a size advantage early on in their evolution over the average size of the ½⟨111⟩ population.

There was no observed trend with increasing dose in the ratio of ⟨100⟩ to ½⟨111⟩ dislocation loops. It could be argued that as the dislocation loop population increases, the frequency of ½⟨111⟩ dislocation loop coalescence should increase, and hence the proportion of ⟨100⟩ dislocation loops should increase. However, at higher doses, dislocation loops would be larger, and would be less mobile than smaller dislocation loops and clusters, so coalescence of large loops would be a rarer occurrence. As was discussed earlier, this finding also means growth would be dominated by the absorption of smaller dislocation loops and clusters. Therefore, the rate of ⟨100⟩ dislocation loop formation should decrease at higher doses because most defects would be absorbed by the existing larger dislocation loops.

The simulations by Xu et al. [16] only demonstrate ⟨100⟩ formation from the coalescence of identically sized ½⟨111⟩ loops and at a single intersection point; they did not address what the resultant loop Burgers vectors might be from the coalescence of ½⟨111⟩ loops of different sizes, and from intersections at various points within the loop. Therefore, it may be presumptuous to expect this to be the dominant ⟨100⟩ formation mechanism observed here.

### *4.2. The role of composition*

Dislocation loop density was found to be slightly higher in the lower-Cr alloys, with Fe-18%Cr-3%Al having the fewest dislocation loops. The proportion of dislocation loops with **b**=⟨100⟩ fell, on average, with increasing Cr content. The proportion of ½⟨111⟩ and ⟨100⟩ dislocation loops is important because ⟨100⟩ dislocation loops are highly immobile compared with ½⟨111⟩ dislocation loops [16]. They act as stronger barriers to dislocation motion than ½⟨111⟩ dislocation loops and hence have a larger potential for hardening [4,15].

Previous thin foil irradiation experiments on FeCr [8] suggest that the total dislocation loop density in Fe and FeCr binary alloys does not change substantially with Cr content, and that the average sizes are smaller in higher-Cr alloys. But in FeCr alloys irradiated before thinning to electron transparency ("bulk irradiated" specimens), it was found that the dislocation loop densities are higher and dislocation loop sizes smaller in high-Cr than in low-Cr alloys, [17,19,40]. It has also been reported that in thin foil [8] and bulk [7,17–19] ion-irradiated alloys, the ⟨100⟩ population is larger in Fe than FeCr, and there is a trend of an increasing proportion of ½⟨111⟩ dislocation loops with increasing Cr content. The suggested explanation [8] for this trend in thin foils is that more ½⟨111⟩ dislocation loops are lost to the foil surface via glide in lower-Cr alloys. However, the same trend has been seen in bulk irradiations [7,17–19], which suggests that a reduction in defect mobility with increasing Cr content in FeCr alloys [41] leads to a reduction in ⟨100⟩ dislocation loop formation. This is supported by the simulations by Xu et al. [16] of ⟨100⟩ dislocation loop formation from ½⟨111⟩ dislocation loop coalescence.

The majority of dislocation loops were found to be ½⟨111⟩ for all the alloys examined here at all doses. This finding is different from what was reported in the room-temperature in-situ ion-irradiated Fe-9%Cr and Fe-18%Cr binary alloys studied by Yao

[8], in which the majority of dislocation loops were found to be ⟨100⟩. Since the higher irradiation temperature of the FeCrAl alloys should favour ⟨100⟩ formation [14], the larger proportion of ½⟨111⟩ dislocation loops in these alloys could be attributed to the Al content or differences in impurities.

The fraction of ½⟨111⟩ dislocation loops present was largest in the Fe-15%Cr-4%Al alloy and smallest in Fe-10%Cr-5%Al. It would be expected that the highest-Cr alloy would have the largest fraction of ½⟨111⟩ dislocation loops. However, there was a small decline in the proportion of ½⟨111⟩ dislocation loops from Fe-15%Cr-4%Al to Fe-18%Cr-3%Al. The Fe-15%Cr-5%Al sample initially had very distinguishable grooves along the surface, as shown in the unirradiated micrograph (Figure 1). These FIB induced features on the surface may have altered how the ½⟨111⟩ loops interact with the free surface, and could have constrained ½⟨111⟩ loop loss, leading to their growth. The difference could also be related to the change in Al content, although the mechanism is unclear. In these alloys, as the Cr content increased, the Al content decreased, so it is difficult to say with any confidence what the real effect of Al was. Al remains dissolved in the matrix under irradiation and hence could have an effect on the irradiated microstructure [20], but due to the variances in the Al content, a distinct designation on the role of Al could not be confirmed. Since this change in Al remains small compared to that of Cr for the alloys, one would expect that if the magnitude of the effects of each element were roughly the same, the majority of the effect seen in our study would be due to Cr changes between the alloys.

A reduction in defect mobility should also, via a reduction in loop coalescence, lead to an increase in dislocation loop density and to smaller dislocation loops; but that is not what was observed. The Fe-18%Cr-3%Al alloy had the lowest dislocation loop population of the alloys, but the median dislocation loop size was ~8 nm at 2.5 dpa and was not significantly different from that in the other alloys. It is possible that there existed a large population of smaller dislocation loops that were not resolvable in our micrographs, which would reduce the average dislocation loop size and increase the overall population observed. This explanation seems likely, considering previous studies of defect evolution in FeCr alloys [8] showing that increasing Cr content leads to an increase in dislocation loop population and a reduction in average dislocation loop size. The majority of the errors in dislocation loop counts were from loops <5 nm in size, supporting the idea that there could be a large population of smaller dislocation loops hidden in the background noise of the micrographs. In a typical micrograph, a dislocation loop with strong contrast is easy to identify; but other features are much more difficult to identify as possible dislocation loops, noise, oxides, or sample preparation artefacts. In practice, identifying weak-contrast dislocation loops was subjective, as automated counting via thresholding was not adequate here for distinguishing the smallest and weakest dislocation loops from the background.

Field et al. [4] investigated the same alloys irradiated with neutrons to 1.8 dpa at 382°C and found dislocation loops 25–30 nm in diameter, of which 70–80% had **b**=½⟨111⟩. In comparison, the dislocation loops reported here were ~6–8 nm in diameter on average and 50–80% had **b**=½⟨111⟩. There are multiple possible contributing factors to the differences observed. These differences could be attributed mainly to the dose rates being a factor of ~$10^3$ higher in this study, which meant defects had less time to develop, agglomerate, and grow; and to dislocation loop loss to the surface, which meant a significant proportion of the ½⟨111⟩ dislocation loop population would be lost from the foil as a result of glide in the direction of the ⟨100⟩-oriented foil surface. In addition to these potential causes, the higher temperature of the neutron irradiation would favour ⟨100⟩ formation [13,14]. The alloys studied by Field et al. were not annealed like the alloys used for this study; the neutron-irradiated cold-worked FeCrAl alloys would contain a rich density of dislocations, which act as local sinks for radiation-induced defects. All of these factors impact loop growth, loop-loop interactions, and the formation of ⟨100⟩ loops.

## 5. Summary

The evolution of radiation damage under ion irradiation in thin foils of Fe-10%Cr-5%Al, Fe-12%Cr-4.5%Al, Fe-15%Cr-4%Al, and Fe-18%Cr-3%Al model FeCrAl alloys was investigated, using $Kr^{++}$ irradiation in-situ with TEM at doses up to 2.5 dpa ($1.4\times10^{11}$ ions $cm^{-2}$) at 320°C.

1. For all alloys, the dislocation loop population increased up to a maximum dose of 2.5 dpa between ~$7\times10^{15}$ $cm^{-3}$ in Fe-10%Cr-5%Al and ~$3\times10^{15}$ $cm^{-3}$ in Fe-18%Cr-3%Al. The increase in loop population slowed at higher doses for all alloys, which implies the defect population could have been approaching some upper limit.
2. At each dose, there was a slight trend of decreasing dislocation loop number density from Fe-10%Cr-5%Al to Fe-15%Cr-9%Al, but there was a much lower dislocation loop density in Fe-18%Cr-3%Al. At 2.5 dpa, the dislocation loop population decreased slightly from ~$7\times10^{15}$ $cm^{-3}$ to ~$5.5\times10^{15}$ $cm^{-3}$ as the Cr content was increased from 10 to 15%, and then decreased by a factor of ~0.5 from 15%Cr to 18%Cr to ~$3\times10^{15}$ $cm^{-3}$.

3. Dislocation loops with Burgers vectors of ⟨100⟩ coexisted with ½⟨111⟩ dislocation loops in all alloys. The proportion of ½⟨111⟩ dislocation loops was ~50% in Fe-10%Cr-5%Al and increased with increasing Cr content, although the largest proportion was found in Fe-15%Cr-4%Al with a population of ~80% ½⟨111⟩ dislocation loops. This result implies a reduction in the frequency of ⟨100⟩ formation with increasing Cr content. The cause of a small increase in the proportion of ⟨100⟩ dislocation loops from Fe-15%Cr-4%Al to Fe-18%Cr-3%Al is unclear; it could be statistically insignificant, but it may be influenced by the change in Al content.
4. The diameter of the largest dislocation loop images was ~20 nm. The median diameter of loops was ~6 nm at 2.5 dpa in Fe-10%Cr-5%Al and ~8 nm in Fe-12%Cr-9Al and Fe-15%Cr-8Al. Dislocation loops grew most quickly with increasing dose in Fe-10%Cr-5%Al, but the median dislocation loop diameter did not change significantly above 1.0 dpa. The median dislocation loop diameter in all other alloys increased with dose up to the final dose of 2.5 dpa.
5. The ⟨100⟩ dislocation loops grew faster than ½⟨111⟩ dislocation loops in all materials. The ratio of ⟨100⟩ to ½⟨111⟩ median dislocation loop size increased with dose, but there was no clear compositional effect.
6. The rate of dislocation loop nucleation peaked at ~1 dpa, indicating a transition from a nucleation-dominated to coarsening-dominated regime. The rate of dislocation loop loss to the foil surface was small compared with the rate of nucleation. Losses to the foil surface were more frequent at higher doses.
7. Nature analysis was conducted by the inside-outside method on the Fe-18%Cr-3%Al alloy; it was found that all of the ⟨100⟩ dislocation loops were interstitial,

whereas 50% of the ½⟨111⟩ dislocation loops had vacancy-like contrast. It remains possible that the dislocation loops identified as vacancy-type retained habit planes close to {110}, in which case interstitial loops can show vacancy-like contrast (or vice versa).

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**Figures**

Figure 1: Typical unirradiated condition of (a) Fe-10%Cr-5%Al, (b) Fe-12%Cr-4%Al, (c) Fe-15%Cr-4%Al and (d) Fe-18%Cr-3%Al, each imaged in a strong two-beam condition with g=011.

Figure 2: TEM micrographs taken at g=011 (marked as an arrow on image) as a function of dose and composition. The thickness is indicated for each alloy as measured by CBED, with an error of ±5%.

Figure 3: (a) Loop volume number density as a function of dose. Time indicated is the time the ion beam was on the sample. 1 dpa = $5.75\times10^{14}$ ions $cm^{-2}$. For clarity, data points have been shifted by ±0.05 dpa on the dose axis to separate data points for each Burgers vector. (b) and (c) Percentage of ½⟨111⟩ loops as a function of composition and dose. For clarity, data points have been shifted on the composition and dose axes to separate data points.

Figure 4: Maximum signal as a function of time and successive 1/15 s frames of the same loop nucleating in Fe-10%Cr-5%Al at ~0.2 dpa.

Figure 5: Example of loops being lost from the foil to the surface in Fe-10%Cr-5%Al between 2.3 and 2.4 dpa.

Figure 6: Loop nucleation rate and loss rate for each alloy. For the purpose of clarity, data points have been shifted by ±0.05 dpa on the dose axis so as to separate data points for gains and losses. Error bars are calculated as $\sqrt{\boldsymbol{N}}$, where N=number of loops counted.

Figure 7: (a) Loop image sizes as a function of dose for each Burgers vector and composition. The size distribution is presented as a box plot with a KDE overlaid for each plot. (b) The ratio of ⟨100⟩ to ½⟨111⟩ median loop size as a function of dose. For clarity, data points have been shifted by ±0.05 dpa on the dose axis to separate data points for alloys.

Figure 8: Loop population estimation as a function of the loop size, N, where N=number of atoms contained within a loop. To assist clarity, data points have been shifted on the size axis to separate data points. Error bars were calculated from the square root of the number of loops counted in each bin.

Figure 9: (a)–(d) Images of ⟨100⟩ loops at different imaging conditions. (a) and (b) are each composite images of ±g, where g=002 (a) and 020 (b). This is done to reduce background contrast. (c) and (d) are −g and +g, respectively , where g = 112, imaged close to [$\mathbf{3\bar{1}\bar{1}}$]. In this condition, [001] and [010] point toward the oncoming electron beam. For an interstitial loop, the Burgers vector is defined (here) as pointing toward the oncoming electron beam. Inside-outside contrast tells us (g·n)·$s_g$ >0 is true when contrast is "outside" and (g·n)·$s_g$<0 is true when contrast is "inside." Contrast of all indicated loops is outside when g=112 and inside when g=$\mathbf{\bar{1}\bar{1}\bar{2}}$. For this to be true, the Burgers vectors of the indicated loops all must be either +[010] or +[001], and so they must be interstitial.

Figure 10: Illustration of defect tendencies in nucleation- and coarsening-dominant regimes.

Figure 11: Illustration of the glide path of a large loop and small loop.

## Tables

| Alloy | Heat ID | Composition (wt %) | | | | | |
|---|---|---|---|---|---|---|---|
| | | Fe | Cr | Al | Y | C | S |
| **Fe-10Cr-4.8Al** | F1C5AY | 85.15 | 10.01 | 4.78 | 0.038 | 0.005 | 0.001 |
| **Fe-12Cr-4.4Al** | B125Y | 83.56 | 11.96 | 4.42 | 0.027 | 0.005 | 0.0013 |
| **Fe-15Cr-3.9Al** | B154Y-2 | 80.99 | 15.03 | 3.92 | 0.035 | 0.005 | 0.0004 |
| **Fe-18Cr-2.9Al** | B183Y-2 | 79.52 | 17.51 | 2.93 | 0.017 | 0.005 | 0.0006 |
| | | | **O** | **N** | **P** | **Si** | **Mo** |
| **Fe-10Cr-4.8Al** | F1C5AY | | 0.0013 | 0.0003 | 0.006 | <0.01 | <0.01 |
| **Fe-12Cr-4.4Al** | B125Y | | 0.0017 | 0.0009 | 0 | 0.01 | <0.01 |
| **Fe-15Cr-3.9Al** | B154Y-2 | | 0.0025 | 0.0007 | <0.002 | 0.01 | <0.01 |
| **Fe-18Cr-2.9Al** | B183Y-2 | | 0.0015 | 0.0011 | <0.002 | <0.01 | <0.01 |

Table 1: Summary of FeCrAl alloy compositions (in wt %) used in this study

| Dynamic effect | Description | Observations |
|---|---|---|
| **Nucleation** | Formation of new loops from planar clusters of point defects | Loops mostly appeared quickly within a 1/15 s frame. Other times they appeared gradually, likely owing to poor contrast regions of the captured video |
| **Movement** | Migration of the loop via glide and/or slip | Slight movement when small. Not seen in loops larger than ~ 5nm besides surface losses. Loops did not show signs of string alignment |
| **Hopping** | Rapid one-dimensional jump from one position to another | Not seen. Loops were observed to hop only once. Hopping distance typically <4 nm within a 1/15 s frame |
| **Growth (i)** | Growth of individual loops via absorption of mobile clusters and point defects | Presumed to be the dominant growth mechanism |
| **Growth (ii)** | Growth via merging of two or more dislocation loops | Rare event. <10 examples observed throughout the irradiation of each alloy |

| | | |
|---|---|---|
| **Loss to free surface** | Free surface of the thin foil acts as a large sink for loops and freely migrating defects | Dominant mechanism for loop loss, but still an infrequent observation in comparison with nucleation |

Table 2: Qualitative summary of the dynamic mechanisms observed in the in-situ video