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## On the Origins of Tin-Opacified Ceramic Glazes: New Evidence from Early Islamic Egypt, the Levant, Mesopotamia, Iran, and Central Asia

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### Abstract

It has been generally accepted that the beginnings of tin-based opacification of ceramic glazes is associated with the white glazed wares excavated in Iraq and western Iran and dated to the ninth century AD (so-called ‘Samarra-type’ pottery). This paper focuses on an earlier stage in the technological development of tin-opacified glazes, that is, the yellow and white glazed wares produced from the eighth century AD in Egypt and the Levant. In addition, the compositional data was extended for the subsequent spread of tin-opacified glazes into Mesopotamia in the ninth century and Northern Iran and Central Asia in the tenth century. Using SEM-EDS and SR-Micro-XRD, the chemical composition and microstructure of eighty-five samples of opaque yellow and white glazed wares from Egypt (Fustat), the Levant (Madaba, Aqaba, Al-Mina and Raqqa), Mesopotamia (Samarra, Kish, Basra and Susa), Northern Iran (Takht-i Suleiman), and Central Asia (Nishapur, Merv and Samarkand) were investigated. These data confirmed that the yellow and white glazes were opacified by lead-tin-oxide ( $\text{PbSnO}_3$ ) and tin oxide ( $\text{SnO}_2$ ) particles respectively. Replication experiments were then conducted to imitate typical compositions of the analysed opaque yellow and opaque white glazes. Overall, the results posited the beginning of tin-based opacification of glazes in the eighth century in Egypt and the Levant, and provided explanations as to how the production of opaque yellow and white glazes in the Levant and Mesopotamia might have been technologically linked.

**Keywords:** Glazed ceramics; Opacification; Technological Transfer; Tin oxide; Lead-tin-oxide; Calx

## 1. Introduction

The beginning of the Islamic period brought about stylistic and technological revolutions in the production of glazed ceramics. During the preceding Parthian and Sassanian periods in Iraq and Iran, the range of glazing techniques was limited to transparent and semi-opaque monochrome green or turquoise glazes of alkali-lime type (Hedges and Moorey 1975; McCarthy 1996; Hill *et al.* 2004; Pace *et al.* 2008). Likewise, in the Early Byzantine period in the Eastern Mediterranean, transparent glazes of high lead type, mostly in green, orange/yellow or colourless, were used to cover vessels (Rice 1930; Armstrong *et al.* 1997; Vroom 2004).

Archaeological excavations since the early twentieth century have yielded evidence of the first phase of Islamic glazed ceramics in Egypt, Jordan, Palestine and Syria from as early as the eighth century, namely Coptic Glazed ware (CGW), Hijazi ware and Cream ware (Whitcomb 1989); and the further developments in the eighth/ninth centuries, known as the Yellow Glaze Family (YGF) (Watson 1999). Despite their significance, the mention of these wares hardly goes beyond archaeological reports and classifications, and they remain largely unexplored. In fact, these wares have been overshadowed by the results of excavations in Samarra, Iraq, during 1912-13. The Samarra ceramic findings, published by Sarre (1925), had an enlightening, yet somewhat blinding, impact on research in the field of Islamic ceramics. A diverse range of exceptionally fine ceramics covered by opaque white glazes and influenced, in shape and decoration, by imported Chinese Tang porcelain and stoneware were found in the Abbasid capital city of Samarra, all dated to the ninth century AD. As a result, it became widely assumed that Islamic ‘fine’ glazed pottery was invented first in Iraq or western Iran in the ninth century, under the influence of imported Chinese wares, and from there it was spread to east and west. Although such an assumption brought new

interest into the field, an adverse consequence was that it relegated to a peripheral position other types of early Islamic glazed ceramics.

In a critical review, Watson (2014) highlighted the significance of eighth-century glazed ceramic findings from Egypt and the Levant and suggested that these wares, rather than the Samarra pottery, form the first chapter in the history of Islamic glazed ceramics and demonstrate the development of a pre-ninth-century market for fine ceramics in the Near East. Following upon this, Tite *et al.* (2015) conducted a series of preliminary chemical analysis using SEM-EDS, hand-held XRF, and PIXE on early Islamic opaque yellow glazed wares from Egypt, Syria, Iraq and Iran demonstrating that the yellow glazes were opacified by lead-tin-oxide ( $\text{PbSnO}_3$ ) particles. The study also suggested that the origins of tin-based opacification of yellow glazes in the early Islamic period may lie in early Byzantine glass making practices which also used lead-tin-oxide particles to opacify and colour yellow glasses, particularly in the form of mosaic tesserae (see also, Freestone *et al.* 1990; Mass *et al.* 1998; Heck and Hoffmann 2002; Heck *et al.* 2003; Marii 2012; Neri *et al.* 2012).

The first aim of this paper is to provide an extended set of chemical compositional data of both opaque yellow and opaque white Islamic glazed wares from the eastern Mediterranean, across to Mesopotamia, and into Iran and beyond. These data include, for the first time, a comprehensive dataset for glazed ceramics from Egypt (Fustat) and the Levant (Madaba, Aqaba, Raqqa, Al-Mina); an extension of previously published data (Kleinmann 1986; Mason and Tite 1997) for glazed ceramics from Mesopotamia (Samarra, Kish, Susa, Basra); and, in an Appendix, data for a small number of ceramics from Northern Iran (Takht-i Suleiman) and Central Asia (Nishapur, Merv, Samarqand) which provide preliminary evidence of the further spread of this opaque glaze technology (Fig. 1). The glazes and bodies of the ceramic were both studied using SEM-EDS, but

the bodies were not subjected to either thin-section petrology or trace element analysis (e.g., ICP-MS), in part because the former would have required significantly larger samples, and the latter, would have required samples from many more ceramics from each context for statistical treatment. Therefore, the tentative separation between locally produced and imported ceramics was made on the basis of the major and minor element compositions of the bodies as determined by EDS, and the body textures and distinctive inclusions as observed in the SEM.

The second aim of this paper is to integrate the chemical compositional data for the glazes with primary textual accounts of the glaze production processes, and thus, replicate the opaque yellow and opaque white glazed wares in the laboratory. According to accounts by Abu'l Qasim Kashani in 1301 (Allan 1973, 113; Afshar 2006, 342-3) and Isfahani in the nineteenth century (1888, 4, 9), the production of the opaque glazes would take place in two stages. First, a mixture of lead and tin was calcined (oxidised) by heat to produce the 'calx' powder. The ratios of Pb/Sn is stated to be around 16 for opaque yellow glazes (Isfahani 1888, 9) and between 1.5 and 4 for opaque white glazes (Allan 1973, 113; Afshar 2006, 342-343; Isfahani 1888, 4). In the second stage, to produce the opaque yellow glaze, the calx powder was mixed with a source of silica (e.g., quartz) and heated (Isfahani 1888, 9). To produce the opaque white glaze, alkaline frit was also added to the mixture of calx and silica and then again heated (Isfahani 1888, 3-4; Allan 1973, 113; Afshar 2006, 342-343).

The combination of the analytical data from the study of Islamic glazed ceramics and from the replicated samples that we present in this paper suggest that there are critical technological links between the production processes of tin-opacified yellow and white glazes. In particular, our study

provides evidence that sheds light on the processes of invention and development of these glazing techniques in relation to one another.

<Figure 1>

## **2. Archaeological background to the production of Islamic glazed ceramics**

### *2.1. Coptic Glazed Wares (CGW)*

Coptic Glazed Wares (CGW) is a term first used by Rodziewicz (1976a, 63-4; 1976b, 209; 1978; 1983) to describe a certain group of ceramics found in Alexandria, Egypt, that occur immediately after the late Roman levels in the archaeological sequence, in layers containing the most ancient Islamic coinage. Rodziewicz suggested Egypt as the original production centre for the CGW given that their shapes and modes of decoration closely imitate those of the local late Roman wares.

CGW is classified into two distinct decorative types: first, a painted type covered with an overall translucent amber glaze, and second, an opaque yellow, green, and/or brown glazed ware. The latter is the focus of this paper. The characteristic of this type is that, rather than an overall decoration, it is covered by glaze in discrete bands with some areas left unglazed (Fig. 2.a). Such vessels were recorded also from Fustat (Scanlon 1998; Gayraud 2009), Abu Mina (Engeman 1990), Tod (Joel 1992), Aqaba (Whitcomb 1989; 1991, 48-56), Tiberias (Stacey 1995, 164-166, 286), Pella (Walmsley 1995, 664, 667-8; Walmsley 1997, 2-3), Yoqne'am (Avissar 1996) and Caesarea (Arnon 2008, 35 and 400).

### *2.2. Yellow Glaze Family Wares (YGF)*

Further development and use of CGW decorative techniques is characterised in ceramics excavated in, amongst other sites, Tell Aswad, near Raqqa (Watson 1999), Al-Mina (Vorderstrasse 2005, 75-8), Qinnasrin (Whitcomb 1999, 81-3), Antioch (Waage 1948), and Tarsus (Bagci 2016). The wares were labelled 'Yellow Glaze Family' (YGF) by Watson (1999, 81) after his

classification of the sherds from Raqqa. Despite its name, the YGF encompasses not only opaque yellow glazed wares but also opaque white glazed wares with green and/or brown decorations (Watson 1999, 82-3) (Fig. 2.b). Of all the ceramics found in the aforementioned sites, YGF wares make up one of the main types in the corpus; for instance, three-quarters of the excavated glazed sherds in Raqqa (Watson 1999).

### 2.3. *Samarra-type pottery*

Since the excavations led by Ernst Herzfeld and Friedrich Sarre at Samarra before the First World War and the publication of *Die Keramik von Samarra* by Sarre (1925), the so-called ‘Samarra-type’ pottery, securely dated to the ninth century AD, has become a major focus of research. Delicately fine with a radically new range of decorations, the corpus represented a turning point in the history of Islamic ceramic manufacture. Characteristic wares were covered by a background opaque white glaze, sometimes plain, but generally with blue, splashed green or lustre over-glaze decoration (Kervran 1977, 88, 152-3, Table II; Whitehouse 1979; Northedge and Kennet 1994; 25-34; Northedge 2001, 211-213; Priestman 2011, 90-95). Other groups of glazed pottery, such as opaque yellow glazed wares were found in relatively smaller quantities. A wider range and more significant quantities of opaque white and yellow glazed wares were found in another key archaeological assemblage of Samarra-type pottery in Susa, Iran (Koechlin 1928; Rosen-Ayalon 1972; 1974; Kervran 1977; 1979; Guillermina *et al.* 2005). Bowls with runs of green and/or brown from the rim are found amongst both the opaque yellow and opaque white wares of Susa (Figs. 2.c and 2.d).

Previous chemical analysis of Samarra-type glazes has been focused on opaque white glazes and, except for few individual samples, opaque yellow glazes have not been previously examined and the technological aspects of their production has not been addressed (e.g., Sarre 1925, 95-100;

Crowe 1977; Jones 1980; Tite 1988). Kleinmann (1986) undertook comprehensive study of a range of Samarra-type wares using optical microscopy and electron microprobe. Subsequently, Mason and Tite (1997) published compositional data for opaque white glazed ceramics from Basra, Samarra and Nippur, as well as for later opaque white glazed ceramics from Fustat in Egypt. Both studies suggested two main groups of tin-opacified white glazes: that is, high lead-alkali glazes with approximately 50 wt% PbO, and alkali-lead glazes with 5-10 wt% PbO.

<Figure 2>

#### *2.4. Dating of CGW, YGF, and Samarra type wares*

The precise dating of early Islamic pottery in Egypt and the Levant is still far from clear, and the attempts to “extract reliable dates amidst the confusion” is well described by Stacey (2004, 11 and cpt 3). The assumption of the precedence of ninth-century Samarra for the beginnings of all glazed wares has much distorted the view taken by archaeologists and art-historians in the past and is still current in certain quarters. However, in general terms, the dating of the CGW and YGF to the eighth century, and the Samarra horizon wares to the ninth is well supported by archaeological data. In particular the evidence for CGW as the first elaborate glazed ware is clear from excavations at Alexandria and supported by finds from Aqaba (Whitcomb 1989). Finds of YGF across the Levant and Syria in sites of early Abbasid date point to the spread of this technique to new centres of production in the second half of the eighth century, the major finds at Raqqa being contemporary with the development of that city as the Islamic capital under Harun al-Rashid in the very late eighth and early ninth century (Watson 1999).

### **3. Experimental procedures**

#### *3.1. Archaeological glazed ceramic samples*

The following sherds were selected for examination: one CGW sherd from Fustat (the Alexander Kaczmarczyk collection at the Research Laboratory for Archaeology and the History of Art, Oxford); one CGW sherd from Madaba (University of Toronto); thirty-six YGF sherds from Aqaba (The Oriental Institute Museum, Chicago), Raqqa (Museum für Islamische Kunst, Berlin) and Al-Mina (Victoria and Albert Museum, London); and thirty-three Samarra-type sherds from Mesopotamia, of which sixteen sherds were from Samarra (twelve from the Victoria and Albert Museum, London, four from the Museum für Islamische Kunst, Berlin), twelve from Susa (Musée du Louvre, Paris), four from Basra (Metropolitan Museum of Art, New York) and one from Kish (Victoria and Albert Museum, London). The sherds were examined in the SEM, and all the bodies were analysed by EDS. However, due to weathering, the condition of some of the glazes was not suitable for chemical analysis. Photographs of the sherds (Fig. S1) and the accession numbers (Table S1) are given in the online supplement.

### 3.2. SEM-EDS

Polished sections through the glazes and into the bodies were prepared for investigation under scanning electron microscope (SEM). A JEOL JSM- 5910 SEM with Oxford Instruments EDS (INCA 300 System) was used for examination of the archaeological and replicated samples. The system was operated at 20 kV with 120s measuring time for EDS analyses. Glaze and body microstructures were studied and recorded in backscattered electron mode (BSE), in which the different phases present could be distinguished on the basis of their atomic number contrast.

The Oxford Instrument system was calibrated using primary mineral standards. Both before and after a series of analyses, the accuracy of the system was checked by three analyses of Corning A and Corning B glass standards (Table 1), obtaining agreement better than 25% for  $\text{Al}_2\text{O}_3$ ,  $\text{FeO}$ ,



MnO, Sb<sub>2</sub>O<sub>3</sub> and PbO, and better than 10% for the rest of the elements. Analytical precision was better than 5% for major elements and better than 25% for minor elements.

Due to the problem of quantifying a heavy element in a light matrix (i.e., Pb in a Si matrix; see Goldstein *et al.* 2003, 403-421), major deviations in analytical totals occurred consistently for very high lead (>60 wt% PbO) and high lead-alkali (35-50 wt% PbO) glazes with compositions respectively close to the DLH1 and Corning C glass standards. The ‘measured values’ for PbO and SiO<sub>2</sub> in the DLH1 and Corning C glass standards deviated from the ‘most probable values’ by  $\pm 6\%$  relative, resulting in totals of around 90-100% (Table 1). To compensate for this effect, the Pb-M $\alpha$  and Si-K $\alpha$  peaks in the DLH1 glass standard (with a similar matrix to opaque yellow glazes) were used as the primary standards for the calibration in measuring the composition of the high lead glazes, and the Pb-M $\alpha$  and Si-K $\alpha$  peaks in the Corning C glass standard (with a similar matrix to opaque white glazes) were used as the primary standards for the calibration in measuring the composition of lead-alkali glazes. The resulting totals ranged between 98-102%, as given in the glaze analysis tables in the Results section.

< Table 1 >

EDS measurements on the bodies were performed by analysing three areas of approximately 1 mm x 0.8 mm. Depending on the degree of porosity, the totals ranged between 68-75%. The average of the resulting normalised totals were reported as the chemical composition of sherd bodies. For the glazes, bulk EDS compositions were determined by analysing areas up to about 0.4 mm x 0.4 mm. The problem of inhomogeneity of glazes due to the presence of scattered opacifying particles, undissolved quartz/feldspar grains, air bubbles and, in some cases, extensive weathering was of considerable importance. To help moderate this effect, three to five areas were analysed in each glaze and the average normalised compositions were reported. The standard deviations for

different oxides in bodies and glazes were very variable, but to give some indication of variation, average SD values from three to five readings were given in the body and glaze analysis tables in the Results section.

SnO<sub>2</sub> and PbO were about 0.6%, and for P<sub>2</sub>O<sub>5</sub>, ZnO and As<sub>2</sub>O<sub>3</sub> around 0.2%. For all other oxides, values below 0.1% were unreliable and were not taken into account. Typical average standard deviations for bodies, opaque yellow glazes, and opaque white glazes

### 3.3. *SR- Micro-XRD*

SR-Micro-XRD was performed at the Xaloc beamline at the ALBA synchrotron, Barcelona, Spain, using 12.661 keV energy (0.979  $\mu\text{m}$  wavelength) and 50 x 6  $\mu\text{m}^2$  spot size. The small beam size facilitated the acquisition of information from micro-crystalline compounds. A series of diffraction patterns from thin cross-sections of the samples (between 50 and 100  $\mu\text{m}$ ) were obtained, the cross sections being scanned vertically at steps of 5 microns to allow some overlapping. A virtually noise free Pilatus 6M (Dectris) detector, with a large active area (424x435 square millimeters, 6 Mpixels) protected by a small beam stop, was used, typical acquisition times varying between 0.2 to 1 s. The powder diffraction patterns of the layers were obtained by radial integration of the images obtained by this two-dimensional detector. This setup provides a high sensitivity to poor diffracting compounds present in very low amounts, and a large  $2\theta$  range ideal for compound identification.

### 3.4. *XRD*

Qualitative X-ray diffraction analysis was conducted to identify crystalline phases in calx powders using a Bruker D8 Advance XRD with a vertical goniometer and Bragg-Brentano  $\theta$ - $2\theta$

configuration for powder diffraction measurements. Identification of the compounds has been performed based on the Powder Diffraction File (PDF) database from the International Centre for Diffraction Data (ICDD).

### *3.5. Replication experiments*

#### *3.5.1. Calx powders*

The three calx recipes presented in Table 2 were chosen and prepared at the University of Vic, Spain. Pure lead in the form of sheets and pure tin in the form of granules were used in the recipes. The mixtures were put in dish-like crucibles (15 cm in diameter), made out of clay from La Borne, France (known as grès français) which had been biscuit fired. Lead was first put in crucibles and fired up to 400°C in a Hobersal MF22-134 electrical furnace. Tin was then added to the resulting melt. After the addition of tin, the mixtures were stirred using steel rods. Figure 3 shows the firing schedule and the three stages of sampling (i.e., 400°C, 600°C and 800°C).

Samples of calx powders were ground using an agate mortar and pestle, passed through a 240-mesh sieve (BSS 63 µm), and then pressed into pellets and mounted on a holder for XRD examination.

<Table 2>

#### *3.5.2. Replicate glazed ceramics*

In order to imitate the calcareous clays of the Levantine and Mesopotamian bodies, British Gault clay was used for the body pastes (Table 3). Clay pastes were wedged and kneaded for a few minutes. The resulting lumps were flattened and formed into discs using a plastic mould 35 mm in diameter. The resulting tabular discs were left to dry for 24 h and then biscuit-fired to 900°C.

Three glazes were prepared using mixtures of the C7.600 (C7 calx fired to 600°C, see Fig. 3) and C30.600 (C30 calx fired to 600°C, see Fig. 3) calces with silica, and the C7.600 calx with silica,

alkaline frit and calcium carbonate (Table 4). The compositions of the components used in these glaze mixtures are given in Table 3. Glaze ingredients were weighed and mixed, and the resulting mixtures were ground in a porcelain mortar and passed through a 240-mesh sieve (BSS 63  $\mu\text{m}$ ). The glazing powders were mixed with water and 10 ml syringes were used to apply the mixture to the surface of the biscuit-fired clay discs. The glazed discs were then fired to 850°C and 950°C in a Lenton electrical furnace at the Research Laboratory for Archaeology and the History of Art (RLAHA), Oxford, according to the firing schedule shown in Figure 4.

<Tables 3 and 4>

<Figure 4>

## 4. Results

### 4.1. Body

The chemical compositions of the bodies are presented in Tables 5 and 6. For the Egypt and Levant ceramics, the classification of the sherds into different groups is based on the typology suggested by Watson (1999), and for the Mesopotamia ceramics that by Northedge and Kennet (1994) and Priestman (2011).

The EDS analyses indicate that all the Levantine bodies were produced from calcareous clays (10-26 wt% CaO and 2-9 wt% MgO) which on firing results in a fairly porous structure. In contrast, the Egyptian Fustat.1 CGW sherd was produced from aluminium-rich clay (23.4 wt%  $\text{Al}_2\text{O}_3$ ), typical of the use of Nile silt (Fig. 5.a), which on firing results in a more compacted structure.

The body fabrics of the Levant sherds examined were either medium-grained, with inclusion sizes between 50 to 200  $\mu\text{m}$ , or coarse-grained, with inclusion sizes greater than 200  $\mu\text{m}$ . Non-plastic inclusions mainly include quartz, K-feldspar, mica, plagioclase, pyroxene, or a combination of the minerals as a rock fragment, as well as more occasional microfossils (Fig. 5.b). However, the non-

plastic inclusions are not sufficiently distinct to classify the body fabrics into groups by SEM examination, with the exception of the body of Madaba.5 CGW sherd which is made essentially of clay pellets within a porous calcareous clay (Fig. 5.c), and bodies of the Raqqa.22 kiln rod and Raqqa.20 sherd which contain added fragments of high lead glass (Fig. 5.d).

<Table 5>

<Figure 5>

In contrast, the majority of body fabrics of the Mesopotamian sherds examined, including plain white, splashed, blue-on-white, lustre and yellow-glazed wares, were fine-grained, with non-plastic inclusions being less than 50 µm in size, (labelled 'f' in Table 6; Fig. 6.a). Non-plastic inclusions in these bodies, previously referred to as the 'Basra fabric'<sup>1</sup>, were mainly too fine for identification in the SEM. Exceptions were one sample of lustre ware (Samarra.26) and one sample of yellow polychrome glazed ware (Susa.1) that contained peloidal phosphate phases with the composition  $\text{Ca}_3(\text{PO}_4)_2$  (labelled 'f+phos' in Table 6; Fig. 6.b). The abundance of this phase in the Samarra.26 sample raised the bulk  $\text{P}_2\text{O}_5$  content to about 6%. Some of the Basra samples, including one kiln rod, contained a small number of dispersed quartz and feldspar inclusions of about 300 µm in size (labelled 'f+q/feld' in Table 6; Fig. 6.c).

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<sup>1</sup> On the basis of the study of twenty-seven ceramic fragments and kiln rods from Basra at the Metropolitan Museum of Art (see Table 6 and Fig. 6.c for Basra bodies), the term 'Basra fabric' has sometimes been used (e.g., Mason and Keall 1991; Hallett 2000; Mason 2004) to describe the typical fine-grained calcareous bodies from the southern part of Mesopotamia and Western Iran. However, the homogenous geology of the Southern Mesopotamian region means that the petrographic fabrics and compositions of the bodies are not distinctive enough to make secure suggestions concerning provenance, and as a result, the same clay types could have been used in various production centers over wide geographic areas (Philip 1968; Berry *et al.* 1970). Therefore, it is entirely possible that the same clay was used in Basra as well as in other Southern Mesopotamian production centres. As a result, the more general term 'Southern Mesopotamia body' is preferred to the site-specific term "Basra fabric".

Only a relatively small number of the Mesopotamian samples examined, including yellow-glazed, plain white and splashed wares, were either medium-grained (50-200  $\mu\text{m}$ ) or coarse-grained (more than 200  $\mu\text{m}$ ), (labelled 'm', 'm/c', or 'c' in Table 6). These bodies included a high content of identifiable non-plastic inclusions, such as quartz, feldspar, mica and pyroxene, but further differentiation of petro-fabrics by SEM was again not feasible. These bodies appear similar in their microstructures to those of the Levantine wares. The body of the splashed green sherd Samarra.5 included relicts of alkali-lead glass (labelled 'c+glass' in Table 6; Fig. 6.d).

The bodies of all the Levantine sherds and the Mesopotamian sherd with very high lead or high lead-alkali glazes contain up to a maximum of about 2 wt% PbO. This is probably due to the impregnation of the ceramics by lead vapour during firing.

<Table 6>

<Figure 6>

#### 4.2. Glaze

The chemical compositions of opaque yellow and white glazes are given in Tables 7 and 8 and those of overglaze green, black and blue decorations are given in Table 10. The distinct grouping of the glazes on the basis of the PbO, SiO<sub>2</sub> and SnO<sub>2</sub> concentrations normalised to 100 wt% is illustrated in Figure 7.

##### 4.2.1 Yellow glazes

The chemical composition of all the opaque yellow glazes, whether from Egypt, the Levant or Mesopotamia, differ from the opaque white glazes in containing very high lead oxide (>60 wt% PbO) and negligible alkali (<2 wt% Na<sub>2</sub>O+K<sub>2</sub>O) (Tables 7 and 8; Figure 7). Their tin oxide contents are less than 5 wt%, and their Pb/Sn ratios, representing the calx composition, vary

between 16 and 54. The microstructures of glazes as seen by SEM show a distribution of bright lead-tin-oxide particles, air bubbles and quartz grains through the glaze thickness (Figs. 8.a and 8.b). Micro-XRD analyses (Fig. S2) and EDS compositions (Table 9) of the lead-tin-oxide particles indicate that they are of the form lead-tin-oxide type II (i.e.,  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$ ) in which the Sn/Si ratio ranges between 4.5 and 10.5.

SEM images show that for yellow glazed sherds from Egypt, the Levant and Mesopotamia, glaze thicknesses vary between 50 and 300  $\mu\text{m}$ . The body-glaze interaction layers in yellow glazed sherds from Raqqa and Al-Mina is typically around 20-40  $\mu\text{m}$  (Table 7) and include secondary crystals of wollastonite ( $\text{CaSiO}_3$ ) and diopside ( $\text{CaMgSi}_2\text{O}_6$ ) (Fig. 8.a). In contrast, the interaction layers for sherds from Fustat, Madaba, Aqaba and Mesopotamia (Susa and Kish) are negligible ( $<10\text{ m}$ ; Fig. 8.b; Tables 7 and 8).

#### 4.2.2 White glazes

The opaque white glazes fall into three major compositional groups. The first group, which is exclusively used in the Levant and to a lesser extent in Mesopotamia (Tables 7 and 8), consists of high lead-alkali glazes with lower lead oxide contents (34-46 wt% PbO) than the opaque yellow glazes, but containing significantly higher concentrations of alkalis (3-6-wt%  $\text{Na}_2\text{O}+\text{K}_2\text{O}$ ) and  $\text{SnO}_2$  (6-16 wt%). The Pb/Sn ratios representing the calx composition vary between 2.5 and 9. The microstructure of glazes as seen by SEM shows a distribution of bright tin oxide particles, as confirmed by micro-XRD and EDS analyses, air bubbles and quartz grains (Fig. 8.c).

The remaining two compositional groups of opaque white glazes, which were only used in Mesopotamia (Table 8), are alkali-lead glazes (10-12 wt%  $\text{Na}_2\text{O}+\text{K}_2\text{O}$ ; 2-9 wt% PbO) and alkali glazes (12-15 wt%  $\text{Na}_2\text{O}+\text{K}_2\text{O}$ ;  $<1$  wt% PbO). The alkali-lead glazes, which are associated with the plain white, blue-on-white, and lustre wares from Samarra and Susa, are again opacified with

tin oxide particles, air bubbles and quartz grains (Fig. 8.d), but exhibit Pb/Sn ratios approximately equal to 1. Some splashed green wares might also fall into this group but no sherds of this type were included among those selected for analysis as part of this study. The alkali glazes, which are associated with blue-on-white, splashed and lustre wares from Samarra, Susa and Basra, do not contain any tin oxide particles but are instead opacified with a proliferation of undissolved quartz grains and air bubbles (Fig. 8.e).

The glaze thicknesses as seen by SEM images is about 100-550  $\mu\text{m}$  for the Levantine and Mesopotamian high lead-alkali white glazed sherds but is significantly thicker (up to 1000  $\mu\text{m}$ ) for Mesopotamian alkali and alkali-lead white glazed sherds. The body-glaze interaction layers vary between 30-80  $\mu\text{m}$  for white glazed sherds from Raqqa and Al-Mina (Fig. 8.c), but are negligible for sherds from Aqaba, Susa and Samarra (Figs 8.d and 8.e; Tables 7 and 8); with the exception of Samarra.2 and Samarra.13 for which interaction layers are nearly 20  $\mu\text{m}$  thick.

<Tables 7, 8 and 9>

<Figures 7 and 8>

#### *4. 2.3 Green, brown and blue glazes*

The compositions of the green, brown and blue decorations are given in Table 10, and show that Cu, Mn and Co colorants were used for the green, brown and blue respectively.

Fe and Zn were associated with the Cu for the Egyptian/Levantine and Mesopotamian green glazes respectively. The compositions of the green glazes were similar to those of their underlying yellow and white glazes. However, compared to the underlying yellow or white glazes, the green glazes contained fewer or no opacifying particles (Fig. 9.a) which suggests that calx was probably not included in the green glazing mixtures. The brown glaze in the case of Raqqa.25 had merged with



the previously applied underlaying yellow glaze (Fig. 9.b), and as a result, contained about the same amount of opacifying particles (Tables 7 and 10, Raqqa.25; see Molera *et al.* 2013).

FeO and ZnO were associated with the Co for the Mesopotamia blue glazes. The compositions of blue glazes for Susa.2 and Samarra.19 are again similar to those of their underlying white glazes (Fig. 9.c). In contrast, the compositions of blue glazes for Samarra.20, Samarra.21 and Samarra.23 differ from the compositions of their underlying white glazes in containing measurable amounts of lead oxide (1-18 wt% PbO; Fig. 9.d).

<Table 10>

<Figure 9>

#### **4.3. Relationship between body types and glaze compositions**

Principal Components Analysis indicates that, although all made from calcareous clays, the bodies can be grouped on the basis of the K<sub>2</sub>O, MgO and FeO contents (Fig. 10). The fine-grained Mesopotamian bodies (Table 6) which are associated with alkali-lead and alkali white base glazes (see Table 8 and Figure 7), form a fairly tight compositional group with lower K<sub>2</sub>O and higher MgO and FeO contents. In contrast, the medium and medium/coarse-grained Egyptian/Levantine bodies (Table 5) which are associated with very high lead yellow glazes and high lead-alkali white glazes (see Table 7 and Figure 7), form a separate, more dispersed compositional group. It should also be noted that the K<sub>2</sub>O, MgO and FeO contents of the bodies of the kiln rods found at Basra and Raqqa fall, respectively, within these Mesopotamian and Egyptian/Levantine compositional groups.

The Mesopotamian very high lead yellow glazed bodies, three of which are fine-grained and one medium/coarse-grained, form a tight compositional group with higher K<sub>2</sub>O contents than the Mesopotamian fine-grained, white glazed bodies, but similar MgO and FeO contents. This

compositional group therefore falls on the edge of the Egyptian/Levantine compositional group. The medium and coarse-grained Mesopotamian bodies, which are associated with high lead-alkali white base glazes mainly fall within the Mesopotamian fine-grained, white glazed compositional group. The exceptions are the Susa.18 sherd (plain white) which falls within the very broad Egyptian/Levantine cluster, as do Susa.17 (plain white) and Susa.13 (splashed green) for which we do not have glaze analyses. As seen in Figure 10, the three Susa sherds do not form a cluster but are widely dispersed within the Egyptian/Levantine group.

<Figure 10>

### *4.3. Replication experiments*

#### *4.3.1 Replicated calces*

At 400°C, for all three calx mixtures, molten phases were formed in the middle of the crucibles and these were surrounded by dark grey ash-like powders. XRD analysis suggested that the grey colour of the powders was due to the formation of romarchite (SnO) crystals. Other identified crystals were tin (Sn) and cassiterite (SnO<sub>2</sub>) in C0 calx, and lead (Pb), tin (Sn), litharge (PbO, tetragonal structure), massicot (PbO, orthorhombic structure), and cassiterite (SnO<sub>2</sub>) in C7 and C30 calces.

With further heating to 600°C and 800°C, the C0 calx turned into a fine white powder (Fig. 11) which XRD indicated was mainly cassiterite with negligible romarchite. The colour of the C7 and C30 calx powders progressively changed to yellow with increasing temperature. In addition, at 800°C, the C30 calx had reacted with the clay crucible to form a glassy layer on the surface of the crucible. XRD peaks indicated that the powders contained mainly lead-tin-oxide type I (i.e., Pb<sub>2</sub>SnO<sub>4</sub>) and cassiterite, as well as massicot, litharge, and in the C30 calx fired to 800°C, lead-tin-oxide type II (i.e., Pb(Si,Sn)O<sub>3</sub>) (Fig. S3).

<Figure 11>

#### 4.3.2. Replicated glazes

When the glazed sherds were fired to 850°C, glaze mixtures G1 and G2, which contained calces C30 and C7 respectively and only quartz, produced opaque yellow glazes (Fig. 12). In SEM examination, these glazes (G1.850 and G2.850) were seen to contain scatters of lead-tin-oxide type II particles with the formula  $\text{Pb}(\text{Si},\text{Sn})\text{O}_3$ . (Figs. 13.a and 13.c). In contrast, glaze mixture G3, which contained calx C7 and both quartz and alkaline frit, resulted in a white glaze (G3.850) speckled with small yellow spots (Fig. 12), and a microstructure containing tin oxide particles dispersed densely throughout the thickness of the glaze (Fig. 13.e). Air bubbles and unreacted quartz grains could also be observed throughout the thicknesses of all three glazes.

When sherds glazed with mixture G1 were fired to 950°C, a glossy surface resulted which exhibited a translucent brown colour with milky white opaque appearance in the middle (Fig. 12). The microstructure of this glaze (G1.950) was free of tin-based opacifying particles, but air bubbles and partially reacted quartz grains were occasionally seen (Fig. 13.b). The glaze-body interaction layer contained a variety of secondary-formed calcium-lead-silicate crystals. When fired to 950°C, glazing mixtures G2 and G3 also resulted in glossy and smooth surfaces but with these mixtures, both the glazes (G2.950 and G3.950) were an opaque white colour, and their microstructures contained tin oxide particles ( $\text{SnO}_2$ ) (Figs. 13.d and 13.f). The body-glaze interaction layers again contained a variety of secondary-formed calcium-lead-silicate crystals.

As a result of interaction between the glaze and the body, the chemical compositions of all the glazes showed a shift in the  $\text{PbO}$  and  $\text{SiO}_2$  contents compared to the respective glazing mixture compositions as calculated from their recipes (Table 11), an approximate 6 wt% decrease in the  $\text{PbO}$  and 3 wt% increase in the  $\text{SiO}_2$  contents being observed. The  $\text{Al}_2\text{O}_3$  and  $\text{CaO}$  contents of the

glazes also increased as a result of diffusion from the body into the glaze.

<Figures 12 and 13>

<Table 11>

## 5. Discussion

### 5.1 Replication experiments

#### 5.1.1 Conversion of lead-tin-oxide type I to type II

Chemical analysis of the replicate samples showed that in the calx powders, lead-tin-oxide particles were in the form  $\text{Pb}_2\text{SnO}_4$  (type I) but when analysed in the glaze, they had been converted into the form  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$  (type II). Previous studies by Rooksby (1964), Kühn (1968) and Clark *et al.* (1995) had demonstrated that conversion of lead-tin-oxide type I to type II would take place when silica ( $\text{SiO}_2$ ) is added to the lead-tin-oxide type I and the mixture is fused between 800°C and 950°C. During this process  $\text{Pb}_2\text{SnO}_4$  incorporated variable amounts of  $\text{SiO}_2$  which substituted for  $\text{SnO}_2$ . These earlier results were further confirmed in the present work by heating the C30 calx to 800°C. In the glass layer produced as a result of partial reaction with the clay crucible, both lead-tin-oxide types I and II were detected (C30.800). This observation is the result of the reaction of  $\text{SiO}_2$  from the crucible body with the  $\text{Pb}_2\text{SnO}_4$  in the calx which resulted in the formation of  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$ .

#### 5.1.2 Dissolution of lead-tin-oxide type II and precipitation of tin oxide

The use of both opaque yellow and opaque white glazes during the early Islamic period was facilitated by a crucial change occurring during firing of the glaze. As demonstrated by Molera *et al.* (1999) and Tite *et al.* (2008), in a glaze composed of  $\text{PbO}$ ,  $\text{SiO}_2$  and  $\text{SnO}_2$ , the lead-tin-oxide type II ( $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$ ) crystals are stable at up to a temperature in the range from approximately 750°C to 1000°C, depending upon the exact composition of the glaze. At higher temperatures, the

$\text{Pb}(\text{Sn},\text{Si})\text{O}_3$  crystals dissolve and subsequently, tin oxide ( $\text{SnO}_2$ ) crystals precipitate from the melt. Hence, the colour of the glaze changes from yellow to white.

In the G1 glaze, produced using calx C30 and fired to about  $950^\circ\text{C}$ , the dissolution of  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$  and formation of secondary  $\text{SnO}_2$  crystals seemed to have started, but had not yet been completed. As a result, the microstructure of G1.950 showed no opacifying particles due to the complete dissolution of the  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$  crystals, and the colour of the G1 glaze had changed from yellow at  $850^\circ\text{C}$  to brownish yellow with milky halos at  $950^\circ\text{C}$ . The milky halos seemed to represent the areas of the glaze in which conversion to  $\text{SnO}_2$  particles had taken place, although no such particles were observed in the SEM. This might have been due to the very small sizes of the  $\text{SnO}_2$  nucleates formed. In contrast, in the glaze G2, produced using calx C7 and fired to about  $950^\circ\text{C}$ ,  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$  particles dissolved in the glaze and  $\text{SnO}_2$  particles were extensively formed. Thus, the colour of the glaze changed from yellow at  $850^\circ\text{C}$  to completely white at  $950^\circ\text{C}$ .

The addition of alkalis and alkaline earths to the glazing recipe significantly decreased the temperature of conversion. Thus, glaze G3, again produced using calx C7 but containing approximately 8 wt% alkalis and alkaline earths ( $\text{Na}_2\text{O}+\text{K}_2\text{O}+\text{CaO}+\text{MgO}$ ), exhibited a white colour with only a few yellow speckles in the center when fired at  $850^\circ\text{C}$ . Then, when fired to  $950^\circ\text{C}$ , the yellow speckles had disappeared and the resulting glaze was completely white.

#### *5.1.3 Role of lead in calcination of tin*

Kingery and Vandiver (1986, 116) had previously argued that when tin was heated alone, it resulted in a grey powder that was not suitable for use in glaze production. In contrast, when a combination of lead with tin was heated (e.g.,  $\text{Pb}/\text{Sn}=3$ ), the resulting mixture was a white powder. They therefore claimed that this was the reason why lead always accompanied tin in the calcination process.

Contrary to this assumption, the calcination experiment with the C0 mixture demonstrated that metallic lead is not necessary for calcination of tin. When heated to 400°C, the metallic tin in the calx C0 turned into tin oxide (SnO) and did not proceed to form tin dioxide (SnO<sub>2</sub>) in significant quantities. The result was a dark grey powder as pointed out by Kingery and Vandiver (1986, 116). However, at 600°C, C0 was fully calcined to tin dioxide, resulting in a fine white powder, thus demonstrating that the presence of metallic lead is not always necessary for complete calcination of tin.

## *5.2 Glazes*

On the basis of the replication experiments, it can be argued that there are close technological links between the first invention and use of tin-opacified yellow and white glazes. Thus, to produce a white glaze, the PbO content used in the production of a yellow glaze was reduced and the SnO<sub>2</sub> content was increased, such that the Pb/Sn ratio for the white glaze was reduced to 2.5-9, as compared to 16-54 ratio for the yellow glaze. In addition, the alkali content of the white glaze was increased to 3-6 wt% as compared to a negligible alkali content in the yellow glaze. As discussed in Section 5.1.2, the increased alkali content used in the production of white glazes results in the formation of a completely opaque white glaze at a lower firing temperature than would have been the case if the very high lead glaze type had been used.

The compositions of all tin-opacified yellow glazes from Mesopotamia examined in this paper are of the very high lead type, and thus, directly comparable to those produced in Egypt and the Levant. The relief-moulded (Susa.7, Susa.12) and the green-and-brown-on-yellow glazed wares from Susa (Susa.1) definitely seem to be local Mesopotamian productions, in that their bodies are fine-grained and on the basis of their K<sub>2</sub>O, MgO and FeO contents, they form a tight compositional group (Fig. 10). The earlier dates for the Egyptian and Levantine yellow glazed wares suggest that

they are the precursor of the Mesopotamian yellow glazed wares. The similarity in shape and decorative patterns of opaque glazed ware finds from Susa to those from Egypt and Levant, essentially with flat bases or low footrings and steep walls, also attests to both technological and stylistic links between these productions (Fig. 2; see Watson 2014). In contrast, the body of the yellow glazed

sherd from Kish is medium/coarse-grained, and although its  $K_2O$ ,  $MgO$  and  $FeO$  contents match those of the Susa sherds, they are also consistent with those of Egyptian and Levantine bodies. Therefore, it is not impossible that the Kish ceramic was imported into Mesopotamia from the Levant.

Tin-opacified white glazes of Mesopotamia fall into two compositional groups, confirming results previously published by Kleinmann (1986), and Mason and Tite (1997). Firstly, there is a high lead-alkali type with calx  $Pb/Sn$  ratios between 2.5 and 9, and secondly, an alkali-lead type with calx  $Pb/Sn$  ratios around 1. In addition, there is a third Mesopotamian white glaze group with an alkali type glaze which is not tin-opacified but instead opacified by a combination of bubbles, quartz and other mineral particles.

The high lead-alkali Mesopotamian glazes include plain white and splashed green decorated wares reported above, as well as two previously analysed *sancal*-type sherds which were considered to have been produced in Mesopotamia (Rawson *et al.* 1988). The  $K_2O$ ,  $MgO$  and  $FeO$  contents of the bodies of these Samarra sherds fall within or close to the compositional group for fine-grained Mesopotamian bodies associated with alkali-lead and alkali white base glazes. Therefore, it seems probably that the high lead-alkali white glazed wares found at Samarra were locally produced in Mesopotamia, but at a different production centre to that of the fine-grained, alkali-lead and alkali glazed wares discussed below. However, the medium/coarse-grained bodies of these sherds, the

high lead-alkali composition of their glazes and the thicknesses of their body-glaze interaction layers are directly comparable to those for white glazed ceramics found in the Levant, and thus, their import into Mesopotamia from the Levant remains a possibility. In contrast to these Samarra sherds, the  $K_2O$ ,  $MgO$  and  $FeO$  contents of the plain white Susa.18 sherd, which also has a high lead-alkali glaze and medium-grained body, falls within the Egypt/Levantine compositional group. Therefore, it is more likely that it was imported into Mesopotamia from the Levant.

Unlike the high lead-alkali glazed wares, the alkali-lead white glazes, which were applied to plain white, splashed green, blue-on-white, and lustre wares, have not been used on Egyptian and Levantine wares, and are only seen on Mesopotamian sherds, and always associated with fine-grained fabrics (Table 8; see also Wood *et al.* 2007). On this basis, one can conclude that these wares represent local Mesopotamian productions. One point to note about the plain white sherds is that they might in fact be fragments from a blue (and green)-on-white or splashed green decorated piece where no coloured decoration was applied, or a fragment of lustre ware where the lustre decoration had disappeared by weathering. Therefore, the type of the decoration associated with such samples cannot be firmly established from the available sherd. Other features of the alkali-lead white glazes that distinguishes them from the high lead-alkali glazes are their comparatively larger thicknesses (up to 1000  $\mu m$ ) and the abundance of air-bubbles and undissolved quartz grains. These microstructural features were also widely observed among the non tin-opacified alkali white glaze associated with blue-on-white, splashed and lustre wares, again with fine-grained bodies, as well as with pre-Islamic green and turquoise glazes in Mesopotamia (Hill 2004; Pace *et al.* 2008). Even though the dating for Mesopotamian non tin-opacified alkali and tin-opacified alkali-lead glazes are not sufficiently definite to establish the sequence of production, the results may suggest that the Mesopotamian potters began the initial



stage of producing blue-on-white and lustre wares by creating the opacification effect through the inclusion in the glaze of air-bubbles and undissolved quartz grains, in a manner widely practiced during the pre-Islamic period.

The compositions of the over-glaze blue decorations suggest that, in contrast of the copper green decoration, the cobalt is sometimes incorporated in a different glaze composition to that of the underlying white glaze (Table 10). The composition of Samarra.20, Samarra.23 and Samarra.21 glazes contain higher PbO contents than their base white glazes which indicates that, as previously discussed by Wood *et al.* (2007), the cobalt pigments used for these glazes were either mixed or fritted with lead oxide, and then applied over the base white.

In summary, it would appear that the technologies for both the opaque very high lead yellow glaze and the high lead-alkali white glaze were transferred from Egypt/Levant into Mesopotamia. However, in addition, separate but parallel white alkali glazes without tin-opacification and white tin-opacified alkali-lead glaze technologies were introduced into Mesopotamia. As discussed by Tite *et al.* (2015, 89), it seems possible that these latter technologies were related to the production of Islamic opaque white glass which was of the tin-opacified alkali-lime type containing only a few wt% PbO. White glasses opacified by SnO<sub>2</sub> from the Abbasid period are found in Samarra and Ctesiphon, Iraq (9<sup>th</sup> century; Schibille *et al.* forthcoming; M. Wypyski, personal communication), and in Nishapur, Iran (8<sup>th</sup>-10<sup>th</sup> century; Pilosi *et al.* 2012; Wypyski 2015), and were produced using calces with Pb/Sn ratios in the range from 0.1-2.4 which is similar to the calces in the range from 0.6-2.3 used in the production of opaque white alkali-lead glazes from Mesopotamia. Since both opaque white glass and opaque white alkali-lead glazed ceramics with similar calx Pb/Sn ratios were concurrently used in Mesopotamia during the 9<sup>th</sup> century, it is

reasonable to argue that the white tin-opacified glass and glaze technologies developed in conjunction with one another.

### *5.3. The spread of tin-opacified yellow and white glazes*

The initial spread of Islamic glazed wares to Iraq in the ninth century may be related to the movement of the Islamic capital from Damascus, Syria, to Baghdad, Iraq in 762 AD after the Umayyads (661-750 AD) were overthrown by the Abbasids (750-1258 AD), as a result of which potters and other craftsmen might have been migrated to the new capital. It seems that, during this transition, the composition of lead-tin calx and glazing mixtures for yellow glazes did not change. This lack of variations is partly due to the sensitivity of the manufacturing process of tin-opacified yellow glazes to the firing temperature and the amount of alkalis and alkaline earths. The technique of tin-based opacified yellow glazing continued eastwards to Iran and Central Asia during the tenth century (see Appendix for samples from Takht-i Suleiman, Merv and Nishapur) and was also used during medieval Islamic period in India (Gill and Rehren 2011; Gill *et al.* 2014) and in Iran until the nineteenth century (Isfahani 1888).

The emergence of white glazes in Mesopotamia seems to have had a particularly major impact, in that during the ninth century, the technique became widely used as base white glazes for Samarra-type pottery, produced essentially in response to imported Chinese stoneware and porcelain (Watson 2014). The white glaze also provided an appropriate background for Islamic blue and lustre over-glaze decoration. However, unlike the situation with opaque yellow glazes, the dominant opaque white glaze types employed in Mesopotamia (Samarra, Susa and Basra) were the alkali and alkali-lead types and not the high lead-alkali glaze type employed in Egypt and the Levant.

Subsequently, opaque white glazing technology transferred eastwards to Iran, Central and Eastern

Asia (Priestman 2011) (see Appendix for samples from Samarkand). However, more significant for future development of opaque white ceramic glazes was the spread westwards to Egypt, Tunisia, Spain, Italy, France, Netherlands, Germany, Britain and the rest of Europe (see e.g., Caiger-Smith 1973, 127-140; Whitehouse 1978; 1980; Fourest 1980) where it continued as a main glazing technique until the 18th century. In modern ceramic manufacture, tin oxide is chiefly replaced by zirconium and titanium compounds, but is still employed by artists and craftsmen for the particular opacification and glossy effect it provides.

It appears from published data that changes in the composition of tin-opacified white glazes, particularly the PbO/alkalis ratios of glazes and the Pb/Sn ratios of calces can vary dramatically by region and time period. For example, on the basis of the glaze compositional data for Islamic opaque white glazes summarized in Tite *et al.* (2008, 69), the average Pb/Sn ratios for the associated calces and the average PbO/(Na<sub>2</sub>O+K<sub>2</sub>O) ratios for the glazes vary from 1.2 and 0.4 respectively for Abbasid Mesopotamia; through to 3.3 and 5.0 for Fatimid Egypt, and 4.0 and 2.1 for Seljug Iran; and up to 7.8 and 17 for tenth-century Spain and 6.6 and 22 for twelfth-century Spain. Thus, the glazes used begin in Mesopotamia as the alkali-lead type, convert in Egypt and Iran to the lead-alkali type, and ultimately, in Spain, more-or-less match the high lead type originally used in the Levant, for which the average Pb/Sn ratios for the calces and the average PbO/(Na<sub>2</sub>O+K<sub>2</sub>O) ratios for the glazes are 6.5 and 11 respectively.

## **6. Conclusion**

The SEM-EDS analyses presented here is the largest corpus of data published thus far on the bodies and glazes of Egyptian and Levantine CGW and YGF wares dated to eighth century AD. Combined with analysis of bodies and glazes of ceramics from Mesopotamia, Northern Iran and Central Asia, the data revealed that tin-opacification of glazes began in the eighth-century in Egypt

and the Levant, nearly a century before the hitherto earliest examples of Samarra-type pottery. Yellow glazes of very high lead composition began production in Egypt and the Levant and continued through to Mesopotamia, Northern Iran and Central Asia. White glazes of high lead-alkali composition were also produced in Egypt and the Levant, and seemingly continued through to Mesopotamia, but did not become as prevalent in this region. In addition, and more abundant, was a type of tin-opacified white glaze with alkali-lead composition produced in Mesopotamia from the ninth-century, which appear to have been largely exclusive to the region and was not found anywhere else. Finally, non tin-opacified white glazes, which included quartz grains, air bubbles and occasionally secondary crystals, were found to have alkali compositions in Mesopotamia.

Replication experiments demonstrated that for the manufacture of both tin-opacified yellow and white glazes, the lead-tin calx was the principal ingredient. It is apparent from the glaze compositional data that the Pb/Sn ratios used in the calcination processes were specifically chosen for each glaze type: i.e., Pb/Sn greater about 16 for yellow glazes, Pb/Sn between about 2.5 and 9 for high lead-alkali white glazes, and Pb/Sn equal to about 1 for alkali-lead white glazes. Subsequently, the calx was mixed with silica and fired to about 700°C (Tite *et al.* 2008, 72-5), during which lead-tin-oxide type I converted to type II (i.e.  $\text{Pb}_2\text{SnO}_4$  to  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$ ), rendering a yellow colour to the glaze. By increasing the temperature to nearly 850°C or to above 950°C depending on the exact composition of the glaze, the  $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$  crystals decomposed and eventually re-crystallised in the form  $\text{SnO}_2$ . The colour of the glaze therefore changed from yellow to white. Addition of alkaline salts to the glazing mixture decreased the temperature of the formation of  $\text{SnO}_2$  crystals and played a key role in the change of glaze colour from yellow to white.

In summary, this paper aimed to provide data on a comprehensive range of early Islamic ceramics, to consider anew the origins of tin-opacification of glazes, and to understand and highlight the relationships in the manufacturing processes between opaque yellow and white glazes. There is much work still required to distinguish different manufacturing sites for early Islamic tin-opacified glazed wares, especially by means of trace element analysis of bodies, and to explore further the links between the eastwards movement of this technology and the shift of the Islamic capital from Damascus, Syria, to Baghdad, Iraq in 762 AD. The relationship in the composition and production technology between tin-opacified yellow glasses and glazes in the Levant and in Mesopotamia, as well as between the opaque white glasses and opaque white alkali-lead glazes in Mesopotamia, also offer important directions for future research. Finally, more comprehensive studies of the typology and technology of tin-opacified glazed wares from Northern Iran and Central Asia would provide valuable insights into the later production and development of these wares.

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## **APPENDIX: Tin-opacified glazed sherds from Northern Iran and Central Asia**

The compositions of bodies and glazes of fourteen samples from Takht-i Suleiman, Nishapur, Merv and Samarkand (see Figs.1 and S1, and Table S1) dating to around the tenth century AD are given in Tables A1 and A2 respectively.

The bodies were all produced using calcareous clays containing 7-16 wt% CaO and 2.5-5 wt% MgO. However, as a result of the different geology of the regions in which they were produced, the body fabrics were very variable in terms of the types and sizes of their inclusions.

The yellow glazes, which were found at Nishapur, Merv and Takht-i Suleiman, were of the very high lead compositional type (calx Pb/Sn in range 10-28) and were opacified by lead-tin-oxide particles, air bubbles, and occasionally undissolved quartz grains (Fig. A1.a and A1.b). Some of the sherds from Takht-i Suleiman (i.e., Takht.3 and Takht.6) were peculiar in that they were decorated by a combination of opaque yellow and white glazes. The white glazes were of either high lead-alkali or high lead types and were not opacified by tin oxide particles; instead, undissolved quartz grains and a small number of air bubbles were used to achieve semi-opacification (Fig. A1.c). The white glazes of Samarkand sherds were lead-alkali (14-19 wt% PbO, 7-10 wt% Na<sub>2</sub>O+K<sub>2</sub>O; calx Pb/Sn in range 3-5) and opacified by tin oxide particles, undissolved quartz grains and air bubbles (Fig. A1.d).

<Tables A1 and A2>

<Figure A1>

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## Captions

Table 1. *Composition of glass standards; measured values represent the average of three EDS analyses (wt%).*

Table 2. *Calx mixture recipes.*

Table 3. *Chemical compositions of raw materials (normalised to 100%).*

Table 4. *Recipes for replicate glazing mixtures (wt%).*

Table 5. *EDS analysis of Egyptian and Levantine bodies (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Table 6. *EDS analysis of Mesopotamian bodies (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Table 7. *EDS analysis of Egyptian and Levantine opaque yellow and white glazes (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Table 8. *EDS analysis of Mesopotamian opaque yellow and white glazes (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Table 9. *EDS analysis of lead-tin-oxide crystals (normalised to 100%).*

Table 10. *EDS analysis of green, brown and blue glazes (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Table 11. *Calculated chemical compositions of replicate glazing mixtures (wt%) and EDS analysis of replicate glazes (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Table A1. *EDS analysis of bodies of sherds from Northern Iran and Central Asia (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Table A2. *EDS analysis of yellow and white glazes from Northern Iran and Central Asia (wt%; obtained averaging three to five area measurements for each sample; normalised to 100%).*

Figure 1. Map showing location of sites from which ceramic samples were obtained.

Figure 2. Examples of Coptic Glazed Ware (CGW), Yellow Glazed Family (YGF) ware, opaque yellow and opaque white glazed wares; (a) CGW from Fustat (Ashmolean Museum, University of Oxford, acc. no. EA1974.48); (b) YGF sherd from Al-Mina (Victoria and Albert Museum, acc. no. C242A-1937); (c) bowl from Susa with green decoration on opaque yellow glaze (Musée du Louvre, acc. no. MAO S.576 - photo reproduced from Watson (2014, 141)); (d) bowl from Susa with green and brown decoration on opaque white glaze (Musée du Louvre, acc. no. MAO S.439 - photo reproduced from Watson (2014, 141)).

Figure 3. Calx firing schedule and stages of sampling.

Figure 4. Firing schedule for replicate glazes.

Figure 5. Backscattered SEM photomicrographs of Egyptian and Levantine bodies. (a) Fustat.1: showing a matrix of compacted aluminium-rich clay with a variety of non-plastic inclusions; (b) Al-Mina.6: showing a matrix of porous calcareous clay with a variety of non-plastic inclusions and microfossils; (c) Madaba.5: showing clay pellets embedded within a porous calcareous clay; (d) Raqqa.22-kiln rod: showing a variety of non-plastic inclusions as well as high lead glass fragments added to a porous calcareous clay. Q: Quartz, Px: Pyroxene, Pl: Plagioclase, Fld: Feldspar, Bi: Biotite, Mus: Muscovite, Ti-Mag: Titanium magnetite.

Figure 6. Backscattered SEM photomicrographs of Mesopotamian bodies. (a) Samarra.22: showing fine-grained, porous calcareous clay; (b) Samarra.26: showing phosphate inclusions in a fine-grained, porous calcareous clay matrix; (c) Basra.4: showing dispersed quartz and feldspar (plg) grains in a fine-grained porous calcareous clay matrix; (d) Samarra.5: showing coarse-grained, porous calcareous clay matrix showing a variety of non-plastic inclusion including added fragments of glass. Q: Quartz, Pl: Plagioclase.

Figure 7. Ternary plot of the normalised  $PbO-SiO_2-(Na_2O+K_2O+CaO+MgO)$  contents in tin-opacified yellow and white glazes from Egypt, the Levant and Mesopotamia (data from Tables 7 and 8).

Figure 8. Backscattered SEM photomicrographs of sections through yellow and white glazes; sherds (a) Al-Mina.7 (green-and-brown-on-yellow glazed ware): yellow glaze showing a scatter of lead-tin-oxide particles (white), air bubbles and undissolved quartz particles, as well as secondary crystals of wollastonite and diopside formed in the interaction layer; (b) Susa.7 (yellow relief-moulded ware): showing a scatter of well-formed lead-tin-oxide particles (white), quartz/feldspar particles (dark grey) and air bubbles in the glaze; (c) Raqqa.26 (plain white glaze): showing a scatter of tin oxide particles (white) and air bubbles, as well as secondary crystals of wollastonite and diopside formed in the interaction layer; (d) Samarra.26 (polychrome lustre ware): showing high concentration of tin oxide particles (white), air bubbles and quartz/feldspar particles (dark grey) distributed through the glaze; (e) Susa.2 (blue-on-white ware): white glaze showing a scatter of air bubbles and quartz grains (grey) through the glaze.

Figure 9. Backscattered SEM photomicrographs of section through green, brown and blue glazes: sherds (a) Al-Mina.3 (green-on-yellow glazed ware): showing fewer lead-tin-oxide particles (white) in upper green glaze layer as compared to the underlying yellow glaze layer; (b) Raqqa.25 (green-and-brown-on-yellow glazed ware): brown glaze showing quartz grains, hematite and

various oxidised forms of manganese and manganese-lead bearing particles – tin oxide particles scattered through glaze are barely visible; (c) Samarra.19 (blue-on-white glazed ware): showing double glaze layer with a scatter of cobalt-bearing minerals (white) in the outer blue layer, and quartz/feldspar particles (dark grey) and air bubbles throughout the glaze thickness; (d) Samarra.21 (blue-on-white glazed ware): showing double glaze layer with inner white alkali glaze containing a scatter of tin oxide particles (white), quartz/feldspar particles (dark grey) and air bubbles, and outer blue lead-alkali glaze. Q: quartz.

Figure 10. Scatter plots for body compositions;  $K_2O$  vs  $FeO$  and (b)  $K_2O$  vs  $MgO$ .

Figure 11. Calx samples C0.600, C7.600 and C30.600.

Figure 12. Glazes G1.850, G2.850, G3.850, G1.950, G2.950 and G3.950.

Figure 13. Backscattered SEM photomicrographs of sections through replicate glazes; (a) G1.850: yellow glaze showing lead-tin-oxide particles (white), with secondary crystals of wollastonite (grey) formed in the interaction layer, (b) G1.950: translucent brownish glaze showing no opacifying particles, but with secondary crystals of wollastonite (light grey) formed in the interaction layer; (c) G2.850: yellow glaze showing lead-tin-oxide particles (white), with secondary crystals of wollastonite (grey) formed in the interaction layer; (d) G2.950: white glaze showing scatters of tin oxide particles (white), with secondary crystals of wollastonite (grey) formed in the interaction layer; (e) G3.850: white glaze showing scatters of tin oxide particles (white); (f) G3.950: white glaze showing scatters of tin oxide particles (pale grey) as seen at higher magnification (x2300).

Figure A1. Backscattered SEM photomicrographs of sections through Northern Iranian and Central Asian glazes. Sherds (a) Merv.1 (brown-on-yellow glazed ware): yellow glaze showing scatters of lead-tin-oxide particles (white) throughout the glaze; (b) Takht.3 (yellow-and-green-and-brown-on white glazed ware): double glaze layer showing a proliferation of quartz grains and some air bubbles in white inner glaze, and lead-tin-oxide particles (white) scattered through yellow outer glaze ; (c) Takht.3 (yellow-and-green-and-brown-on white glazed ware): double glaze layer showing a proliferation of quartz grains through the inner white layer, with the outer transparent glaze being free of particles; (d) Samarqand.3 (green-on-white glazed ware): showing dispersed tin oxide particles, air bubbles and undissolved quartz grains throughout the white glaze.

#### **Online supplementary files**

Table S1. List of Sherds.

Fig. S1. Photographs of sherds.

Fig. S2. SR-Micro-XRD patterns corresponding to the crystalline phases in Fustat.1 glaze.

Fig. S3. XRD peaks for the calx C7.600.



Table 1

Jlass Standard	Values		SiO <sub>2</sub>	PbO	NaO <sub>2</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	MnO	FeO	P <sub>2</sub> O <sub>5</sub>	Cr <sub>2</sub> O <sub>3</sub>	CuO	As <sub>2</sub> O <sub>3</sub>	NiO	CuO	TiO <sub>2</sub>	ZnO	Sb <sub>2</sub> O <sub>3</sub>	Totals
	Measured	Avg																				
CORNING A	Most Probable Values*		66.56	0.12	0.19	14.3	2.87	5.03	2.66	1.00	1	1.21	0.13	0.001	0.17	nr	0.02	1.17	0.79	0.044	1.75	99.02
	Measured	Avg	67.54	bd	bd	14.28	3.20	5.17	2.70	0.85	1.02	0.94	bd	bd	0.23	bd	bd	1.21	0.98	bd	1.73	99.85
	Values	Sd	0.11	-	-	0.30	0.03	0.13	0.01	0.08	0.06	0.05	-	-	0.06	-	-	0.06	0.04	-	0.25	-
CORNING B	Most Probable Values*		61.55	0.61	0.04	17.00	1	8.56	1.03	4.36	0.25	0.38	0.82	0.005	0.046	nr	0.1	2.66	0.089	0.19	0.46	99.15
	Measured	Avg	62.46	0.73	bd	15.80	1.18	8.43	0.97	3.77	0.19	0.26	0.85	bd	bd	bd	bd	2.53	bd	bd	0.53	97.70
	Values	Sd	0.73	0.04	-	0.25	0.03	0.11	0.15	0.13	0.01	0.06	0.09	-	-	-	-	0.06	-	-	0.17	-
CORNING C	Most Probable Values*		34.87	36.7	0.19	1.07	2.84	5.07	2.76	0.87	0.82	0.38	0.14	0.001	0.18	nr	0.02	1.13	0.79	0.052	0.03	87.91
	Measured	Avg	34.11	35.85	0.52	0.91	3.10	5.17	2.47	0.70	bd	0.36	bd	bd	0.17	bd	bd	1.16	5.20	bd	bd	89.72
	Values	Sd	0.19	0.15	-	0.08	0.04	0.12	0.03	0.09	-	0.12	-	-	0.10	-	-	0.13	0.08	-	-	-
DLHI	Most Probable Values**		24.96	67.04	nr	1.01	0.96	1.04	0.33	3.98	nr	0.98	nr	nr	nr	nr	nr	nr	nr	nr	nr	100.30
	Measured	Avg	26.55	64.54	bd	0.92	0.98	1.02	0.23	3.82	bd	1.11	bd	bd	bd	bd	bd	bd	bd	bd	bd	99.17
	Values	Sd	0.39	0.44	-	0.11	0.09	0.06	0.09	0.05	-	0.08	-	-	-	-	-	-	-	-	-	-

\* Brill 1999.  
\*\* British Museum Research Laboratory, 1. Freestone, pers. comm., June 2014.

bd: below detection.  
nr: not reported.

Table 2

Calx mixtures	Pb:Sn	Pb (wt%)	Sn (wt%)
C0	0	-	100
C7	7	87.5	12.5
C30	30	96.8	3.2

### Table 3

[illegible]

Table 4

Glaze recipes	Silica	C7.600	C30.600	Frit	Calcium carbonate
G1	23.3	-	76.7	-	-
G2	38.7	61.3	-	-	-
G3	23.9	51.5	-	22.1	2.5

Table 5

Type	Sample	body fabric	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	FeO	P <sub>2</sub> O <sub>5</sub>	SO <sub>2</sub>	TiO <sub>2</sub>	PbO
Yellow and Yellow-and-Green/Brown Glazed Wares	Fustat. 1	silty/ m	65.8	23.4	1.2	0.7	0.7	1.4	4.1	0.5	0.3	1.8	bd
	Madaba.5	clay gr.	57.7	11.2	19.6	1.4	0.6	1.6	5.2	1.8	bd	0.9	bd
	Aqaba.1*	m	58.8	11.2	15.4	3.1	1.2	1.3	5.5	0.9	1.7	0.9	bd
	Aqaba.2*	m	61.5	10.8	11.9	3.1	1.3	2.1	5.2	0.8	2.1	0.9	bd
	Aqaba.4	m	57.7	14.5	9.9	3.2	2.2	2.3	5.3	0.8	3.0	0.8	bd
	Raqqa.5*	m	55.9	11.6	16.2	4.8	1.2	3.0	5.3	0.3	0.7	0.7	bd
	Raqqa.6*	m/c	53.4	11.0	19.3	4.6	1.5	2.9	5.3	0.3	0.6	0.6	bd
	Raqqa.8*	m	46.5	12.9	21.4	5.4	1.7	3.0	6.4	0.6	1.2	0.8	bd
	Raqqa.10*	m/c	50.7	11.8	18.3	5.2	1.5	3.3	6.1	0.4	0.6	0.7	1.4
	Raqqa.16	m	52.6	11.5	19.8	4.6	1.3	2.7	5.7	0.3	0.7	0.7	bd
	Raqqa.23*	m	49.3	11.7	20.2	5.3	1.5	2.9	5.8	0.3	0.6	0.7	1.7
	Raqqa.31	m	51.0	10.9	20.5	5.4	1.7	2.8	5.6	0.3	1.1	0.7	bd
	Al-Mina.3	f/m+m.foss.	53.0	10.6	15.3	9.2	1.0	2.2	7.3	bd	0.7	0.6	bd
	Al-Mina.4	m	49.8	11.3	23.3	5.0	1.0	2.2	5.8	0.3	0.3	0.6	bd
White and White-and-Green Glazed Wares	Aqaba.5	m	53.6	11.5	20.0	4.4	1.1	1.1	5.7	0.5	0.3	0.7	1.1
	Aqaba.7	m	56.3	10.8	16.3	4.3	1.4	1.8	5.8	0.5	1.1	0.9	0.8
	Al-Mina.9	m/c	64.5	7.8	14.5	2.0	0.9	1.3	4.9	1.2	bd	2.1	0.8
	Raqqa.1	m	48.9	11.3	21.9	6.0	1.7	2.7	5.4	0.5	0.9	0.7	bd
	Raqqa.3	m	50.3	11.9	20.1	5.5	1.6	2.6	5.4	0.4	0.5	0.6	1.1
	Raqqa.4*	m/c	51.4	11.8	18.3	6.3	1.5	2.4	5.9	0.3	0.4	0.6	1.1
	Raqqa.17*	m	46.4	13.5	20.0	6.5	1.5	3.0	6.5	0.5	0.6	0.8	0.7
	Raqqa.19*	m+cal	49.4	11.2	20.8	5.4	1.6	2.6	5.7	0.3	0.5	0.6	1.9
	Raqqa.20	m/c+glass	52.9	12.7	17.6	5.2	1.8	2.0	5.8	0.4	bd	0.7	0.9
	Raqqa.26	m+cal	48.9	11.9	20.7	5.2	1.4	2.5	6.5	0.4	0.4	0.9	1.1
	Al-Mina.5	m	51.6	11.5	20.5	3.9	1.3	1.5	5.4	2.8	0.2	0.6	0.7
Yellow Glazed with Painted Decoration	Raqqa.7*	m	50.6	11.5	19.1	5.2	1.5	3.0	6.0	0.6	0.9	0.8	0.8
	Raqqa.11	m/c	52.7	12.1	18.5	4.7	1.2	2.5	6.3	0.5	0.6	0.8	bd
	Raqqa.13*	m	50.2	11.9	19.9	5.3	1.5	2.8	5.9	0.4	0.6	0.7	0.8
	Raqqa.14	m	52.1	11.8	18.6	5.4	1.4	3.0	5.9	0.3	0.7	0.8	bd
	Raqqa.25	m	50.7	11.9	19.8	5.5	1.5	2.6	5.7	0.6	0.9	0.7	bd
	Raqqa.28*	m	50.2	10.8	20.9	4.2	1.3	3.0	5.8	0.8	0.7	0.9	1.4
	Raqqa.29	m	49.1	11.8	22.6	5.3	1.3	2.8	5.1	bd	0.9	0.8	bd
	Al-Mina.6*	m+m.foss.	55.3	10.1	15.9	7.8	1.0	1.9	6.4	0.4	0.5	0.5	bd
	Al-Mina.7	m	58.7	9.7	14.8	6.4	1.3	1.7	5.7	0.4	0.5	0.7	bd
Sparse decorated wares	Raqqa.12*	m	53.9	11.7	18.1	4.9	1.3	2.8	5.5	0.3	0.7	0.7	bd
	Raqqa.18	m	51.3	12.1	17.4	5.6	1.5	3.3	5.7	0.4	0.9	0.8	1.0
Kiln rod	Raqqa.22	m/c+glass	49.9	12.1	18.3	6.0	2.4	2.4	5.2	0.6	0.3	0.7	2.1
Miscellaneous	Al-Mina.8*	m	49.6	12.7	21.7	4.9	0.7	2.4	6.1	0.3	0.4	0.9	bd
Typical Standard Deviation			2.2	0.6	2.0	0.5	0.2	0.1	0.8	-	0.3	0.1	-

\* Samples without a glaze analysis.

m: medium-grained (inclusions between 50-200 µm)

c: coarse-grained (inclusions >200 µm)

bd: below detection

m.foss.: microfossils

glass: glass fragments

cal: calcite grains decomposed during firing

clay gr.: clay grains

Table 6

Type		Sample	Fabric	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	FeO	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	TiO <sub>2</sub>	PbO
Blue on White		Samarra.19	f	48.7	13.1	19.3	6.6	2.3	1.1	6.5	0.7	0.8	0.8	bd
		Samarra.20	f	47.5	12.5	21.3	7.7	2.1	1.1	6.3	0.4	0.3	0.8	bd
		Samarra.21	f	48.9	12.8	18.9	7.5	2.2	1.4	6.4	0.4	0.8	0.8	bd
		Samarra.22	f	49.2	13.1	19.0	7.5	1.6	1.0	6.9	0.3	0.2	1.1	bd
		Susa.2	f	50.6	12.3	19.5	6.9	1.9	1.4	6.2	0.2	0.4	0.6	bd
		Susa.3*	f	50.9	12.6	19.1	6.4	1.5	1.7	6.5	0.2	0.4	0.7	bd
Blue and Green on White		Susa.8*	f	48.9	11.6	22.6	6.2	1.1	1.7	6.4	0.3	0.5	0.7	bd
		Samarra.23	f	45.3	16.6	19.9	6.8	1.6	1.2	6.6	0.3	1.0	0.8	bd
Plain White		Samarra.24*	f	45.0	13.9	23.8	7.1	1.4	1.3	6.1	0.3	0.3	0.7	bd
		Samarra.29*	f	50.8	12.1	20.7	7.0	1.5	0.9	6.1	0.1	0.1	0.6	bd
		Samarra.32*	f	49.9	12.2	19.6	7.4	1.5	1.6	6.3	0.3	0.4	0.7	bd
		Samarra.6	f	46.2	12.4	22.2	7.4	2.5	1.1	6.4	0.2	0.9	0.8	bd
		Susa.11*	f	49.8	12.3	21.2	5.7	1.3	1.6	6.6	0.4	0.4	0.7	bd
		Susa.18	m	48.6	12.2	22.9	5.2	0.6	2.3	5.7	0.3	0.8	0.7	0.7
		Susa.19	f	50.2	12.0	20.9	6.6	1.2	1.2	6.5	0.4	0.2	0.7	bd
		Susa.15	f	49.0	11.8	21.4	6.9	1.9	1.2	6.4	0.2	0.6	0.7	bd
		Susa.17*	c	55.1	10.1	21.3	3.8	1.0	2.6	4.6	0.4	0.5	0.6	bd
Lustre		Samarra.2	f/m+m.foss.	54.9	11.4	16.9	6.2	1.6	1.5	6.2	0.2	0.4	0.6	bd
		Polychrome Samarra.26	f+phos	42.9	15.4	19.6	6.4	1.7	0.9	6.4	5.7	0.3	0.8	bd
		Samarra.35	f	49.6	12.4	19.2	7.2	2.3	1.4	6.2	0.3	0.7	0.7	bd
Splashed		Monochrome Samarra.37	f	50.5	12.3	19.1	7.4	2.2	1.2	6.0	0.3	0.3	0.7	bd
		Green Samarra.13	m	47.9	12.6	21.4	7.5	1.4	0.8	6.5	0.2	0.3	0.7	0.9
		Samarra.14*	m	47.6	12.4	21.6	7.2	0.9	1.7	6.2	0.2	0.6	0.6	0.9
		Samarra.5	m+glass	50.4	12.7	17.7	7.1	1.2	1.6	6.6	0.5	0.7	0.7	0.7
		Susa.13*	m/c	52.5	12.3	19.5	3.6	1.0	3.5	6.1	0.2	0.6	0.7	bd
		Turquoise and Black Basra.4	f+q/feld	51.7	11.1	18.2	6.9	2.6	1.3	5.6	0.2	1.7	0.7	bd
		Turquoise Basra.5	f+q/feld	52.3	11.2	18.9	6.5	2.0	0.8	5.7	0.3	1.7	0.6	bd
Relief Moulded (Yellow)		Susa.7	f	50.6	12.2	18.7	6.0	1.0	2.6	6.6	0.4	0.4	0.7	0.9
		Susa.12	f	47.6	11.3	22.5	6.8	0.6	2.6	6.2	0.4	0.7	0.7	0.7
Green & Brown on Yellow		Susa.1	f+phos	50.2	11.9	18.4	6.2	0.8	2.6	6.1	0.3	0.5	0.8	2.1
		Kish.1	m/c	56.0	13.0	13.2	6.3	1.4	2.7	6.1	0.5	0.1	0.7	bd
Kiln rods		Basra.8*	f	48.4	12.3	19.0	6.5	2.7	1.8	7.1	0.2	1.2	0.8	bd
		Basra.9*	f+q/feld	47.6	12.2	20.2	7.1	3.3	0.5	7.0	0.2	1.2	0.7	bd
Typical Standard Deviation				1.9	0.4	2.2	0.5	0.3	0.1	0.6	-	0.2	0.1	-

\*Samples without a glaze analysis.

f: fine grained.

m: medium grained.

c: coarse grained.

bd: below detection.

phos: phosphate inclusions.

m.foss.: microfossils

glass: added glass fragments.

q/feld: quartz or feldspar grains.

Table 7

Decoration	Sample	Body Fabric	Glass thickness (µm)	Interaction layer (µm)	Associated coloured glaze(s)*	Compositional Type	Opacification	SiO <sub>2</sub>	PbO	SnO <sub>2</sub>	Na <sub>2</sub> O+K <sub>2</sub> O	CaO+MgO	Na <sub>2</sub> O	K <sub>2</sub> O	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	P <sub>2</sub> O <sub>5</sub>	MnO	FeO	TiO <sub>2</sub>	Pb/Sn (ash)	
Yellow Glazes																							
Y: Green on yellow	Postat.1	slty/m	80	-	G	V High Lead	PbSn+sh(few)	27.7	65.3	2.7	0.2	0.3	0.2	bd	0.2	0.1	2.9	bd	0.1	0.6	bd	28.1	
Y: Green on yellow	Raqua.16	m	50	30	G	V High Lead	PbSn+sh+Q	25.2	65.7	3.6	0.5	2.5	0.2	0.4	2.0	0.5	1.3	bd	bd	0.9	bd	21.3	
Y: Green on yellow	Raqua.31	m	50	20	G	V High Lead	PbSn+sh+Q	29.9	62.5	1.6	0.5	2.6	0.2	0.2	2.2	0.4	1.5	bd	bd	1.1	0.2	44.9	
Y: Green on yellow	Al-Minu.3	f/m+cn.foss.	300	20	G	V High Lead	PbSn+Q+sh	25.1	69.1	3.6	0.1	1.0	0.2	bd	0.8	0.2	0.8	bd	bd	bd	22.4		
Y: Brown on yellow	Madaba.5	m	250	-	-	V High Lead	PbSn+Q+sh	33.2	61.2	1.6	0.4	1.8	bd	0.4	1.7	0.1	1.4	bd	bd	0.4	bd	45.1	
Y: Brown on yellow	Raqua.11	m/c	180	50	[Br]/G	V High Lead	PbSn+Q+sh(few)	28.5	66.7	1.9	bd	0.3	bd	bd	0.2	0.1	1.5	bd	bd	bd	1.0	40.5	
Y: Yellow and brown glazed	Raqua.14	m	70/200	40	[Br] & G	V High Lead	PbSn+Q+sh(few)	26.7	67.8	1.5	0.5	1.4	bd	0.5	1.2	0.2	1.0	bd	0.1	0.9	bd	53.7	
Y: Green and brown on yellow	Al-Minu.4	m	140	weathered	[Br] & [G]	V High Lead	PbSn+sh+Q	28.4	63.5	1.8	1.9	2.7	1.2	0.6	2.2	0.5	0.7	bd	bd	0.5	0.2	40.7	
Y: Green and brown on yellow	Raqua.25	m	80/200-600	40	Br & [G]	V High Lead	PbSn+Q+sh	28.0	62.4	2.5	bd	1.2	bd	bd	1.0	0.2	1.0	bd	4.5	0.3	bd	29.0	
Y: Green and brown on yellow	Raqua.29	m	150/300	20	[Br] & G	V High Lead	PbSn+Q+sh	26.9	66.5	2.6	0.6	2.1	0.2	0.3	1.7	0.4	0.5	bd	bd	0.6	bd	29.7	
Y: Yellow and green and brown glazed	Al-Minu.7	m	250	40	[Br] & [G]	V High Lead	PbSn+sh+Q	28.7	63.4	4.7	0.4	1.3	0.2	0.1	1.0	0.3	0.6	bd	0.1	0.6	bd	15.8	
Y: Sparse decorated (yellow and green and brown glazed)	Raqua.18	m	100/200	30	G	V High Lead	PbSn+sh+Q	26.9	48.4	1.7	1.5	9.8	0.4	1.0	8.8	1.0	1.3	8.6	bd	1.5	0.2	32.8	
Y: Plain	Aqaba.4	m	150	-	-	V High Lead	PbSn+sh+Q	24.7	67.0	4.5	0.7	1.6	0.4	0.3	1.4	0.2	1.1	bd	bd	0.4	bd	17.6	
Typical Standard Deviation								0.2	0.6	0.6	-	-	-	-	0.1	-	0.1	0.1	-	-	-	-	
White Glazes																							
W: Green on white	Raqua.3	m	100/250	30	[G]	High Lead-Alk	Sn+sh+Q	40.0	43.6	7.2	3.8	4.1	2.4	1.4	3.0	1.1	0.8	bd	0.1	0.4	bd	7.2	
W: Green on white	Raqua.20	m/c+glass	550	80	[G]	High Lead-Alk	Sn+sh+Q	45.2	36.9	7.2	4.6	4.5	2.6	2.0	3.5	1.1	0.9	bd	bd	0.6	0.1	6.0	
W: Green on white	Aqaba.5	m	400	-	[G]	High Lead-Alk	Sn+sh+Q	37.5	41.1	10.8	3.2	3.4	1.1	2.1	2.7	0.7	1.6	bd	0.1	2.3	0.1	4.5	
W: Green and purple on white	Al-Minu.5	m+glass	300	40	[G] & [P]	High Lead-Alk	Sn+sh+Q	38.4	43.5	8.1	4.7	3.9	2.3	2.4	2.9	1.0	0.6	bd	0.2	0.4	0.1	6.3	
W: Plain	Aqaba.7	m	400	-	-	High Lead-Alk	Sn+Q+sh	40.4	45.8	6.6	4.6	0.4	3.8	0.8	0.3	0.1	1.6	bd	bd	0.4	0.2	8.1	
W: Plain	Raqua.1	m	450	60	-	High Lead-Alk	Sn+sh+Q	37.7	46.3	7.2	3.8	3.3	2.9	0.9	2.3	1.0	0.5	bd	0.1	1.0	0.1	7.5	
W: Plain	Raqua.26	m+cal	250	60	-	High Lead-Alk	Sn+sh(few)	40.4	41.1	6.5	4.4	3.7	2.7	1.7	2.9	0.8	2.4	bd	0.2	1.2	0.2	7.4	
W: Plain	Al-Minu.9	m/c	350	40	-	High Lead-Alk	Sn+sh	36.0	44.2	12.2	2.8	2.3	1.8	1.0	1.8	0.5	1.9	bd	0.1	0.4	0.1	4.3	
W: Kiln rod**	Raqua.22	m/c+glass	300	80	-	High Lead-Alk	Sn+sh/sec. cry.	44.6	37.7	6.1	4.6	4.9	2.8	1.8	3.7	1.2	1.0	bd	bd	1.9	bd	7.2	
Typical Standard Deviation								0.3	0.8	-	-	-	-	-	0.1	0.1	-	-	-	0.1	0.2	0.1	-

\* See Table 10; glaze colours in brackets were not analysed (either weathered or not sampled)

\*\* indicates that glazes were applied on different sides of the sherd.

^ indicates that the glazes were applied on the same side of the sherd

/: Glaze thickness on different sides of sherd.

\*\* Splashes of opaque white glaze on kiln rod

bd: below detection

G: green glaze

Br: brown glaze

P: purple glaze

Y: yellow glaze

W: white glaze

m.foss.: microfossils

PbSn: Pb(Sn,SiO<sub>3</sub>) crystals

Sn: SnO<sub>2</sub> crystals

ab: air bubbles

Q: undissolved quartz grains

sec. cry.: secondary crystals

Table 8

Decoration	Sample	Body Fabric	Glaze thickness (µm)	Interaction Layer (µm)	Associated coloured glaze*	Compositional Type	Opacification	SiO <sub>2</sub>	PbO	SnO <sub>2</sub>	Na <sub>2</sub> O+K <sub>2</sub> O	CaO-MgO	Na <sub>2</sub> O	K <sub>2</sub> O	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	MnO	FeO	TiO <sub>2</sub>	Pb/Sn (calc)
Mesopotamian Productions [associated with fine-grained bodies]																					
Yellow Glazes																					
Y-Green and brown on yellow	Susa.1	f-phos	80	<10	[G] & [Br]	V High Lead	PbSa+ab (few)	27.9	62.3	3.3	2.0	2.4	0.6	1.4	1.9	0.5	1.0	0.2	0.5	0.4	22.2
Y-relief moulded	Susa.7	f	50	<10	-	V High Lead	PbSe+Q+ab	26.1	67.5	3.6	0.9	0.9	0.5	0.4	0.9	bd	0.4	0.1	0.4	0.2	22.3
Y-relief moulded	Susa.12	f	130	<10	-	V High Lead	PbSa+ab (few)	24.8	68.5	3.1	0.3	2.0	bd	0.3	1.6	0.4	0.7	bd	0.4	0.2	26.4
White Glazes																					
W-Plain	Susa.15	f	300/700	-	-	Alkali-Lead	Sn+Q+ab	68.4	4.7	5.2	10.3	8.8	5.5	4.8	5.7	3.2	1.4	0.4	0.7	0.1	1.1
W-Plain	Susa.19	f	500	<10	-	Alkali-Lead	Sn+Q+ab	70.2	4.3	2.2	11.9	8.4	6.9	5.0	5.1	3.2	1.9	0.4	0.6	0.1	2.3
W-Plain	Samarra.6	f	300/1000	-	-	Alkali-Lead	Sn+Q+ab	70.3	1.9	4.0	11.8	9.3	7.4	4.4	5.6	3.7	1.6	0.4	0.6	0.2	0.6
W-Blue on white	Samarra.21	f	700	-	[Bl]	Alkali-Lead	Sn+Q+ab	71.8	1.8	3.4	12.0	8.9	7.0	5.0	5.9	3.0	1.6	bd	0.5	bd	0.6
W-Blue on white	Samarra.22	f	700	-	[Bl]	Alkali-Lead	Sn+Q+ab	67.8	4.3	3.8	11.9	8.4	7.6	4.3	4.9	3.6	2.8	0.4	0.6	bd	1.3
W-Blue on white	Samarra.19	f	700	-	[Bl]	Alkali	Q+ab	72.2	bd	bd	14.3	8.6	9.0	5.3	5.6	3.0	3.2	0.2	0.6	1.0	-
W-Blue on white	Samarra.20	f	400/500	-	[Bl]	Alkali	Q+ab	73.5	bd	bd	13.3	10.2	7.8	5.5	5.9	4.3	1.7	0.2	0.4	0.6	-
W-Blue on white	Susa.2	f	1000	-	[Bl]	Alkali	Q+ab	77.0	bd	bd	11.9	7.7	7.7	4.2	5.0	2.7	2.2	0.3	0.2	0.7	-
W-Blue and green on white	Samarra.23	f	600/800	-	[G] & [Bl]	Alkali	Q+ab	72.1	bd	bd	15.0	8.9	9.4	5.6	5.3	3.6	2.3	0.3	0.6	0.7	-
W-Lustre	Samarra.26	f-phos	200/1000	-	[Lustre]	Alkali-Lead	Sn+Q+ab	60.4	8.6	9.5	10.7	7.7	6.9	3.8	4.5	3.2	1.9	0.4	0.8	0.1	1.1
W-Lustre	Samarra.35	f	400	-	[Lustre]	Alkali-Lead	Sn+Q+ab	69.6	4.7	3.6	11.2	8.2	6.3	4.9	5.0	3.2	1.7	0.2	0.7	0.1	1.5
W-Lustre	Samarra.37	f	600	-	[Lustre]	Alkali	Q+ab	74.7	0.7	bd	12.3	9.5	8.4	3.9	5.8	3.8	2.0	0.3	0.4	0.7	-
W-Splashed turquoise and black on white	Bavra.4	f-q/fidd	500/650	-	[Turq] & [Blk]	Alkali	Q+ab	71.5	bd	bd	13.3	10.8	7.9	5.4	6.9	3.9	3.3	0.2	0.1	0.7	-
W-Splashed turquoise on white	Bavra.5	f-q/fidd	350	-	[Turq]	Alkali	Q+ab	73.6	bd	bd	12.5	10.3	7.0	5.5	6.7	3.6	2.8	bd	bd	0.7	-
Typical Standard Deviation								0.4	0.8	0.7	-	-	-	-	0.1	-	0.2	-	0.1	-	-
Non-Mesopotamian/Different Local Productions (?) [associated with medium/coarse-grained bodies]																					
Yellow Glazes																					
Y-Green and brown on yellow	Kish.1	n/c	150	-	G & [Br]	V High Lead	PbSa+ab (few)	25.0	65.6	4.5	1.2	1.9	0.5	0.7	1.5	0.4	0.6	0.5	0.5	0.2	17.2
White Glazes																					
W-Plain	Samarra.2	f+m-m.foss.	350/550	20	-	High Lead-Alk	Sn+Q+ab	39.2	33.8	16.0	5.2	4.7	3.5	1.7	2.9	1.8	0.8	0.1	0.3	bd	2.5
W-Plain	Susa.18	m	150	-	-	High Lead-Alk	Sn+Q+ab	39.2	41.1	11.2	4.8	2.7	0.9	3.9	2.0	0.6	0.7	bd	0.3	bd	4.3
W-Splashed Green on white	Samarra.5	m-glass	300	<10	G	High Lead-Alk	Sn+ab (few)	37.9	45.6	5.9	3.2	5.1	1.5	1.7	3.8	1.3	1.3	0.1	0.8	0.1	9.1
W-Splashed Green on white	Samarra.13	m	350	20	G	High Lead-Alk	Sn+ab (few)	41.6	37.5	8.7	5.8	4.1	2.4	3.4	3.1	1.0	1.7	bd	0.5	0.1	5.1
Typical Standard deviation								0.3	0.7	0.5	-	-	-	0.1	0.1	-	-	0.1	0.3	0.1	-

\* See Table 10; glaze colours in brackets were not analysed (either weathered or not sampled)

"/" indicates that glazes were applied on different sides of the sherd

"/&" indicates that the glazes were applied on the same side of the sherd

/ indicates glazes on different sides of sherd.

bd: below detection

G: green glaze

Br: brown glaze

Bl: Blue glaze

Blk: Black glaze

Turq: Turquoise glaze

Y: yellow glaze

W: white glaze

PbSn: Pb(Sn,Si)O<sub>3</sub> crystals

Sn: SnO<sub>2</sub> crystals

ab: air bubbles

Q: undissolved quartz grains

m.foss.: microfossils



**Table 9**

<b>Sample</b>	<b>SiO<sub>2</sub></b>	<b>SnO<sub>2</sub></b>	<b>PbO</b>
<b>Al-Mina.3</b>	8.2	29.0	62.8
<b>Raqqa.10</b>	5.6	31.3	63.1
<b>Raqqa.11</b>	9.1	24.4	66.5
<b>Raqqa.13</b>	5.5	26.9	67.7
<b>Raqqa.23</b>	5.7	27.6	66.8
<b>Raqqa.28</b>	6.8	30.1	63.2
<b>Raqqa.29</b>	5.5	27.8	66.7
<b>Susa.7</b>	5.4	33.4	61.2
<b>Susa.12</b>	5.1	29.0	65.9
<b>Average</b>	6.3	28.8	64.9

Table 10

Decoration	Sample	SiO <sub>2</sub>	PbO	SnO <sub>2</sub>	CuO	FeO	MnO	Na <sub>2</sub> O+K <sub>2</sub> O	CaO+MgO	Na <sub>2</sub> O	K <sub>2</sub> O	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	ZnO	As <sub>2</sub> O <sub>3</sub>
Green glazes																
Y- Yellow and green on yellow	Fustat.1	26.9	61.3	bd	3.4	1.0	bd	0.5	2.7	0.4	0.1	2.4	0.3	4.1	bd	bd
Y- Green on yellow	Raqq.16	24.6	66.9	2.7	2.2	0.6	bd	0.3	1.8	bd	0.3	1.5	0.3	0.6	0.3	bd
Y- Green on yellow	Raqq.31	27.3	61.3	0.9	4.9	1.0	0.1	0.3	2.6	bd	0.3	2.0	0.6	0.7	0.2	0.6
Y- Green on yellow	Al-Mina.3	26.3	66.9	1.9	1.7	0.8	0.1	0.8	1.0	bd	0.8	0.8	0.2	0.4	bd	0.2
Y- Yellow and brown glazed	Raqq.14	24.5	66.5	bd	3.5	1.0	bd	1.2	1.4	0.2	1.0	0.9	0.5	0.6	1.3	0.1
Y- Green and brown on yellow	Raqq.29	28.5	64.6	bd	2.5	1.1	bd	0.3	2.0	bd	0.3	1.7	0.3	0.6	0.2	0.2
Y-Green and brown on yellow	Kish.1	24.6	65.6	4.6	1.1	0.4	0.3	0.8	1.7	0.3	0.5	1.3	0.4	0.6	0.3	bd
W-Splashed Green on white	Samarra.5	36.8	45.1	6.1	1.2	0.8	0.1	2.7	5.4	1.3	1.4	3.7	1.7	1.5	bd	0.3
W-Splashed Green on white	Samarra.13	38.1	43.9	5.2	1.7	0.7	0.2	4.3	3.8	2.0	2.3	2.9	0.9	1.4	0.4	0.3
Brown glazes																
Y- Green and brown on yellow	Raqq.25	22.8	66.4	2.1	0.6	0.9	5.6	0.1	1.2	bd	0.1	1.0	0.2	0.3	bd	0.1
Blue glazes																
W-Blue on White	Samarra.19	65.4	bd	bd	0.3	9.9	0.2	14.2	5.6	8.8	5.4	3.7	1.9	3.3	1.1	bd
W-Blue on White	Samarra.20	68.7	3.0	0.6	0.2	2.4	0.2	13.1	8.6	8.1	5.0	5.2	3.4	2.3	0.9	bd
W-Blue on White	Samarra.21	62.6	17.7	0.7	0.2	4.2	bd	7.0	4.9	2.8	4.2	3.7	1.2	1.8	0.9	bd
W-Blue and green on white	Samarra.23	62.8	1.4	0.5	0.2	8.7	0.1	14.2	8.2	9.1	5.1	5.4	2.8	3.1	0.8	bd
W-Blue on White	Susa.2	72.9	bd	bd	0.2	4	0.2	12.6	7.1	7.9	4.7	4.8	2.3	2.2	0.6	0.2

Y: yellow glaze  
W: white glaze  
bd: below detection

Table 11

Sherd No.	SiO <sub>2</sub>	PbO	SnO <sub>2</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	FeO
<b>G1.calculated</b>	23.0	73.7	3.0	-	-	0.1	-	0.1	0.1
<b>G1.850</b>	24.4	69.9	2.9	bd	bd	1.6	bd	0.9	0.3
<b>G1.950</b>	25.3	68.3	3.0	bd	bd	1.9	bd	1.1	0.4
<b>G2.calculated</b>	38.3	52.2	9.1	-	-	0.1	-	0.2	0.1
<b>G2.850</b>	39.3	49.9	8.9	bd	bd	1	bd	0.7	0.2
<b>G2.950</b>	40.1	49.1	8.7	bd	bd	1.1	bd	0.8	0.2
<b>G3.calculated</b>	39.4	43.9	7.6	3.7	0.2	4.2	0.1	0.8	0.1
<b>G3.850</b>	42.0	39.8	7.4	3.8	0.3	4.4	0.5	1.7	0.1
<b>G3.950</b>	42.9	38.4	7.2	4.0	0.3	5.1	0.4	1.6	0.1

bd: below detection.

Table A1

Sample	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	FeO	P <sub>2</sub> O <sub>5</sub>	SO <sub>3</sub>	MnO	TiO <sub>2</sub>	PbO
Nishapur.1	62.7	13.7	6.7	2.5	1.8	3.1	5.2	1.1	2.1	0.1	0.9	bd
Nishapur.2	60.8	13.7	10.0	3.3	1.7	3.1	5.0	0.5	1.1	0.1	0.7	bd
Nishapur.4*	60.7	13.3	9.5	4.0	1.7	2.7	5.6	0.8	0.8	0.1	0.6	bd
Nishapur.9	61.2	13.5	10.7	3.6	1.6	2.9	4.6	0.5	0.7	0.1	0.6	bd
Merv.1	55.7	14.0	14.8	3.7	1.2	3.1	5.5	0.4	0.2	0.1	0.6	0.5
Merv.2*	53.3	15.6	15.9	3.9	2.1	1.5	5.7	0.5	0.6	0.1	0.7	bd
Merv.3*	53.7	15.4	15.2	3.7	2.4	2.9	5.3	0.4	0.3	0.1	0.6	bd
Takht.2	63.6	13.9	8.6	3.2	2.2	3.1	4.8	bd	bd	bd	0.6	bd
Takht.3	58.9	14.5	13.0	3.0	1.9	2.7	5.1	0.3	bd	bd	0.6	bd
Takht.5	62.1	14.3	9.2	3.2	2.3	3.07	4.7	0.4	bd	bd	0.7	bd
Takht.6	56.0	15.6	15.0	3.0	bd	3.1	5.9	0.2	0.2	0.1	0.9	bd
Samarqand.1	61.8	11.4	13.0	4.4	1.2	2.7	4.0	0.5	0.2	0.1	0.7	bd
Samarqand.3	62.4	12.0	10.9	5.2	1.2	2.6	4.3	0.4	0.2	0.1	0.7	bd
Samarqand.6	64.3	12.2	8.8	4.3	1.4	2.6	4.6	0.5	0.3	0.1	0.7	bd

\* Sherds for which there is no glaze analysis.

bd: below detection

### Table A2

[illegible]

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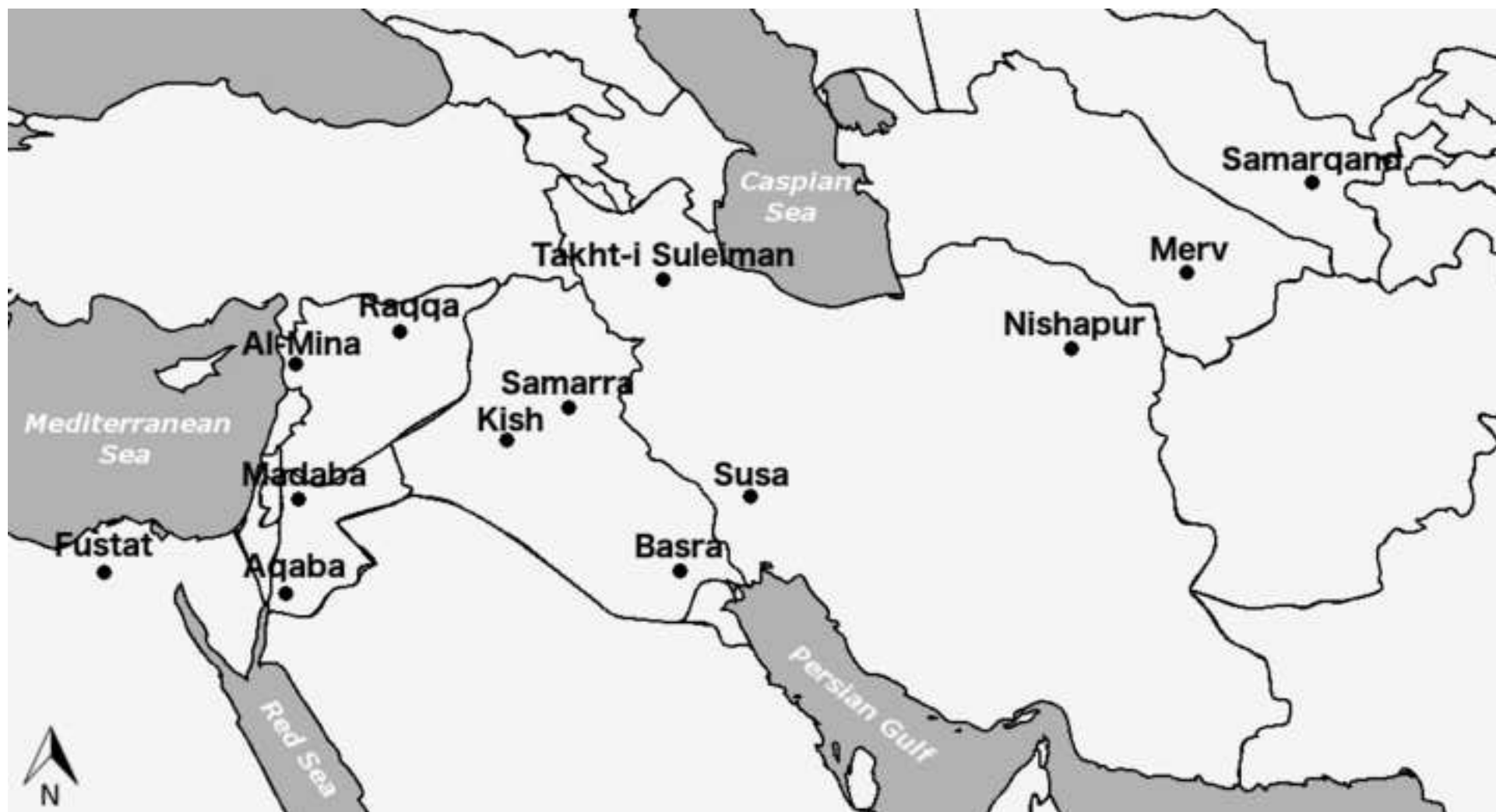


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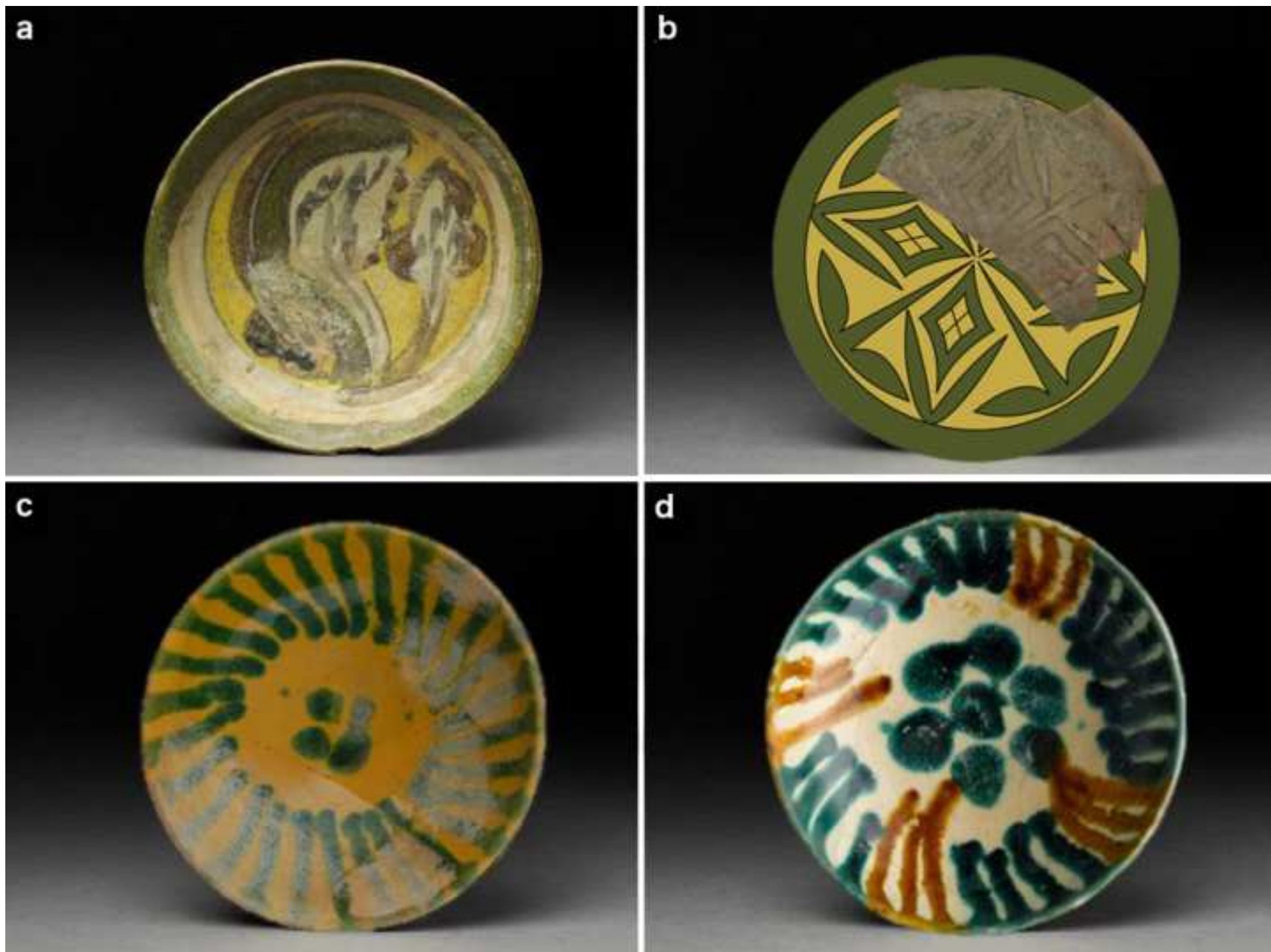


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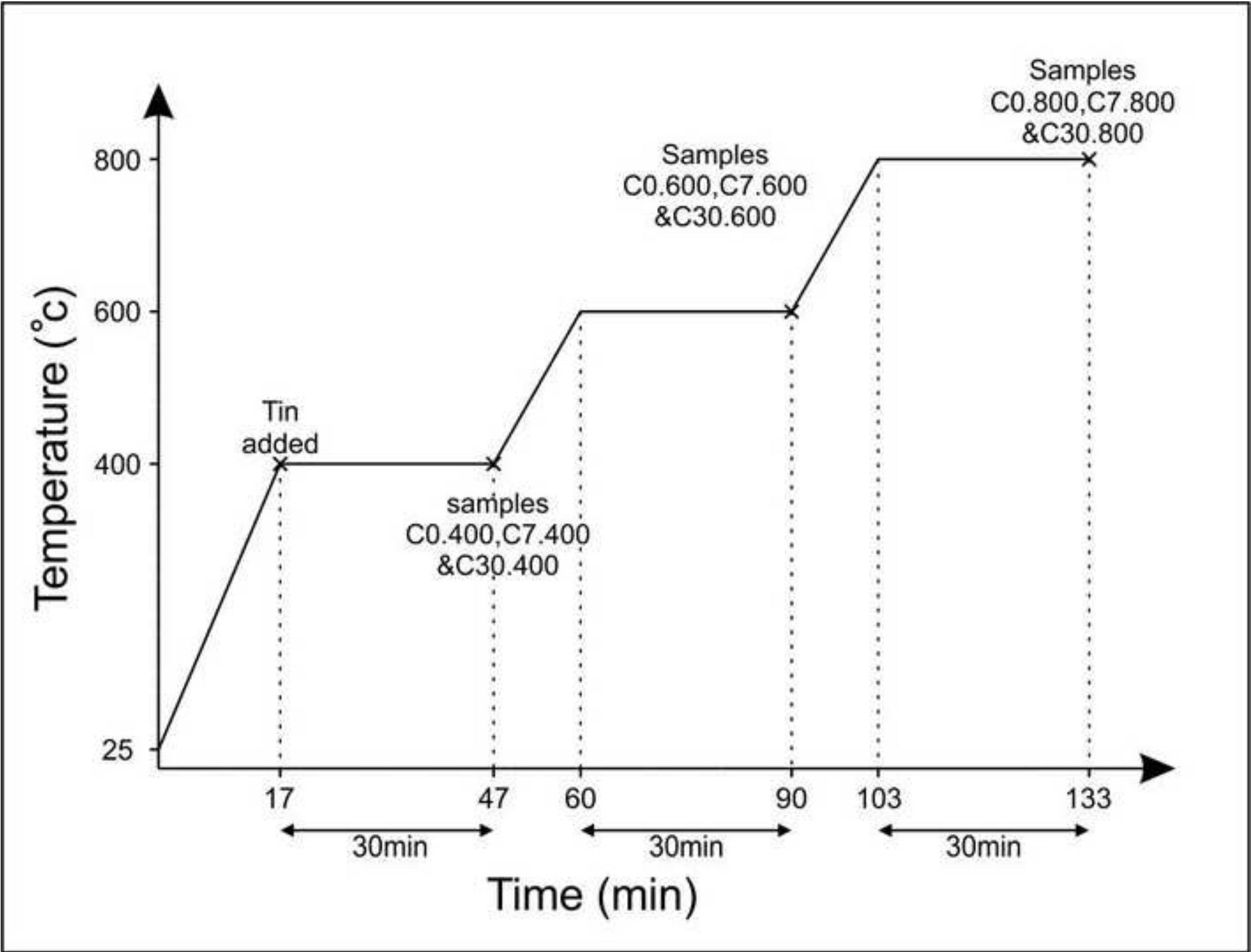




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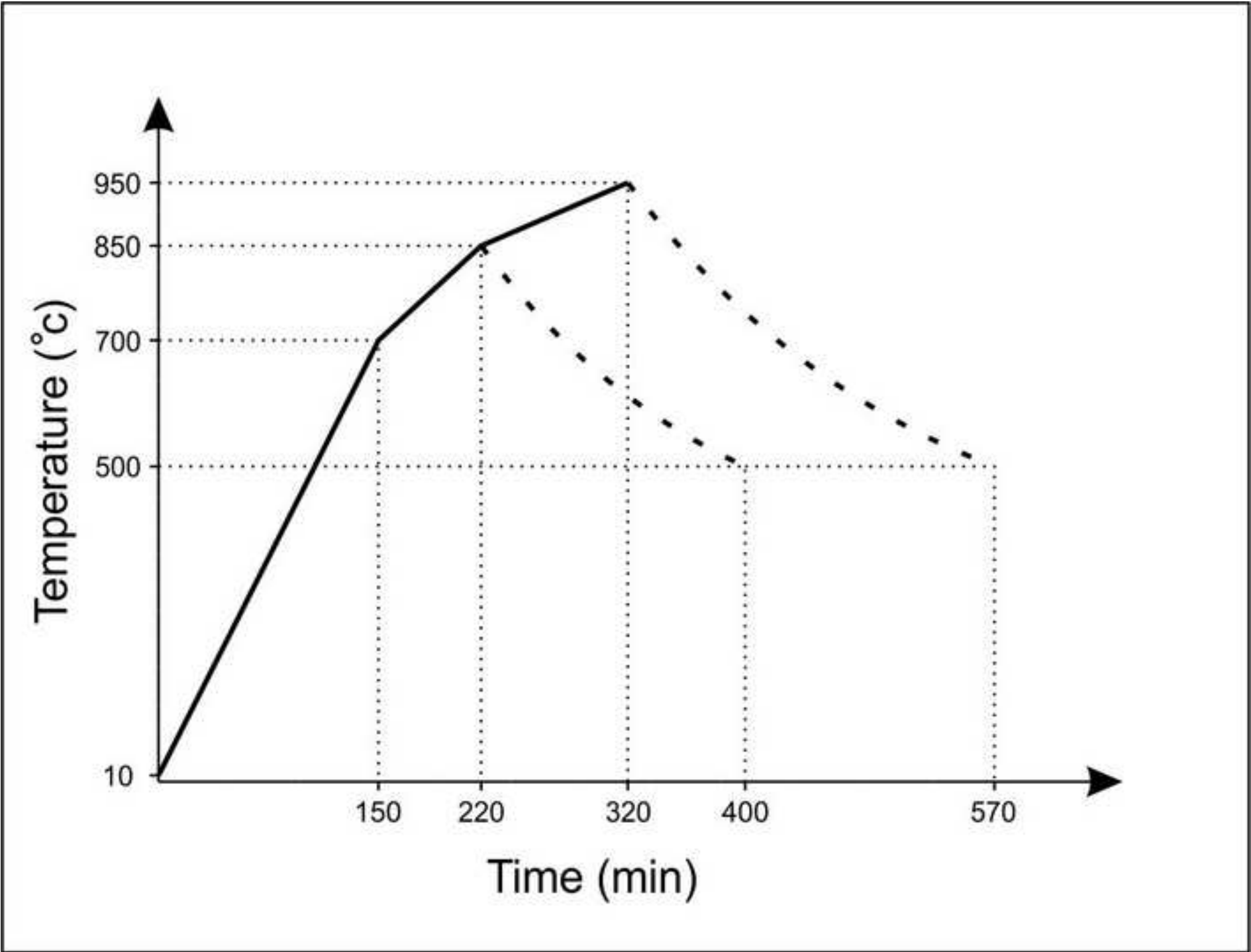


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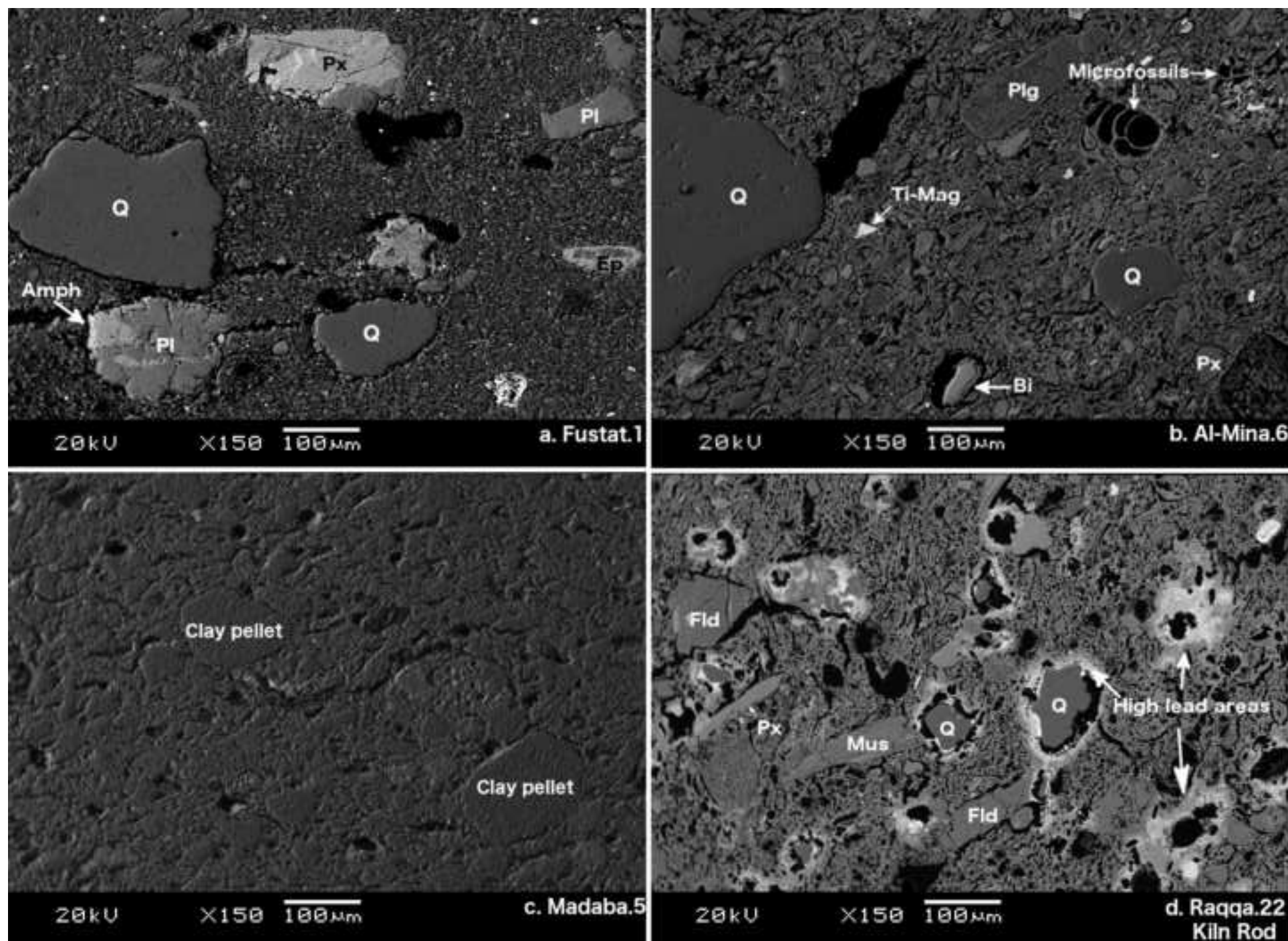




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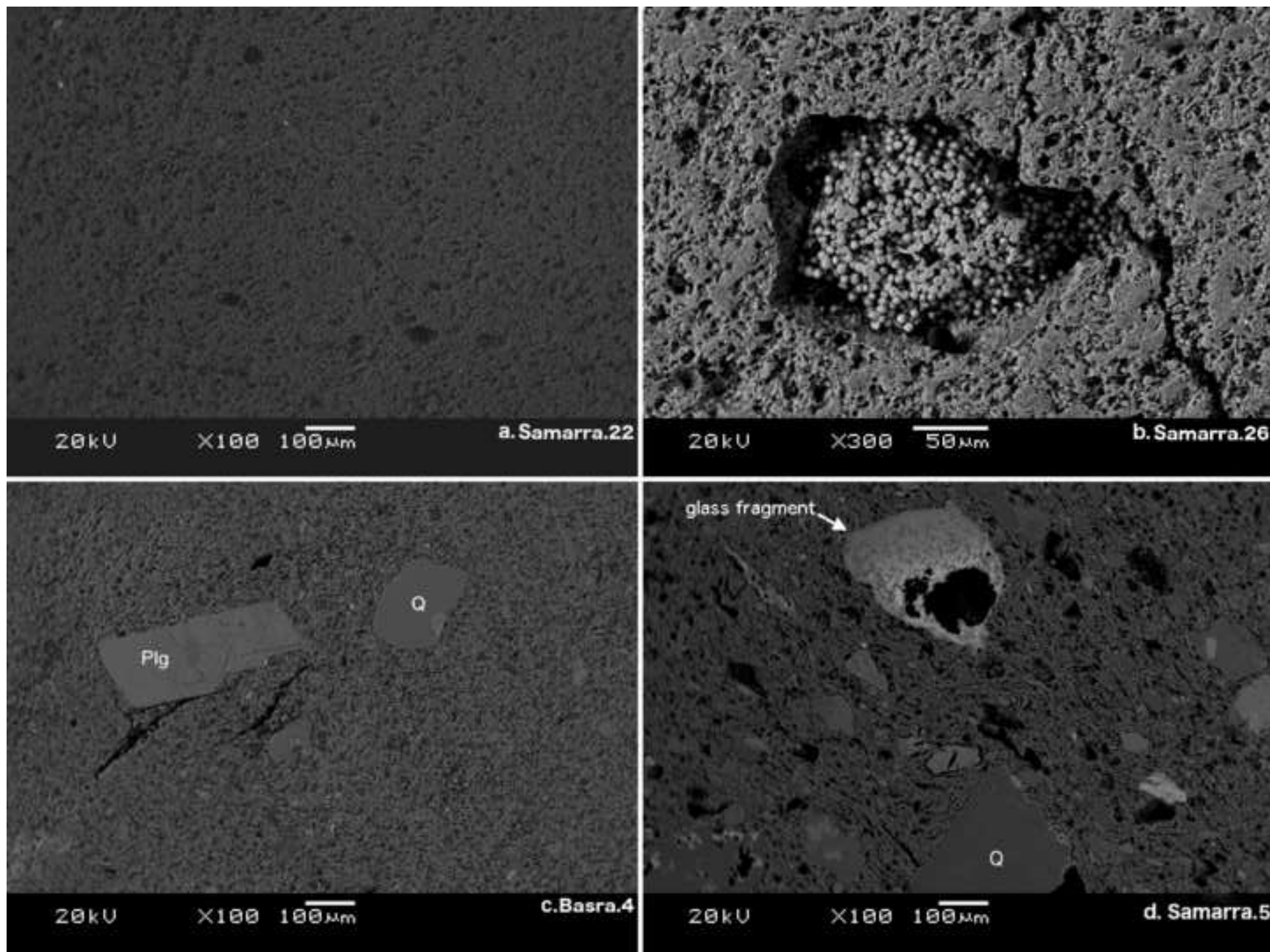


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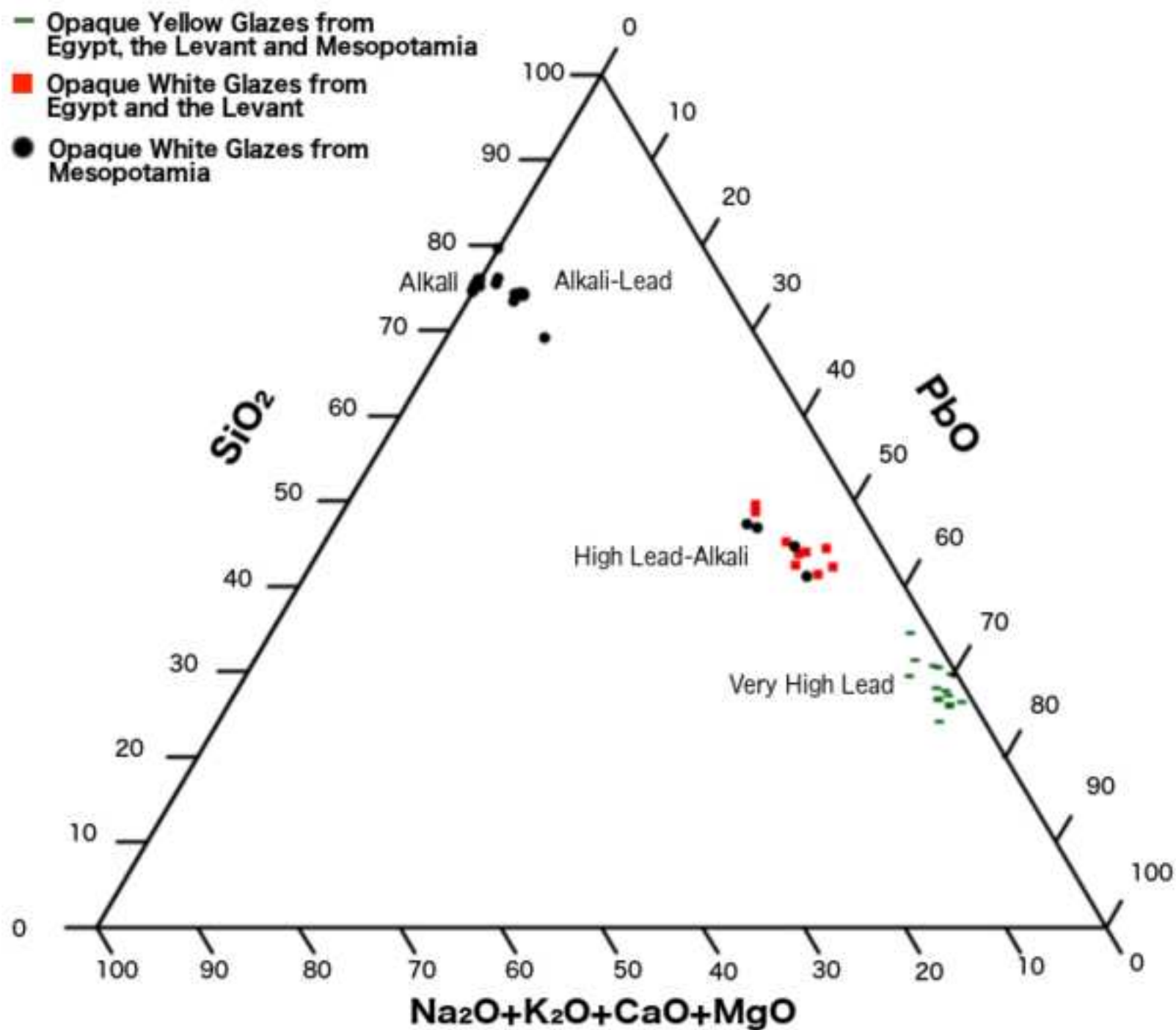


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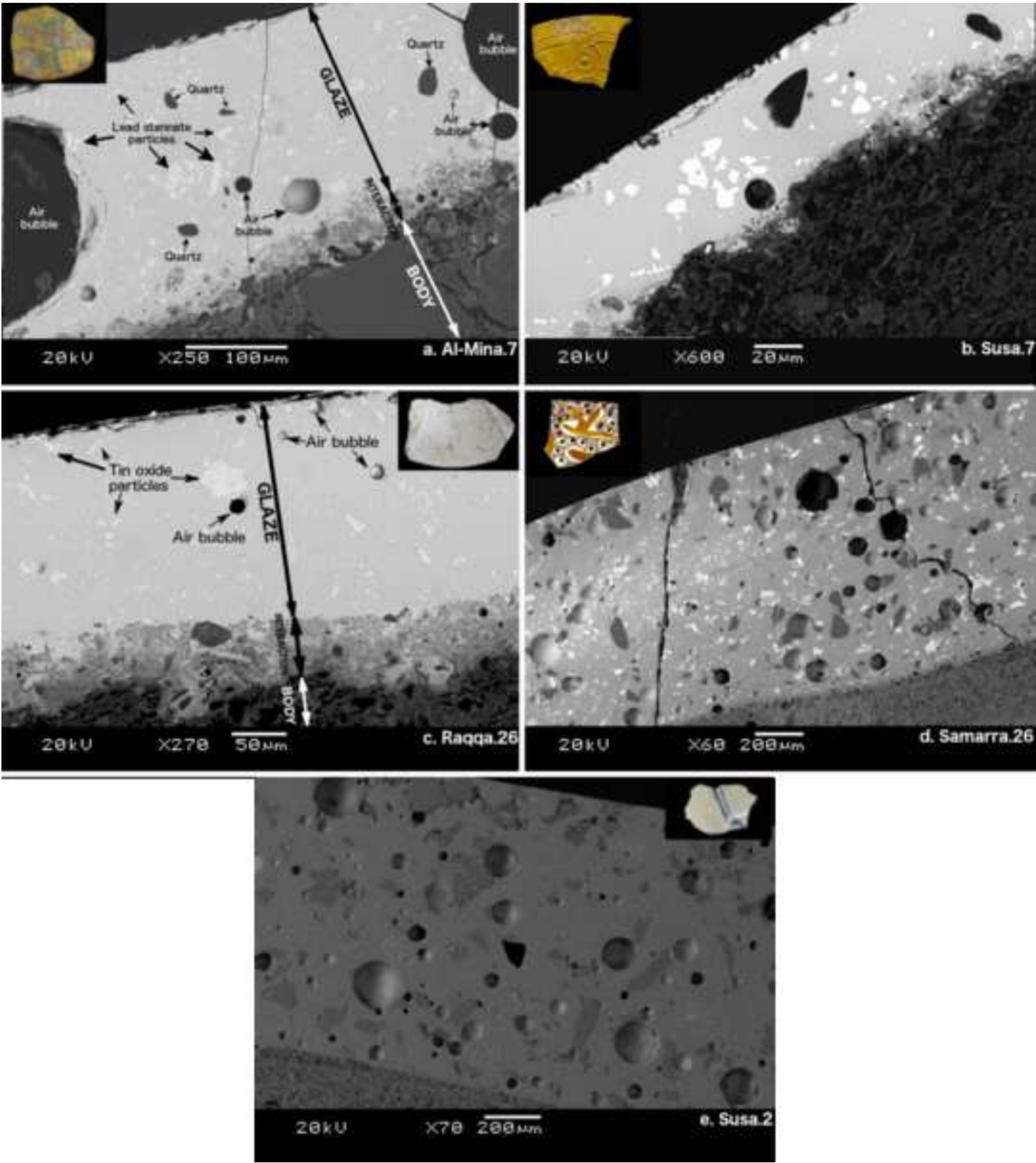




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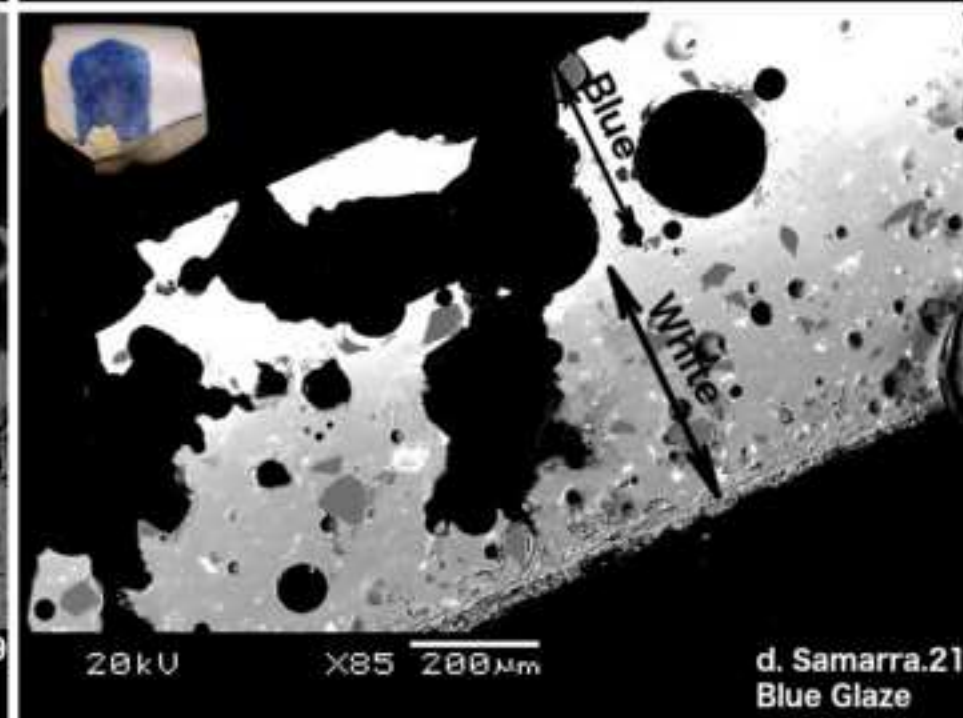
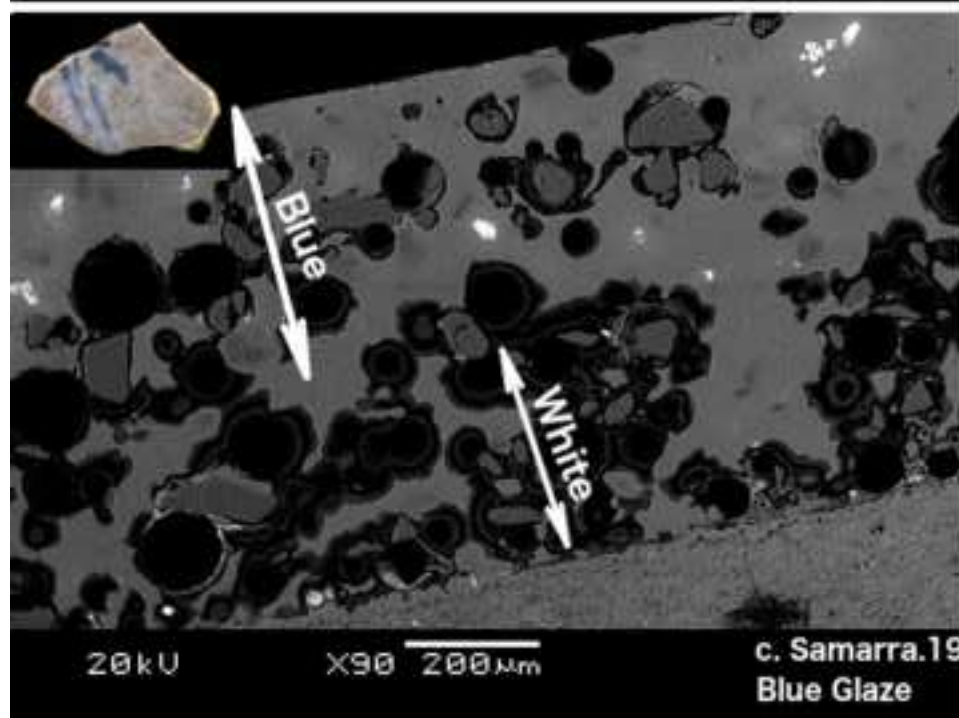
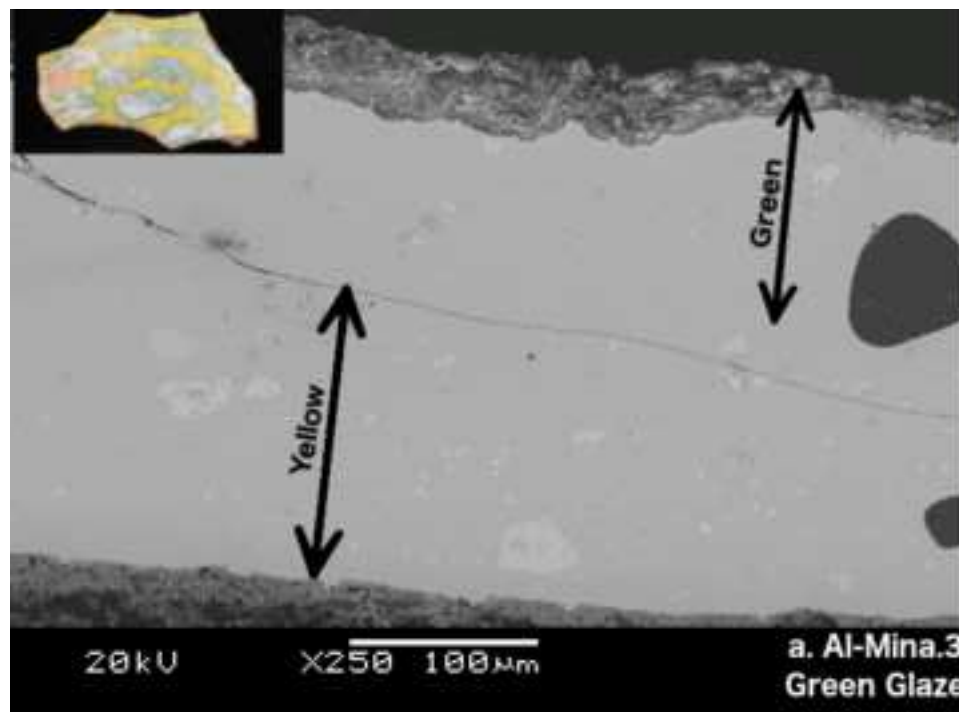


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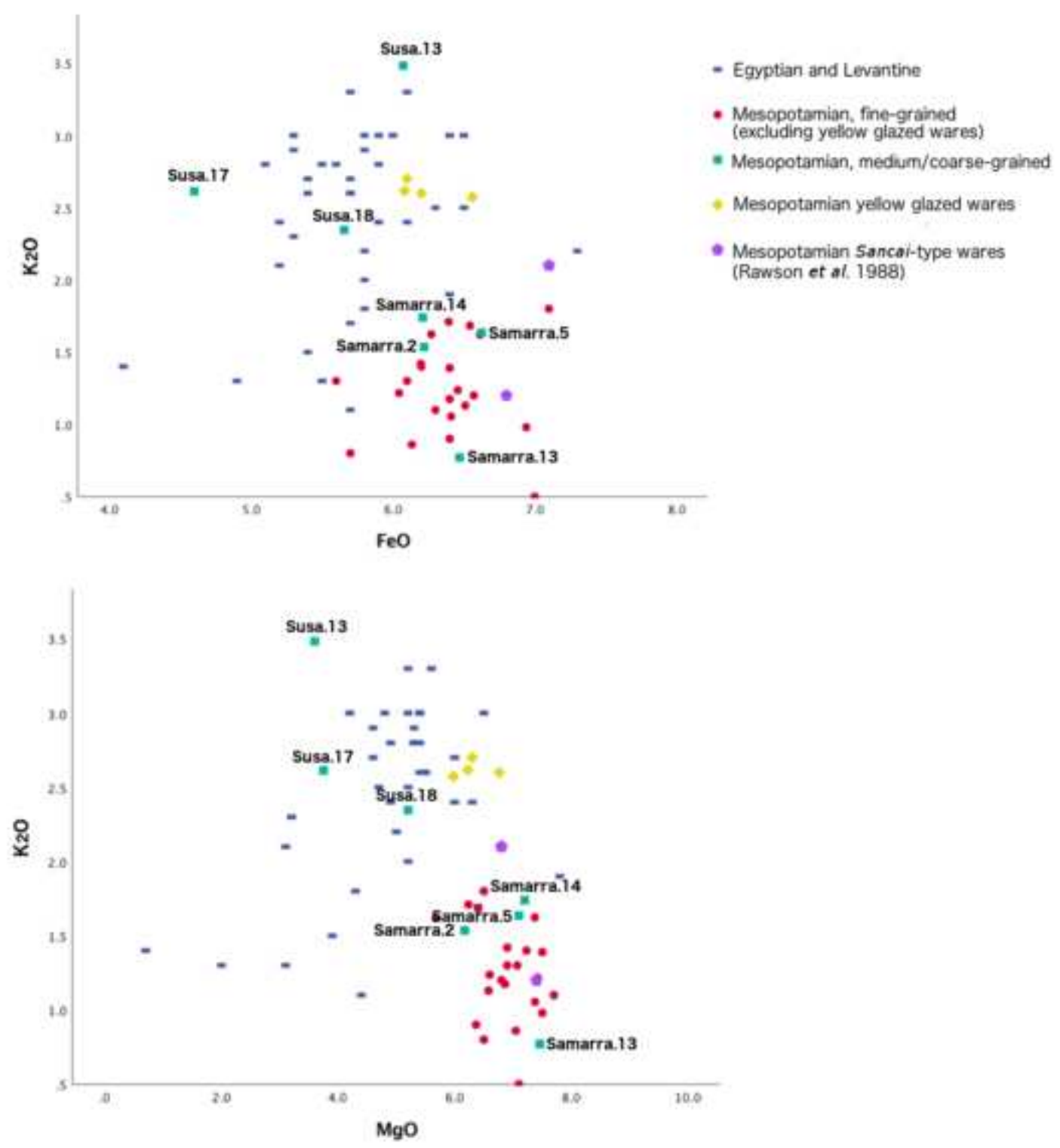


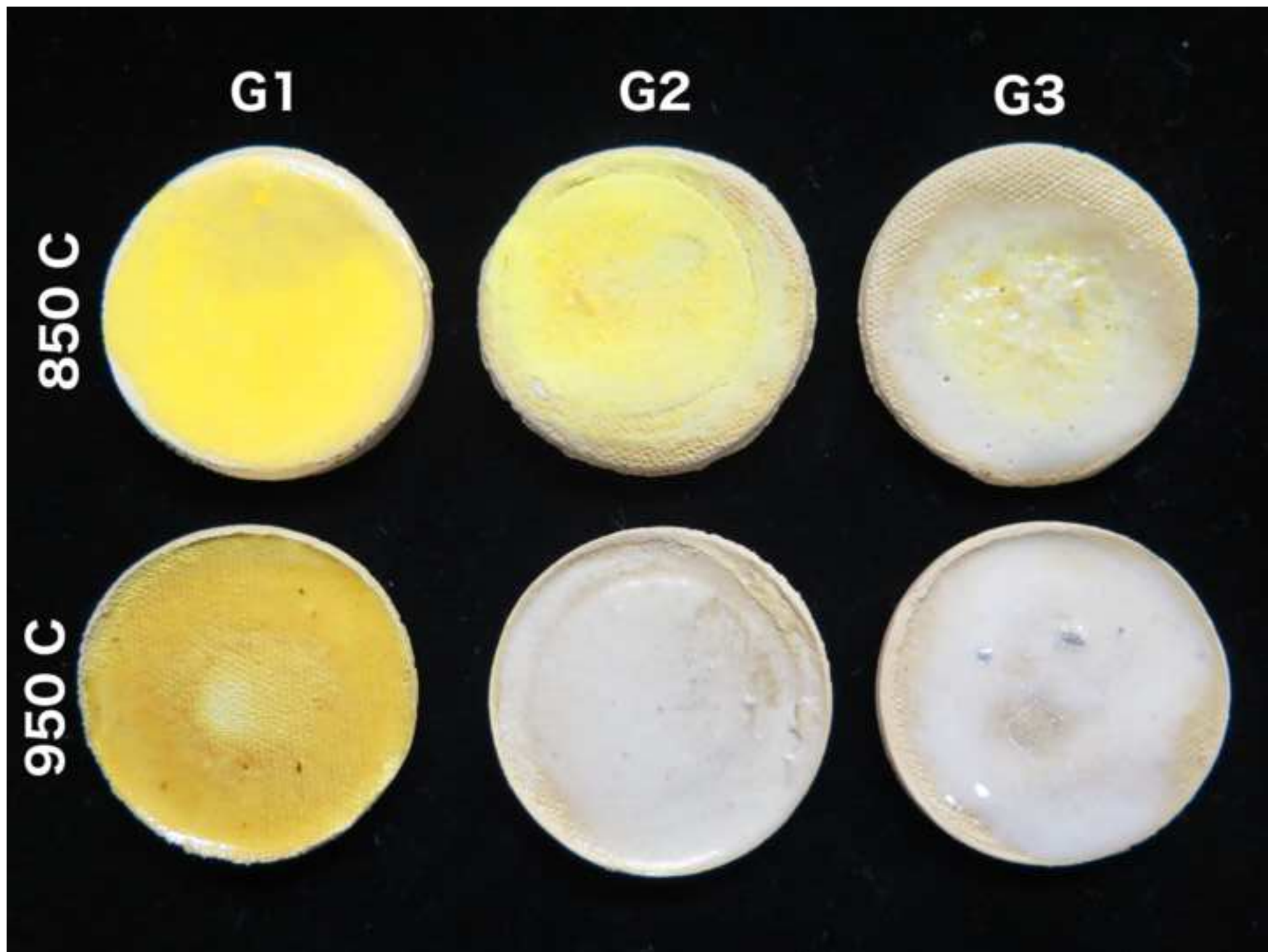
Figure 11

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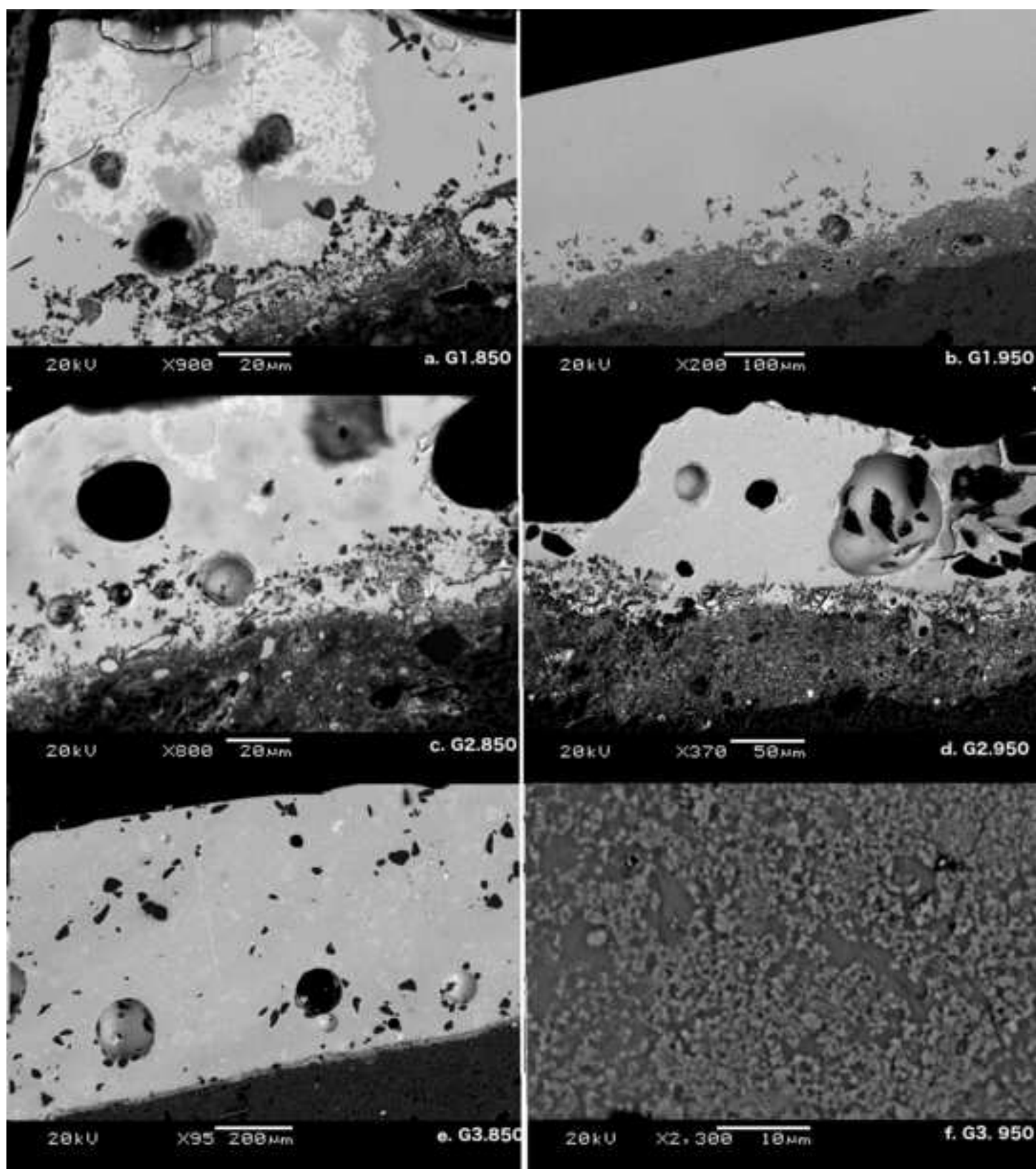
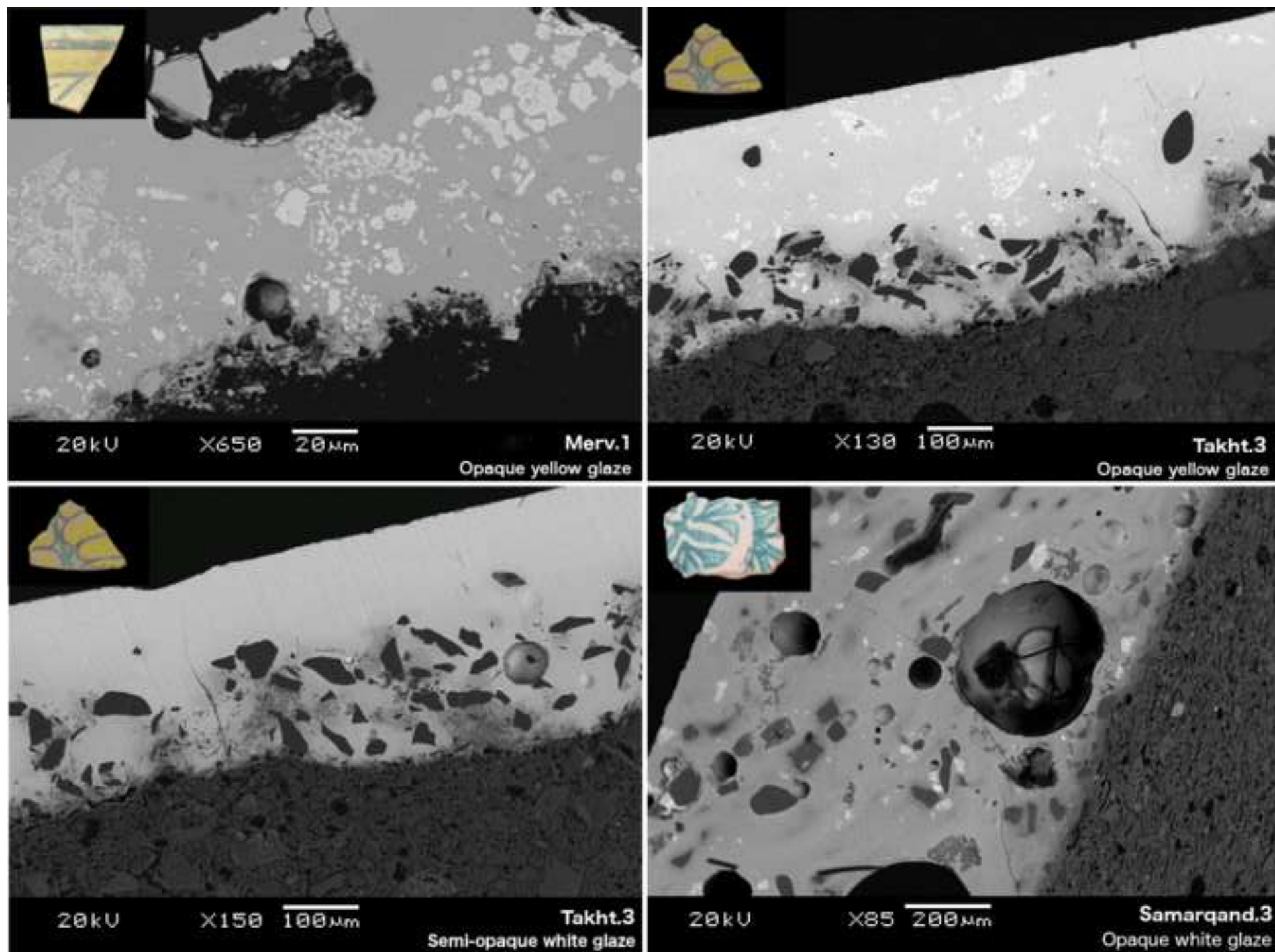


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