

## Fired shards from selected ancient Anatolian ceramics: a brief review of their mineralogical nature and pedological–microstructural evolution

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**Abstract:** This review is concerned with some key features observed within grouped samples of ancient ceramics that provide important evidence concerning the selection of appropriate raw materials and the evolution of the firing technologies employed in their production. It also demonstrates the importance of distinguishing and accounting for the mineralogical and microstructural attributes and identifying the processes responsible for the microstructural evolution of these ancient ceramics. The mineralogy largely reflects both the nature of the raw materials used and the maximum temperatures achieved during firing, deduced from the presence of specific high temperature minerals (HTMs). The microstructural evolution processes, deduced by the micromorphology of these ancient ceramics and displayed in specific features, observed in both the matrix and the slip of these ceramics, are largely controlled by the firing methods used in manufacturing. Thus, we conclude that the application of micromorphological principles, methods, and observations derived from the broad sphere of “pedology” to the study of ancient ceramics, provides valuable insights into the independent evolution of ceramic production methods in ancient societies. Thus, most of the observations recorded here concern identification of the raw materials used to make ancient ceramics and the firing processes used in their manufacture. Our data demonstrate that these ancient potters made use of a variety of temper materials (quartz and chaff, together with fragments of locally available rocks and minerals) that are now preserved in the matrix. Furthermore, analyses of the micromorphological attributes displayed by these ancient ceramics are helpful in determining and explaining the shrinkage features (stress coatings or poro-striated b-fabrics) and the preferred orientation of the elongated pores that have developed after firing in poorly controlled and slow-fired furnaces. In this regard, we finally seek to develop a useful data library ultimately targeting the enhancement of simulated ancient ceramic/pottery production, with an overall objective to apply the mineralogical and pedological properties of Anatolian ceramics researched in this work to globally selected shard specimens. Postburial processes, such as the illuviation-deposition of clay minerals to form the observed clay coatings, probably operated during the wet-dry cycles associated with mid-late Holocene climatic fluctuations. Accompanying calcification-decalcification processes, which may result from an intra cramic leaching-deposition of carbonate present in the source material of the pottery, are also consistent with the known wet-dry cycles of the mid to late Holocene pedogenesis (soil formation) episode and is reflected in the clay coatings and further attested by coeval changes in the soil-faunal activity.

**Key words:** Ancient ceramics, fired shards, mineralogy, micromorphology, pedology, raw material sources

### 1. Introduction

A seminal review of methods of scientific analysis of the materials used in the manufacture of ancient ceramics (Peacock, 1970) led to a significant expansion of studies devoted to such artefacts, most of which were based on criteria and concepts used for typographic descriptions

of modern equivalents. Independently, the introduction of the term “Geoarchaeology” embracing petrology and mineralogy became broadly accepted as a subdiscipline of archaeology (Davidson and Shackley, 1976) and was further advanced by studies that incorporated petrological attributes into ancient pottery research (Peacock, 1977;

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Williams, 1983; Vince, 1984; Whitbread, 1986; Whitbread, 1995; Williams and Vince, 1997).

More broadly, a pioneering petrographic study (Whitbread, 1986) that described and assessed the genetic significance of clay inclusions in archaeological materials also assisted in the task of determining the raw material sources of ancient pottery. Furthermore, with regard to the general interpretation of ceramic fabrics, the Vienna System Framework successfully pioneered the adoption of a terminology that accurately describes the range of fabrics/properties encountered in ceramic materials, ancient and modern (here termed “microstructure”). The present account adopts a mix of pedological-micromorphological terminology (FitzPatrick, 1993; Stoops, 2003) in the text, due to the wide range of properties, including matrix colour and thin section petrography, used to classify ancient pottery (Nicholson and Shaw, 2000) (via the United Kingdom Thin Section Database: UKTS).

Efforts to define the microstructure of Medieval and younger ceramics from Britain represent important milestones in the scientific study of ancient pottery by comparing their petrographic attributes (Vince, 2001). Since 1988, the protocols introduced by the independent Prehistoric Ceramics Research Group (PCRG), using a naked-eye approach to pottery description, have been significantly augmented to incorporate petrologic supplements (PCRG, 2010). A related study combined conventional archaeological naked-eye inspection with stereomicroscopic and submicroscopic observations (Akça et al., 2009a). This study sought to obtain details of form, texture and porosity, colour (Munsell Soil Color Charts), temper (chaff and mineral), and degree of firing in ceramics (Akça et al., 2009a). The estimation of the proportions and distribution of coarse and medium size minerals (grit), shaping (smoothness) and hardness were conducted according to the guidelines presented in the PCRG occasional papers (Akça et al., 2009a).

Moreover, X-ray powder diffraction analyses of the minerals were utilised to complement the limited description of the older (Neolithic) shards that display evidence of degraded morphology, when compared with shards from later periods (Akça et al., 2009a). Furthermore, the pore morphology and matrix modifications (Maniatis and Tite, 1981), utilising terminology familiar to archaeologists (PCRG, 2010), also have been taken into consideration in this study. In addition, a published study detailing the genetic sequences of new minerals formed in ceramic materials under increasing temperature conditions, has contributed significantly to our understanding of ancient furnace technologies and programs for the materials considered here (Maggetti et al., 2011).

Further efforts to identify and describe fabric properties in ancient pottery were undertaken in the Central

Mediterranean Project (FACEM), in which a web-based information Database (UKTS Database) was developed, as described in consecutive reports from the Institute of Classical Archaeology of the University of Vienna. These reports were adopted as the source of the protocols for the microscopic examination of pottery fabrics and were based primarily on stereomicroscopic observations, coupled with petrographic analysis. The relevant information was released in six consecutive reports, published from 2011 to 2018 ([www.facem.at](http://www.facem.at)). These reports were primarily based on descriptions from thin-sections of ancient ceramics that provide detailed information on the type of organic residues and the mineral constituents (size, type, and distribution).

An important recent review of contemporary pedological information related to the identification of the raw material sources for pottery recovered around archaeological soil areas (Nicosia and Stoops, 2017) has significantly enhanced confidence in this aspect. Moreover, the reworked/broken clay coatings of Chromic Luvisols, utilised for the production of the Maya region ceramics of Mexico (Cabadas-Baez et al., 2018), also demonstrate the significance of micromorphology in determining raw material provenance.

Furthermore, detailed knowledge of ceramic mineralogy sheds light on the origin, clay mineral contents and firing temperatures of the raw materials utilized for production. Thus, pedological or micromorphological aspects developed in the ceramics (FitzPatrick, 1972; FitzPatrick, 1993; Stoops, 2003), were recorded, together with the features ascribed to changes that develop during firing in both the mineralogy (e.g., the crystals of the neoformed high temperature minerals-HTMs- and their orientations in the pores) and in the microstructure, along with those postfiring processes that occur following shallow or deep burial of pottery shards. The value of these micromorphological features can also be highlighted in interpreting fragments of pottery by means of their different components such as micromass, inclusions, voids, and, eventually, their coating. The features of each of these components can supply information on the production technology and/or provenance (Maritan, 2017).

Consequently, the principal aim of this brief review is to establish a reliable mineralogical and micromorphological approach to the description and interpretation of the nature (mineralogical) of a range of ancient ceramics, utilizing samples obtained from some world-famous excavation sites in Anatolia (historical name of Asia Minor-Turkey) and also to describe and account for their microstructural processes, i.e. the major attribute of micromorphology. We consider that this brief review may help to explain many of the distinctive mineralogical and micromorphological properties of ancient pottery, as recently documented (Cereda and Fragnoli, 2021).

Ultimately, the studies summarised here also may help to guide studies of these ancient ceramics that are aimed at simulating their processes of manufacture in laboratory-manufactured ceramic specimens.

The principal investigative methods adopted in the studies reviewed here were aimed at detecting and quantifying the primary and high temperature minerals present in these materials and include polarized-light microscopy (PLM), scanning electron microscopy (SEM-BSE EDX), and X-ray powder diffraction of the ground ceramic specimens. Additionally,  $Mg^{++}$  and  $K^+$  saturated and glycolated clay and fine silt-sized particles and/or clay size aggregates randomly oriented on glass slides were X-rayed at room temperature and after heating to 550 °C—the widely adopted procedure for determining the partly or totally uncollapsed clay-size minerals in soils and sediments, i.e. of the raw material sources (Jackson, 1979).

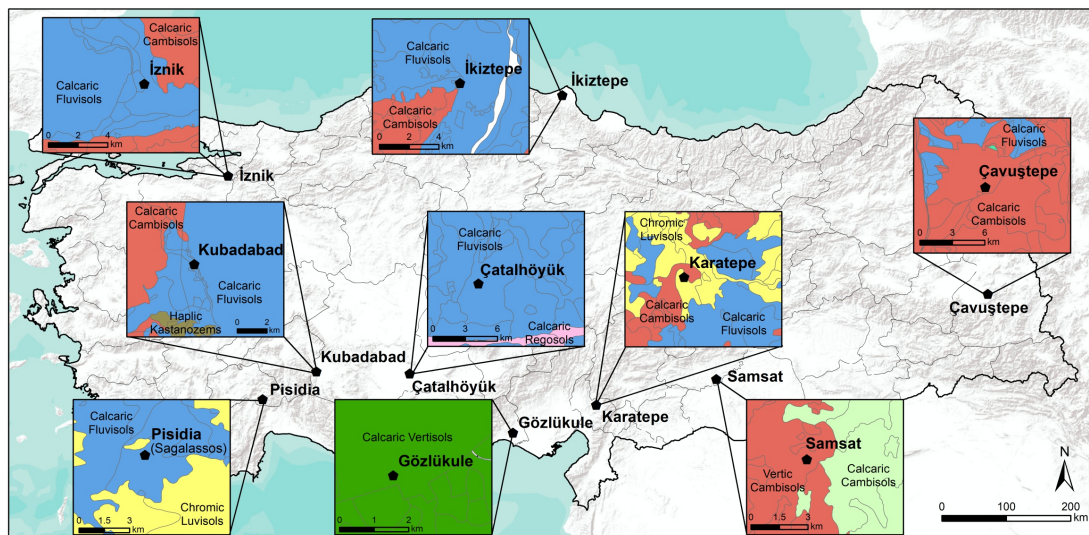
In summary, this review encompasses two major sources that are relevant to the subject matter, namely published national and international papers, together with a summary of hitherto-unpublished results from our four-decades long catalogue of data and relevant theses concerned with research on micromorphology (polarizing microscopy-submicroscopy) conducted in the Soil Mineralogy and Micromorphology laboratories of the Departments of Soil Science and Archaeometry, Çukurova University, Adana, Turkey, the Pedology laboratories of the Plant and Soil Department of the University of Aberdeen, Aberdeen, Scotland, and the Mineralogy & Sedimentology laboratories of the School of Geography, Geology and Environmental Science of the University of Keele, England.

## 2. Ceramic properties related to mineralogy

### 2.1. Provenance of clay-soil/raw material sources and HTMs as indicators of pottery firing temperatures

The durability of the pottery manufactured during the span of time extending from the Neolithic to the mediaeval period appears to be largely dependent on the nature of the raw materials utilised in their manufacture. Thus, determining the provenances of the clays used to make ancient pottery and ceramics has long been a major concern of earth scientists and mineralogists as well as archaeologists. In this context, the examination of petrologic thin sections of pots has yielded valuable visual evidence concerning the overall ceramic composition and characterization of early ceramics (Williams and Jenkins, 1978). Goldberg (1983) had also stated that via the use of ceramic thin sections it would be possible to determine the reuse of broken pottery materials rather than discarded; furthermore, one can usually determine whether the pots themselves were local or imported. Moreover, identification of individual heavy minerals (density  $>2.87\text{g/cm}^3$ ) and distinctive rock fragments within ceramic shards are also valuable tools in tracing the source materials (FitzPatrick, 1993) while information about some coarse components may help to reveal or confirm the origin of the earthenware, i.e. the provenance (Stoops, 2003).

Figure 1 shows the available probable soil-clay sources around the areas where the shards were obtained. Although the excavation site maps cover large areas, they are expected to offer the probable clay/soil sources that might have been utilized by the ancient craftsmen, rather than pinpointing the specific sites of source consumption. This



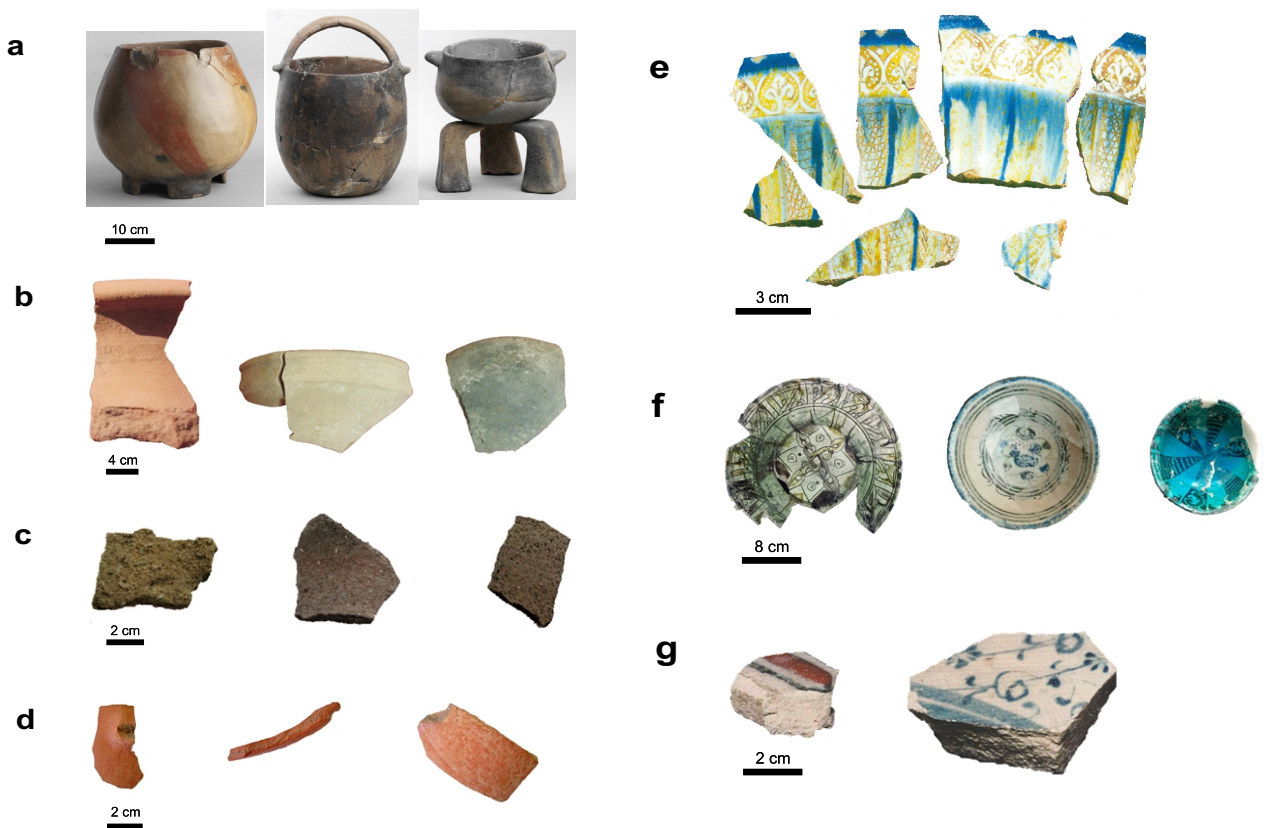
**Figure 1.** The locations of the recovered shards and soils (IUSS Working Group WRB, 2022) of the archeological sites. The site at Pisidia has been included primarily because ‘Sagalassos’ enjoyed great fame in the Classical era and strongly influenced both contemporary and later ideas concerning socioeconomic relations in the ancient Roman sphere of influence (Derygse and Poblome, 2008).

information may be useful in pointing out to the awareness and level of knowledge of the inhabitants concerning the capacity of their soil to cultivate and the appropriateness of the nearby clay/soil sources of pottery. The scale of this map is 1:1,000,000 and it is compiled from local soil maps at 1:25,000 scale (Kapur et al., 2018). In soil maps this enhanced scale might seem to provide a very broad view of the mineralogical characteristics of the excavation sites, but here it is only intended to indicate the large extent of the boundaries of the probable sources since it is difficult to determine the specific point of soil extraction, especially those located within extensive settlement areas.

Fluvisols (also referred to as the Entisols (De Meester, 1970), 7.5 YR 4/3 brown to dark brown in colour) are

represented mainly by the deposits of the Çarşamba River and the ancient wetlands in Çumra (the Konya Basin). These are the probable source materials of the Neolithic (5500 BCE) open-pit fired pottery of Çatalhöyük (Konya, central Turkey) in Anatolia. These soils are dominated by illite and kaolinite type clay minerals with minor smectite and yield a suite of heavy minerals derived from volcanic rocks in the Basin catchment area (De Meester, 1970) (Figures 1 and 2; Table 1).

The red (2.5YR 4/6) to reddish brown colour (2.5YR 3/4) of the more advanced firing products of *terra sigillata* that form the Roman pottery bodies and slips, together with the similar Urartu vessels (5500 BCE, from Van, eastern Turkey) exhibit continuous to extensively vitrified



**Figure 2.** Selected images of ancient Anatolian ceramics:

- (a) Çatalhöyük pottery, non-slipped porous ware;
- (b) Gözlükule ceramic shards, slipped, red ware;
- (c) Hittite ceramic shards, painted, slipped ware and nonslipped, porous ware;
- (d) Urartu ceramic shards, slipped red-brown ware;
- (e) Kubadabad Emirate ceramic shards, scraffito technique: -transparent underglaze decoration;
- (f) Samsat pottery, scraffito, transparent underglaze decoration;
- (g) Ottoman İznik ceramic shards, lead oxide underglaze decoration, iron oxide underglaze decoration. Images of pottery shards from the İkiztepe site were not included due to their amorphous nature. However, despite the similarly amorphous nature of the prehistoric pottery from Çatalhöyük, the images of the restored vessels from this site are provided here for the first time, to show the evolution in shape and form from Neolithic times (Courtesy of the Anatolian Civilizations Museum, Ankara, Turkey and Asst. Prof. Fatih Gülşen, Department of Archaeology, Çukurova University, Adana, Turkey). The images of the pottery shards of Sagalassos were also not included as they were not studied in this paper, but incorporated in Table 1 due to the significance of this site in Anatolian archaeology.

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**Table 1.** The location, geology (MTA, 2020 – <http://yerbilimleri.mta.gov.tr/anasayfa.aspx>), and characteristic soils (IUSS Working Group WRB, 2022) of the sites where the ancient ceramics considered in this analysis were collected, together with brief descriptions of the age and nature of the pottery shards described herein.

Periods of pottery shards	Description	Location	Geology	Soils
Neolithic (ca. 5500 BCE)	Nonslipped, porous ware	37°39'57.59"N-32°49'37.91"E, Çatalhöyük, Konya	Quaternary, alluvial	Calcaric Fluvisols
Bronze Age (ca. 2000 BCE)	Slipped, red ware	36°54'44.52"N-34°53'48.74"E, Gözlü Kule, Tarsus, Mersin	Quaternary, alluvials	Calcaric Vertisols, Calcaric Fluvisols
Early Hittite (1450–1300 BCE)	Painted, slipped ware	41°36'51.08"N-35°52'16.01"E, İkiztepe, Samsun	Quaternary, alluvials	Calcaric Fluvisols, Calcaric Cambisols
Late Hittite (1100–700 BCE)	Nonslipped, porous ware	37°17'41.13"N-36°15'12.26"E, Karatepe, Osmaniye	Quaternary, basalts	Chromic Luvisols
Urartu (900–600 BCE)	Slipped red-brown ware,	38°21'11.20"N-43°27'41.74"E, Çavuştepe, Van	Quaternary, alluvials	Calcaric Cambisol
Classical Hellenistic (500–200 BCE)	Brown-red slipped ware	37°40'10.82"N-30°30'44.16"E, Pisidia, Burdur	Miocene limestones	Chromic Luvisols
Seljuk Kubadabad Emirates (1100–1200 CE)	Scrafitto, transparent underglaze decoration	37°44'37.41"N-31°26'22.12"E, Kubadabad, Konya	Quaternary, alluvials	Calcaric Fluvisols
Seljuk Samsat (1200 CE)	Scrafitto, transparent underglaze decoration	37°30'59.49"N-31°31'52.43"E, Samsat, Adıyaman	Quaternary, alluvials	Vertic Cambisol, Calcaric Cambisol
Ottoman İznik Golden Horn (1500–1600 CE)	Lead oxide underglaze decoration	40°25'46.72"N-29°43'37.93"E, İznik Museum, Bursa	Quaternary, alluvials	Calcaric Fluvisols
Ottoman İznik Coral Red (1560–1610 CE)	Iron oxide underglaze decoration	40°25'46.72"N-29°43'37.93"E, İznik Museum, Bursa	Quaternary, alluvials	Calcaric Fluvisols

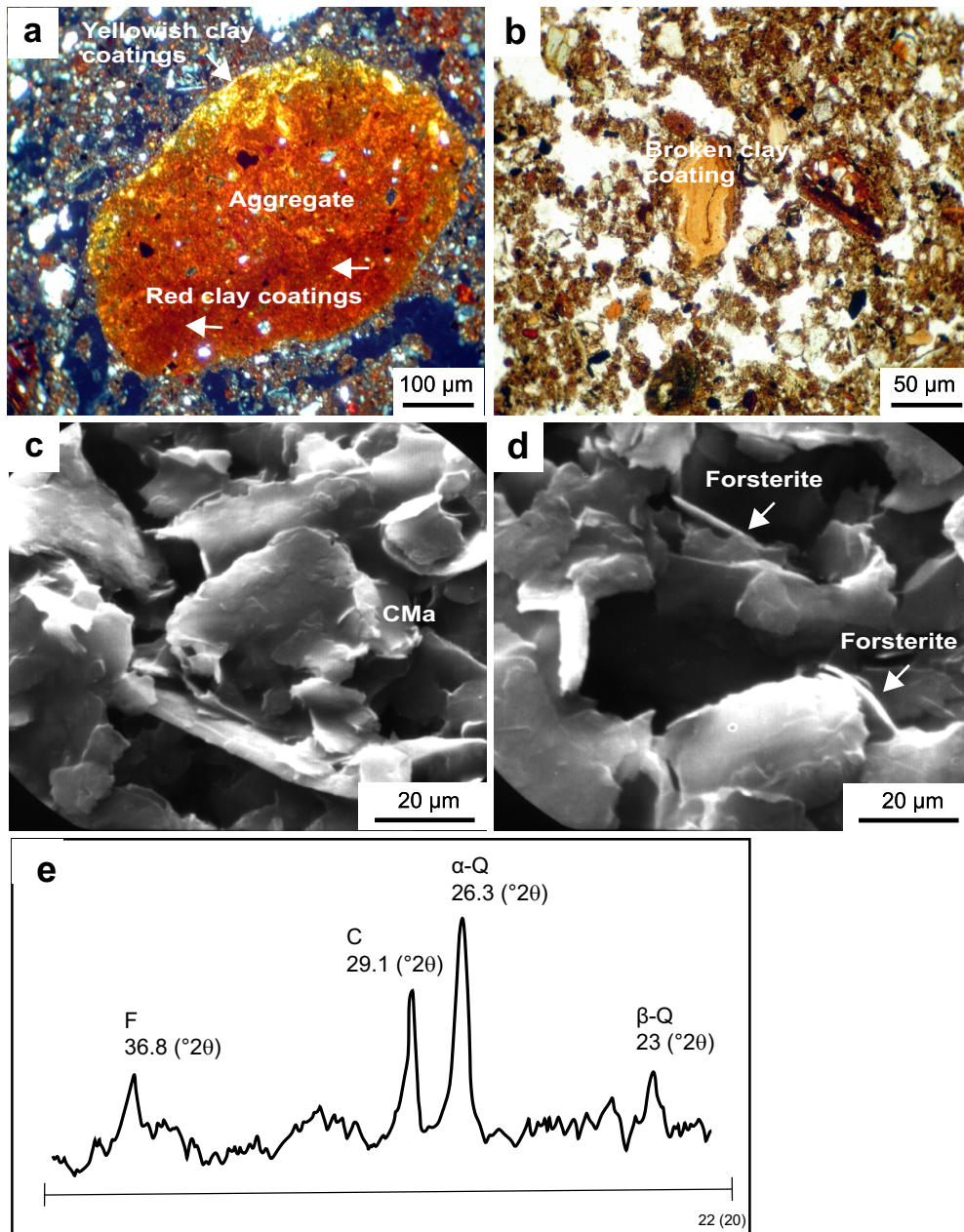
low porosity surfaces (Akça et al., 2010). The probable clay sources of these ceramics were rich in smectite and closely associated with illitic clays, both of which formed from the weathering of mafic volcanic rocks in this area (Kılıç and Çalışkan, 2005). The clay sources utilized for the reddish brown to brown (5YR 4/4 to 7.5YR 5/6) Chalcolithic and Early Bronze age Hittite (Early Hittite) ceramics from İkiztepe (Samsun, northern Turkey) were most likely manufactured from the local Fluvisols and the Cambisols (Chestnut soils) (Kapur et al., 2018) containing kaolinite, chlorite, and illite (Kapur et al., 1992) in the İkiztepe site. The presence of significant quantities of kaolinite in these İkiztepe pottery shards attest to furnace temperatures of less than 550 °C (Figures 1 and 2; Table 1).

On the other hand, the olive brown to brown (2.5Y 3/3–7.5YR 5/6) Late Hittite (1100–700 BCE) ceramics from Karatepe (Kadirli, Turkey) dated by thermoluminescence and optical stimulated luminescence (Atay, 2000), contain locally derived ground basalt rock materials, rich in neoformed kaolinite (Kelling et al., 2020) and mixed with kaolinite-rich basaltic soil with minor illite and smectite, i.e. the ceramic raw materials (750–800 BCE- C-dated) identified in ceramics excavated from the Domuztepe settlement site adjacent to Karatepe (Mermut et al., 2004). These are the Cambisols and/or Fluvisols developed from transported basaltic soil materials (Figure 1) with occasional neoformed smectite (inherited smectitic nodules) developed in basaltic vesicles and incorporated

into the soil after shrink-swell processes that have enhanced the effect of the smectite content in the form of aggregates surrounded by yellowish clay coatings and red pore infills and/or coatings (Kapur et al., 1991) (Figure 3). These broken/reworked smectitic nodules were also occasionally to rarely identified in the basaltic illite-kaolinite rich raw material mixture (Figure 3; Mermut et al., 2004; Akça et al., 2009b) used to make Late Hittite pottery in the Domuztepe site located within the Karatepe excavation area.

The abundant curved clay mineral flakes (clay mineral aggregates-domains of clay mineral crystals) characteristic of these Late Hittite ceramics are most probably responsible for the partially vitrified nature of the ceramic body and the formation of rare forsterite crystals (Akça et al., 2009b; Figure 3). The unusual presence of low amounts of smectite and forsterite in these ceramic shards was determined by X-ray powder diffraction analysis (Akça et al., 2009b), suggesting lower firing temperatures than the 900 °C and shorter periods of firing routinely attained by the Late Hittite furnaces. This also indicates the routine use of Mg-rich basalts or basaltic materials in their production. Our experimental studies conducted on these ceramics revealed a melting point of about 900 °C for basaltic materials (Mermut et al., 2004; Kelling et al., 2020) which also indicates lower firing temperatures than 900 °C (Figure 3).

Moreover, X-ray diffraction analysis has been utilised to determine the fired temperatures by monitoring structural



**Figure 3.** (a) Soil aggregate displaying partial yellowish clay coatings; note also red coatings in pores of the fine-grained matrix of aggregate from Domuztepe ceramic workshop at Late Hittite site, Karatepe; (b) Reworked and/or broken clay coating in raw material mixture, from Domuztepe ceramic workshop at Late Hittite site, Karatepe (Mermut et al., 2004); (c) SEM images of clay mineral aggregates (CMA)- clay domains of curved clay mineral crystals in matrix of Late Hittite ceramics (1100-700 BCE, Karatepe, near Kadirli, Anatolia); (d) SEM image of micropores and rare tabular forsterite in body of Late Hittite ceramic (Akça et al., 2009b); (e) Partial XRD spectrum of this Late Hittite ceramic shard, F: forsterite, C: calcite, Q: quartz, β-Q: β-quartz.

and mineralogical changes as a function of temperature in pottery shards produced by a range of prehistoric and historic societies (Kapur and Bayır, 1981; Kapur, et al., 1998; Rasmussen et al., 2012). The high quality of the extensively vitrified blue and white decorated Damascus

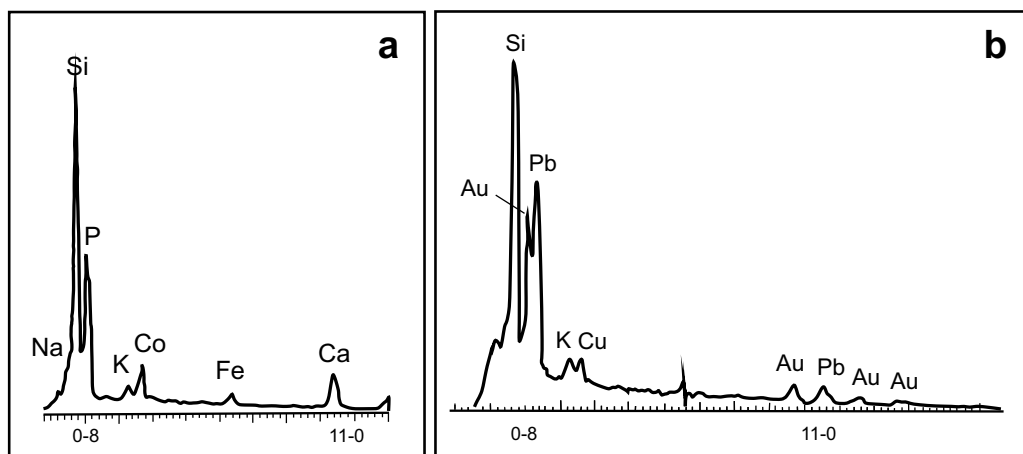
ware ceramics (Ottoman-İznik period, 1500-1600 CE) of western Turkey was most probably achieved by utilization of the kaolinite-rich and iron-free clays from the ancient lacustrine terraces of Lake İznik (Sakarya, 1999). These clays were mixed with ground flint as the main constituent

of the İznik ceramics in the proportion of 10:1:1 for flint (quartz), glass fragments, and clay (Atasoy and Raby, 1989).

Globular hematite is partly responsible for the red colour of the body of the Coral Red İznik ceramics from the same (Ottoman) period. In this context, after firing, hematite has developed in the red clayey body matrix of this shard, under the transparent glaze (Kapur et al., 1998). The uniformly distributed “Coral Red” colour in these unique ceramics and the formation of hematite is probably due to the use of an under-glaze colorant containing ground glass frit mixed with a special red soil material (Aşı Toprağı in Turkish) (Atasoy and Raby, 1989), containing iron oxide (Figure 4a; Kapur et al., 1998). In contrast to the red soil material used in the “Coral Red” ceramic and/or the under-glaze colourant ( $\text{Fe}_2\text{O}_3$ ), the blue colour of the “Golden Horn” vessels is most probably due to the use of lead oxide (PbO) as the under-glaze colourant (Figure 4b). Similarly, Sakarya (1999) determined 6.2 wt.%  $\text{Fe}_2\text{O}_3$  and 12.4 wt.% PbO in the under-glaze colourants of the “Coral Red” and “Golden Horn” specimens, respectively. Our analyses have revealed that the Urartu pottery with characteristic red slip and body colours and vitrified textures was manufactured at low temperatures, compared to the more advanced İznik vessels (Atasoy and Raby, 1989). This was most probably due to the use of Ca-free and Fe-rich amorphous (poorly crystalline) raw material sources in the Urartu pottery rich in illitic and kaolinitic clays with minor smectite (Akça et al., 2010). The Rb contents of the source were likely enhanced by the use of clays rich in lepidolite and spodumene (Akça et al., 2010). Hematite in this mixture may have originated from the alteration of Fe-bearing smectites (Torrent and Cabedo, 1986), like the hematite responsible for the creamy to red

colour changes in the calcareous wall-tiles from Hispano-Moorish workshops (13th to 15th Century AD) in Paterna, Spain (Molera et al., 1998) and also the experimental ceramic replicas from the Bay of Naples (De Bonis et al., 2014). In contrast, hematite formation in Ca-rich ceramics is sometimes hindered by entrapment of Fe in the structure of neoformed high temperature silicates such as gehlenite, thus inhibiting the development of red colour in these ceramics (Kreimeyer, 1987). The red and/or black cores of Early Hittite pottery from İkiztepe reflect abrupt changes, i.e. fluctuations in the firing temperatures and oxidation-reduction conditions during firing (Kapur et al., 1992), as oxidation is indicated by the presence of hematite and reduction by the presence of maghemite determined by XRD as also stated by Özçatal et al. (2014). Our results indicate that the firing temperatures of these Chalcolithic to Early Bronze age pottery vessels from the İkiztepe site varied between 550 °C and 1000 °C, respectively (Kapur et al., 1992).

X-ray powder diffraction has also identified the high temperature minerals (HTMs) such as forsterite ( $\text{Mg}_2\text{SiO}_4$ ), wollastonite ( $\text{CaSiO}_3$ ), spinel ( $\text{MgAl}_2\text{O}_4$ ), and trydimite ( $\text{SiO}_2$ ) in pottery (body and slip-glaze) from the İznik Ottoman period (Kapur et al., 1993, 1998) (Tables 2 and 3), and diopside ( $\text{CaMgSi}_2\text{O}_6$ ) and gehlenite ( $\text{Ca}_2\text{Al}_2\text{SiO}_7$ ) in vessels of the Kubadabad period (Topaksu, 2004) (Figure 6). These are further indicators of a more advanced technology. However, the absence of wollastonite in the “Golden Horn” (16th Century Ottoman) body and glaze-slip and “Coral Red” shard (1560–1610 Coral Red Ottoman) bodies (see Tables 2 and 3) is most probably due to the use of raw materials with low contents of calcium (Figure 4a) and noncrystalline forms of the Ca-Si phase. Alternatively, this peak may be masked by the abundant



**Figure 4.** Microchemical (EDAX) spectra of: (a) fibrous minerals (wollastonite) in the glaze and coating of the Coral Red İznik shard (Ottoman period) (1560–1610 CE) (Table 2); (b) the Golden Horn blue glaze İznik shard (Ottoman period) (1500–1600 CE) glaze and coating (Table 1).

**Table 2.** Mineral contents and crystallinity of Emirate and Ottoman İznik shard bodies (Kapur et al., 1998). The shards provided by Dr T. Tuna (Cerapist, İznik) are listed with their originally assigned dates in Tables 2 and 3. For this study, the two well-known Ottoman shards of the Coral Red and Golden Horn style were reinterpreted by means of micromorphology.

Periods of pottery shards	Quartz		Enstatite		Calcite		Forsterite		Spinel		Wollastonite		Corundum		Feldspar	
	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry
1540–1545 Ottoman	++++	XXX	-	-	-	-	+	XX	++	XX	+	X	-	-	-	-
1530–1540 Ottoman	++++	XXX	+	X	+	X	+	X	++	XX	+	X	-	-	-	-
1510–1520 Ottoman	++++	XXX	+	X	++	XX	-	-	++	XX	-	-	-	-	-	-
17 <sup>th</sup> Century Ottoman	++++	XXX	+	X	+	X	+	X	++	X	+	X	-	-	-	-
1520–1540 Ottoman	++++	XXX	++	XX	++	XX	+	XX	++	XX	+	X	-	-	-	-
17 <sup>th</sup> Century Ottoman	++++	XXX	-	-	+	X	+	XX	+	X	-	-	-	-	-	-
1500–1540 Ottoman	++++	XXX	-	-	+	X	++	XX	++	XXX	+	X	+	XXX	-	-
17 <sup>th</sup> Century Ottoman	++++	XXX	+	X	+	X	+	X	++	XX	+	X	-	-	-	-
11 <sup>th</sup> –12 <sup>th</sup> Century Seljuk Emirates	+++	XXX	++	XX	+	XX	+	XX	+	X	+	X	-	-	-	-
11 <sup>th</sup> –12 <sup>th</sup> Century Seljuk Emirates	++++	XXX	++	XX	+	X	+	XX	+	XX	-	-	-	-	-	-
11 <sup>th</sup> –12 <sup>th</sup> Century Seljuk Emirates	++++	XXX	+	X	+	X	+	XX	+	XX	-	-	-	-	-	-
12 <sup>th</sup> Century Seljuk Emirates	++	XX	+	X	-	-	+	XX	-	-	-	-	-	-	-	-
Seljuk Emirates	+++	XXX	++	X	-	-	+	X	+	X	+	X	-	-	-	-
Seljuk Emirates -1	++++	XXX	-	-	-	-	-	-	++	XX	-	-	-	-	-	-
Seljuk Emirates -2	+++	XXX	++	X	-	-	+	X	+	X	-	-	-	-	-	-
Seljuk Emirates -3	+++	XXX	++	X	-	-	+	X	+	X	-	-	-	-	-	-
12 <sup>th</sup> Century Seljuk Emirates	+++	XXX	+	X	-	-	+	X	+	X	-	-	-	-	-	-
9 <sup>th</sup> Century Byzantine	++	XX	++	XX	-	-	-	-	+	X	-	-	-	-	-	-
16 <sup>th</sup> Century Damascus Ottoman	++++	XXX	+	X	++	XX	+	XX	++	XX	+	XXX	-	-	-	-
16 <sup>th</sup> Century Golden Horn Ottoman	++++	XXX	-	-	-	-	+	X	-	-	-	-	-	-	+++	XXX
1560–1610 Coral Red Ottoman	++++	XXX	-	-	+	X	-	-	++	X	-	-	-	-	-	-
16 <sup>th</sup> Century Coral Red Ottoman	++++	XXX	-	-	-	-	+	X	-	-	-	-	-	-	+++	XXX

Semiquantitative contents: +, low; ++, moderate; +++, high. Crystallinity levels: X, poor; XX, moderate; XXX, high crystallinity. Ab, abundance; Cry, crystallinity.

quartz content in the matrix of the ceramic. In contrast, the wollastonite determined in the Coral Red specimen glaze together with phosphorous (P) and calcium (Ca) (Figure 4a, Table 3) indicate the probable use of bone ash (Şimşek, 2010) for increasing strength and fluxing agent (Ca). Sodium (Na<sub>2</sub>O) and potassium (K<sub>2</sub>O) were also used as alkali fluxes (Sakarya, 1999) in the ceramics of this period.

The absence of forsterite and spinel in these two shards may reflect the illitic and nonsmectitic (absence of montmorillonitic/Mg-rich smectite) character of the

clay source used in production (Kapur et al., 1993, 1998). Moreover, it seems that the use of buried kilns in this period enabled temperatures of around 1000 °C to be achieved through uniform and slowly increased-decreased firing and crystallization from the vapour phase. Firing temperatures under or around 1000 °C in the İznik ceramics are also confirmed by the preservation of angular quartz grains as these grains tend to melt and become rounded at higher temperatures (Kapur et al., 1998).



**Table 3.** Mineral contents and crystallinity of Emirate and Ottoman İznik shard glaze and coatings (Kapur et al., 1998). Shards were provided by Dr T. Tuna.

Periods of pottery shards	Quartz		Enstatite		Calcite		Forsterite		Spinel		Wollastonite		Corundum		Trydmitite		Chrystobalite		Feldspar		İllite		Magnesite		Hematite		
	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	Ab.	Cry	
1540-1545 Ottoman	++++	XXX	-	-	-	-	+	X	++	XX	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
1530-1540 Ottoman	+++	XXX	+	X	-	-	+	X	++	XX	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
1510-1520 Ottoman	++++	XXX	-	-	-	-	+	X	++	XX	+	XX	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
17th Century Ottoman	++++	XXX	+	X	+	X	-	-	++	XXX	-	-	-	-	++	XX	-	-	-	-	-	-	-	-	-	-	-
1520-1540 Ottoman	+++	XXX	-	-	-	-	+	X	++	XX	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
17th Century Ottoman	+++	XXX	+	X	-	-	+	XX	+	X	+	XX	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
1500-1540 Ottoman	++++	XXX	-	-	-	-	+	X	++	XX	+	X	-	-	-	-	++	XX	-	-	-	-	-	-	-	-	-
17th Century Ottoman	++++	XXX	+	X	+	X	-	-	++	XX	+	X	+	X	++	XX	-	-	-	-	-	-	-	-	-	-	-
11-12th Century Seljuk Emirates	++++	XXX	++	X	+	XX	+	X	+	X	+	X	-	-	+	X	-	-	-	-	-	-	-	-	-	-	-
11-12th Century Seljuk Emirates	++++	XXX	++	XX	+	X	+	X	+	X	+	X	-	-	+	X	-	-	-	-	-	-	-	-	-	-	-
11-12th Century Seljuk Emirates	++++	XXX	+	X	-	-	+	X	-	-	-	-	-	-	+	X	-	-	-	-	-	-	-	-	-	-	-
12th Century Seljuk Emirates	++	XXX	+	X	-	-	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Seljuk Emirates	+++	XXX	++	XX	+	X	+	X	+	X	-	-	+	XX	-	-	-	-	-	-	-	-	-	-	-	-	-
Seljuk Emirates-1	++++	XXX	++	XX	+	X	-	-	+	X	-	-	+	XX	-	-	-	-	-	-	-	-	-	-	-	-	-
Seljuk Emirates-2	+++	XXX	++	X	-	-	+	X	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
12th Century Seljuk Emirates-3	+++	XXX	+	X	-	-	+	X	-	-	-	-	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-
Seljuk Emirates	+++	XXX	++	XX	-	-	-	-	+	X	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
9th Century Byzantine	++	XX	++	X	-	-	-	-	+	X	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

Table 3. (Continued).

16th Century Damascus Ottoman	++++	XXX	+	X	+	X	-
16th Century Golden Horn Ottoman	++++	XXX	-	-	+	X	X
1560-1610 Coral Red Ottoman	++++	XXX	-	-	+	-	X
16th Century Coral Red Ottoman	++++	XXX	-	-	+	-	X

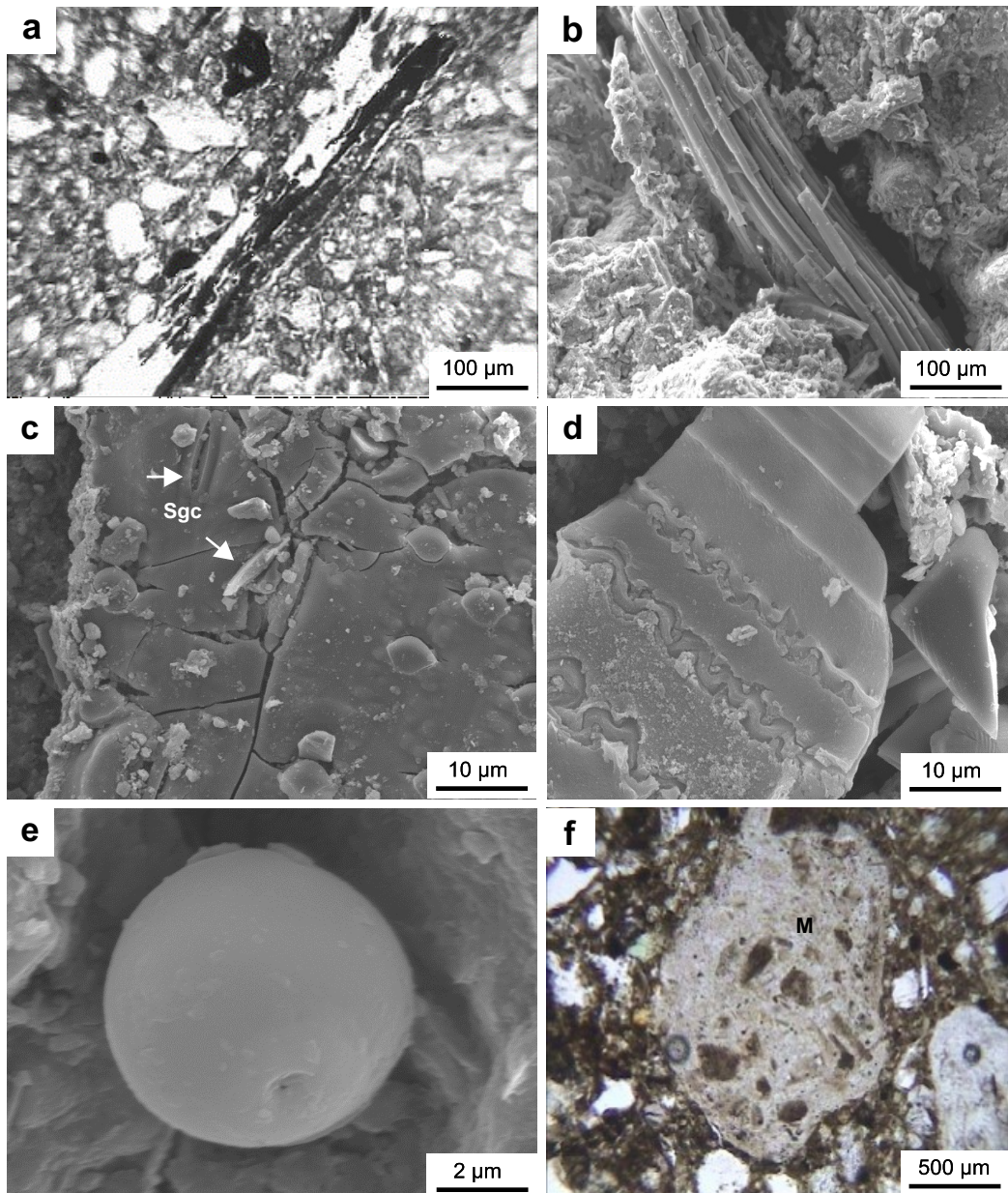
Semiquantitative contents: +, low; ++, moderate; +++, high. Crystallinity levels: X, poor; XX, moderate; XXX, high crystallinity. Ab, abundance; Cry, crystallinity.

## 2.2. Temper

Adding temper to clay in the production of pottery vessels reduces rapid shrinkage and/or expansion during the firing process and promotes the distribution of heat energy through the ceramic paste during firing and/or use of the vessel, i.e. it is the moderating element added to increase strength (Atasoy and Raby, 1989). A wide range of materials has been used as ceramic temper at different times and locations of our study. The nature and proportions of these materials has varied according to the geographic region where the ceramics were being produced and they range from chaff, crushed shell, chamotte, mortar, and stone fragments along with sand-size particles (Figure 5).

Chaff (organic fibres) was also used, in the form of reed fragments (*Phragmites* spp.) seen in the Neolithic Çatalhöyük pottery (5500 BCE) of Konya, central Turkey (Akça et al., 2009a) with linear or undulating orientations (<https://www.cabi.org/isc/datasheet/40514#todistribution>, accessed September 27, 2020) (Figure 5). The prehistoric and primitive open-pit method of firing pottery involved filling pits with clay vessels that were then fired by highly inflammable but low-energy, short-lasting fuels (straw) that reach much lower temperatures (below 550 °C) than the conventional firing methods of later epochs and are thus unable to fully burn organic residues. Ceramics produced at the Çatalhöyük site, alongside partly fresh to burned chaff, contain very fine to coarse quartz, volcanic minerals, mica, feldspar, and calcite (Akça et al., 2009a).

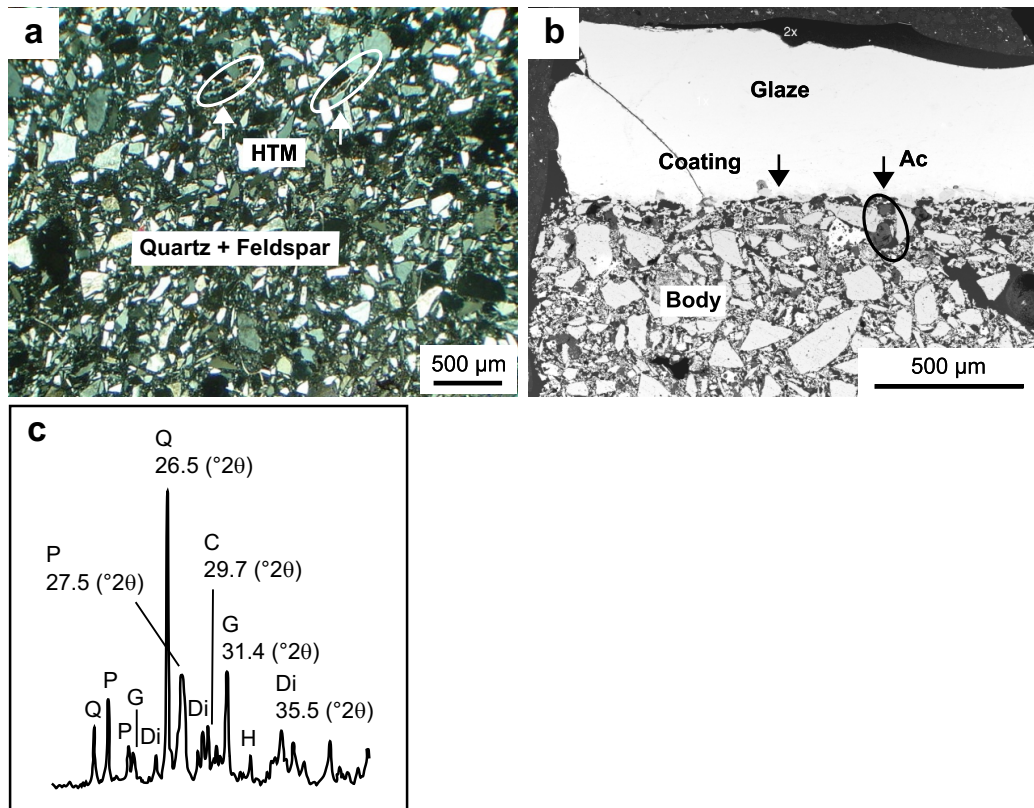
The red to reddish brown weathered ophiolitic raw material sources (Chromic Luvisols) utilised for pottery found in the Roman site of Sagalassos in Pisidia contains smectite together with chlorite and illite and temper elements comprising coarser mineral grains, such as feldspars and high amounts of amphiboles (Table 1) (Dergyse and Poblome, 2008). High contents of quartz were also utilized as temper by Seljuk, Ottoman potters, alongside crushed ceramic (frit) and bone fragments (app. 150 µm size) (Özcan, 1992). Analyses suggest that the Urartu potters (900-600 BCE) of Van, eastern Turkey recognized the value of flux materials low in Ca but containing Li-, Rb-, and Fe-rich mixtures in producing their red slip and body colours with vitrified textures at relatively low temperatures (Akça et al., 2010). Potters producing the Seljuk Samsat (1200 CE) (Dergyse and Poblome, 2008), Kubadabad Emirates (1200 CE) (Sakarya et al., 1990; Kapur et al., 1993) (Figure 6), and Ottoman İznik (1500-1600 CE) (Kapur et al., 1998) ceramics evidently recognised that the addition of angular quartz (broken during the materials mixing process) as temper and the formation of high temperature minerals in their products significantly improved the durability of their ceramics (Kapur et al., 1998). The effect of shape and size of the temper in increasing and/or decreasing basaltic



**Figure 5.** Types of chaff used in Neolithic pottery shards from the Çatalhöyük site (5500 BCE; Konya, central Anatolia) observed under PLM (a, f) and under SEM (b–e): (a) Cellular structure of partly oxidized reed stem-*Phragmites* spp. (plane polarized light—PPL image); (b) Epidermal tissue of *Phragmites* stem; (c) Stomatal guard cells (Sgc) in *Phragmites* stem; (d) Epidermal plant tissue; (e) pollen grain of *Phragmites* spp. in pore; (f) fragment of mortar (M) (PPL image) (modified from (Akça et al., 2009a).

ceramic resistance was also emphasized by Kapur et al. (1995) and Kelling et al. (2000) versus microstructure and high temperature minerals in simulated Late Hittite (1100–700 BCE) vessels. Similarly, the influence of temper on ceramic strength via matrix microstructure was also assessed by Müller et al. (2009) emphasizing the impact of temper shape (platy vs. angular) when replacing phyllite by angular granitic temper. Tite (1989) stated that the bodies

of the İznik ceramics were made from ground quartz with small amounts of white clay and soda-lead frit. In this context, the clay gave the body the plasticity and green-strength necessary for forming the pottery and the frit and contributed to the formation of the interstitial glass which bonds the quartz particles together and gives strength to the fired body. In this respect, the type of clay and frit used are important in the product resistance after firing.



**Figure 6.** (a) PLM image (cross polarized light—XPL) from body of a shard of Kubadabad Emirates pottery (1200 CE, from Konya, central Anatolia), showing abundant fine-crushed, angular quartz and minor feldspar laths (10–50  $\mu\text{m}$  size) together with fibrous HTM phases, all forming the temper in this specimen. The two ellipsoidal outline areas (arrowed) show neoformed needle-like HTMs; (b) BSE image of a Kubadabad period shard with vitreous glaze layer and thin intermediate coating, mixed with fine aggregated clay (Ac) (Topaksu, 2004). (c) X-ray diffractogram of this Kubadabad shard (body and slip-glaze). Q: quartz, P: plagioclase, C: calcite, Di: diopside, H: hematite, G: gehlenite.

### 3. Ceramic properties related to micromorphology-pottery microstructure and pedology

#### 3.1. Evolution of the microstructures of ceramics during firing

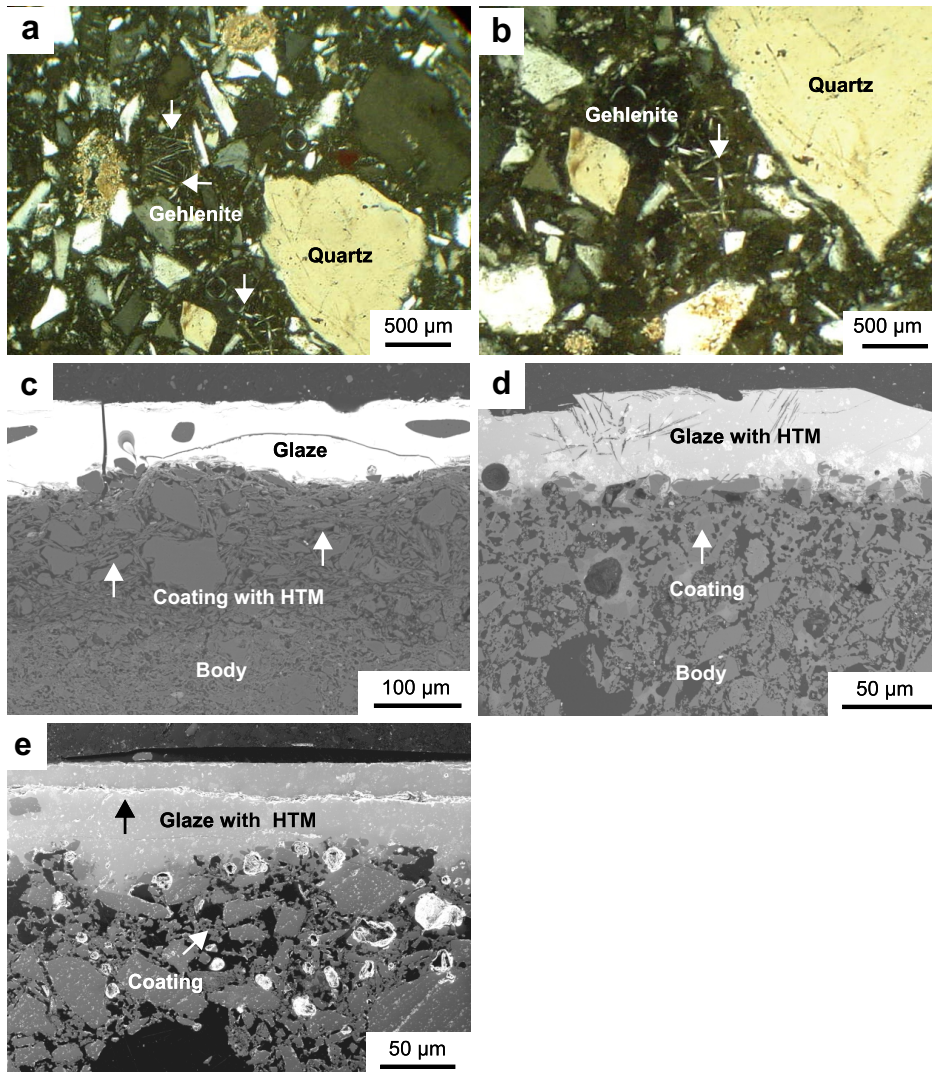
##### 3.1.1. Microstructure: HTMs and their orientation in the body matrix

Gehlenite, initially identified by X-ray powder diffraction (Topaksu, 2004) was also observed under the polarising microscope, where it occurs within strongly altered clinopyroxene grains and displays fibrous morphology and criss-cross orientation (Figure 7). SEM images also revealed the formation of randomly and parallel-oriented fibrous gehlenite in the glaze and coating (Figure 7) of the Kubadabad Seljuk Emirates shards (1200 CE, Konya, central Turkey) (Topaksu, 2004). Wollastonite needles, and radiating clusters in the glaze and body matrix as well as protruding forsterite needles were determined by X-ray powder diffraction and SEM (Atasoy and Raby, 1989; Topaksu, 2004) (Figure 8). These features are most likely the products of an

advanced buried furnace technology involving slow and uniformly increasing-decreasing furnace-firing program and uniform distribution of added minerals, such as quartz and vitreous frit fragments. The protrusion of forsterite needles into the glaze, the pore-filling radial wollastonite clusters and the fibrous gehlenite observed in the body of these Ottoman İznik and Kubadabad Emirates, Seljuk shards are most likely responsible for the increased resistance and long survival of these products in the soil. The blue glaze of the Golden Horn shard was determined by EDAX analysis to contain Pb, Cu, and abundant Si, attesting to the endurance of the glaze decorations against environmental damage.

##### 3.1.2. Microstructure: stress features and porosity

The Chalcolithic and Early Bronze age ceramics from İkoztepe exhibit matrix patterns created by reoriented clay minerals as stress coatings that mimic the porostriated b-fabrics (Stoops, 2003) and the parallel oriented basal planes of the clay minerals (FitzPatrick, 1993) encountered in some soils. Some of these stress coatings [stress cutans

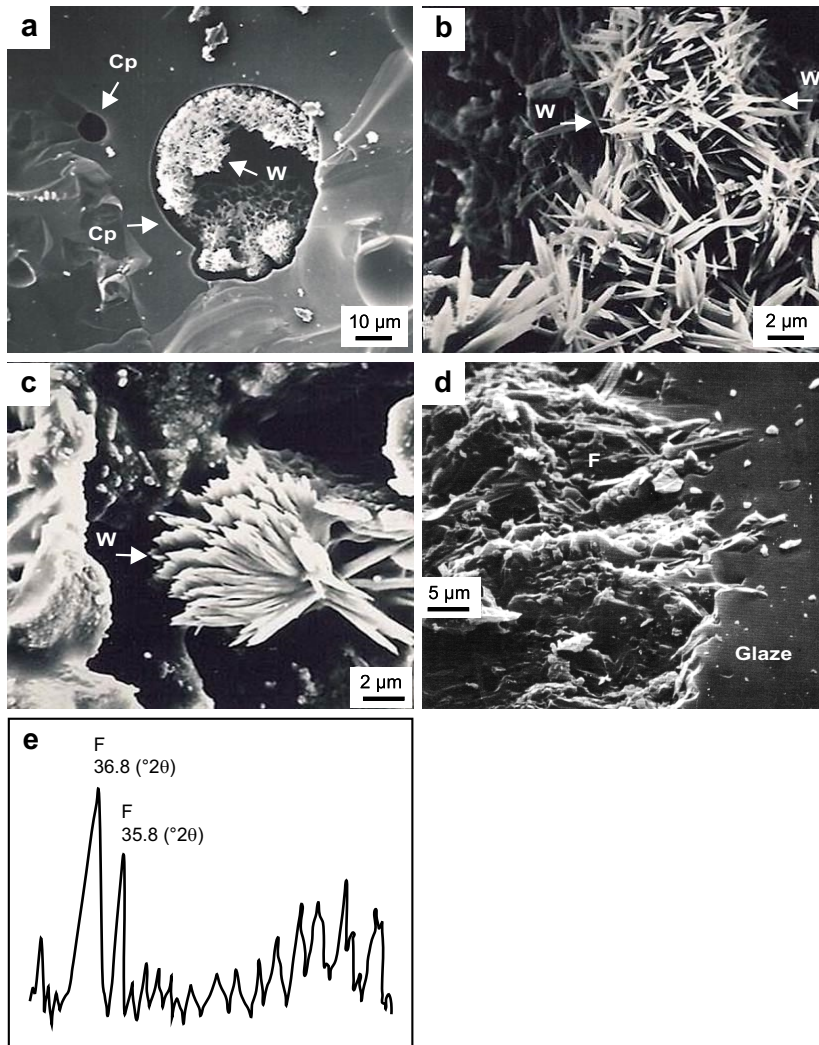


**Figure 7.** XPL (a, b) and BSE (c, d, e) images of thin sections of Kubadabad Emirate Seljuk-ware shards (1200 CE, Konya, central Anatolia). (a, b) strongly altered clinopyroxene grains and gehlenite associated with angular quartz grains; (c) random and parallel oriented fibrous minerals (gehlenite) in coating; (d) occurrence of randomly and parallel-oriented fibrous minerals in the glaze and coating; (e) glaze with layer of parallel oriented HTMs (Topaksu, 2004).

(Kapur et al., 1992)] were developed parallel to the continuous, undulating cracks (i.e. elongated pores, in pedological parlance) within interlayers in the bodies of ceramics, and arguably help to enhance their durability and strength. These fractures reveal a probable rapid rise in prevailing furnace temperatures.

The effects of stresses imposed on ancient ceramics in the course of their manufacture and especially during firing are detectable through a range of measurable properties. Within the shard body, these include the number and size of fissures and fractures and the volume, orientation, and shape of pores. Evidence for shrink-swell phenomena and

imposed stresses affecting body and slip layers are also provided by secondary porosity and cellular structures (Freestone et al., 2009). Thus, the Neolithic shards from Çatalhöyük (Konya, Turkey, 5500 BCE), products of open-pit firing, display occasional fractures (elongated pores) with strong preferred orientation (Figure 9) alongside repeated fine stress coatings (Figure 10). These features are probably attributable to the smectite content of the clay used to make this pottery and probably also result from the hand-shaping process employed, together with the rapid and abrupt increase-decrease patterns of temperatures, i.e. the differential expansion or contraction of the different mineral



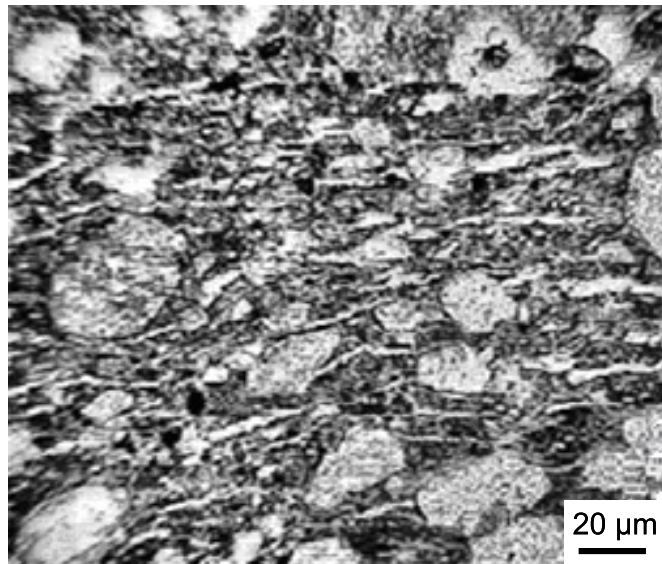
**Figure 8.** SEM images of Ottoman Coral Red İznik ceramics (1560-1610 CE): (a) Occurrence of wollastonite clusters (W) within cellular and/or bloating pores of glaze (Cp) (Sakarya, 1999); (b) and (c) wollastonite (W) forming radial protruding clusters within rounded pore spaces in the glaze; (d) forsterite (F) protruding from body-slip into glaze (modified from Kapur et al., 1998); (e) X-ray powder diffractogram of forsterite crystals observed protruding from body-slip into glaze (d).

phases present in these early open-pit fired pottery vessels (Akça et al., 2009a). Many of the Early Hittite (Late Bronze age) İkištepe ceramics are also characterized by elongated (with preferred orientation) and irregular pores (10–100  $\mu\text{m}$  diameter), together with rare to occasional stress coatings (Kapur et al., 1992). These features emanate from various inclusions and document a manufacturing process that involved moderate quality shaping and open-pit firing with fluctuating temperature rise, as also determined in the Çatalhöyük pottery (Akça et al., 2009a).

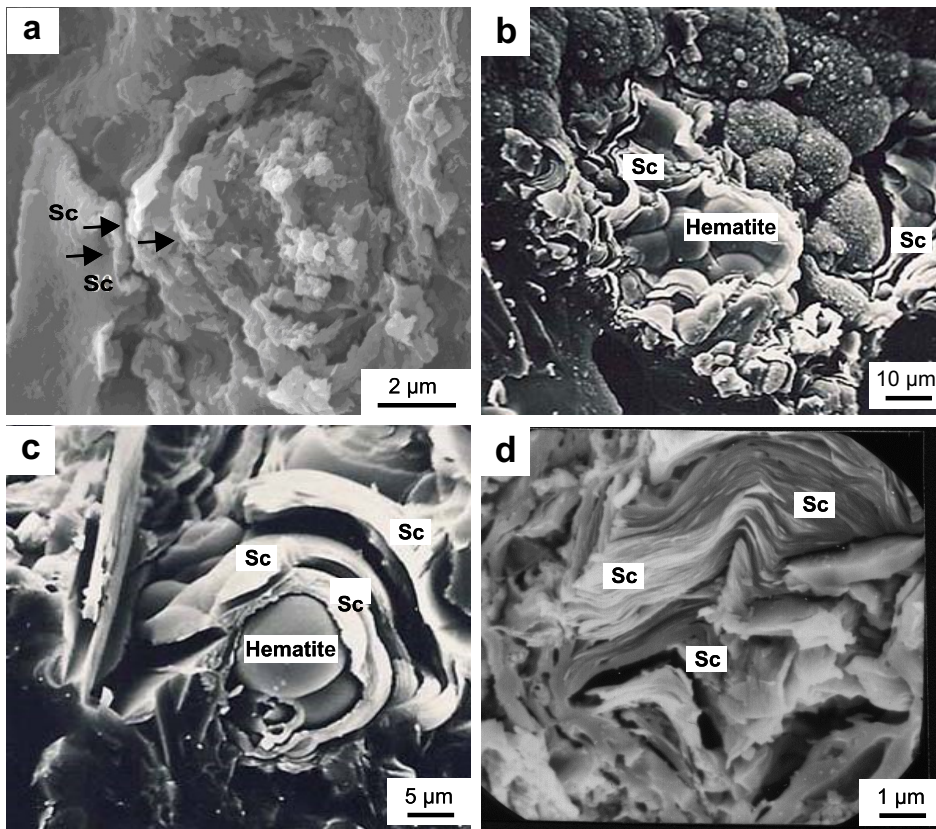
Prominent and well-developed stress coatings were also formed during the firing process around secondary

hematite globules (Figure 10) in the body of the Ottoman İznik ceramics (1500–1600 CE) (Kapur et al., 1998). Much earlier some Late Hittite ceramic shards, produced from the smectitic basaltic soils (Vertisols) of Kadirli (Turkey), reveal the development of multiple V-shaped stress coatings that greatly modify the microstructure despite their illite and kaolinite contents (Figure 10; Akça et al., 2009b).

Pottery from the highly advanced Urartu kilns from Çavuştepe Van, eastern Turkey records probable temperatures of around or over 1000  $^{\circ}\text{C}$  and was produced much earlier (900–600 BCE) than the high-quality Roman ceramics, fired in equally advanced kilns. The body of the



**Figure 9.** PLM image displaying preferred orientation of fibrous minerals and parallel- elongated pores in the body of a Neolithic pot from Çatalhöyük (Konya, Turkey) 5500 CE (Akça et al., 2009b).



**Figure 10.** SEM images showing development of repeated fine stress coatings (Sc) in selected ancient ceramic bodies: (a) Neolithic pottery (5500 BCE) from Çatalhöyük Konya, Turkey) (Akça et al., 2009a); (b, c) Stress coatings around secondary globular hematite (identified by XRD) (Akça et al., 2009a) and developed in the body of Ottoman Coral Red İznik ceramics (1560-1610 CE) from Bursa, western Anatolia (Kapur et al., 1998); (d) Multiple V-shaped stress coatings developed in Late Hittite ceramics from Karatepe (1100–700 BCE, Kadirli, Anatolia) (Akça et al., 2009b).

Urartu ware contains relatively few and fine pores while the slip contains rare rounded to oval pores, attesting to their production by the most advanced contemporary firing systems (Akça et al., 2010). In contrast, the Roman-Byzantine ware contains pores of variable sizes (20–250  $\mu\text{m}$ ) and shapes (oval to rounded) in a matrix displaying randomly oriented fractures, undulating elongated pores, and other fine stress features, all documenting a less sophisticated firing technology (Akça et al., 2010). Similarly, the Ottoman İznik ceramics from Bursa, western Turkey (Figure 11), exhibit well-rounded bloating pores developed by entrapped  $\text{CO}_2$  gas emitted during vitrification of the glaze. Domains of bloating pores in the glaze of İznik ceramics surrounded by fine stress coatings form a “cellular structure” as stated by Tite and Maniatis (1975). This attests to the adoption of rapid cooling but more uniform firing programs and the use of sodium-rich ( $\text{Na}_2\text{O}$ ) transparent glaze materials inducing vitrification. Amounts of  $\text{Na}_2\text{O}$  added as stated by Henderson (1989) and Sakarya (1999) were 13.5 wt.% and 12 wt.%, respectively. The pore sizes and shapes of the Seljuk Emirate and İznik ceramics were also determined, revealing the presence of dominant irregular and rounded pores of 40–60 microns in size. These pores exhibited varying distributions in the matrices of the Emirate shards by 21% and of the İznik shards by 28%–36% (Sakarya 1999). These results indicated consistent and well-controlled furnace temperatures in the use of the İznik kilns.

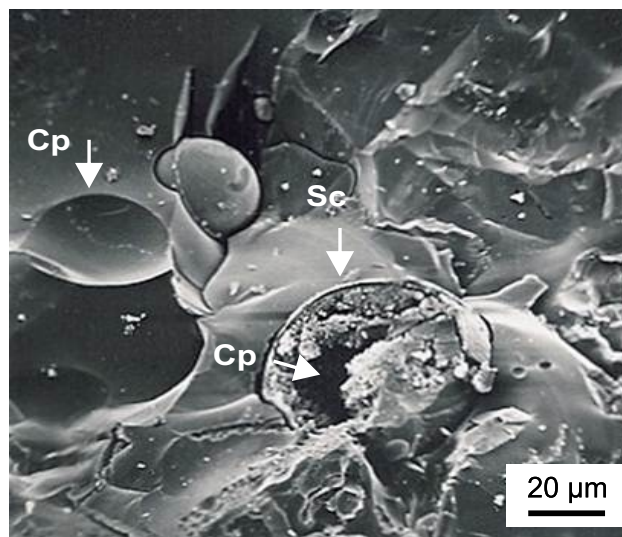
### 3.2. Development of postburial features in ceramics

#### 3.2.1. Clay coatings

Postburial features in ceramics may also provide clues concerning soil formation processes (pedogenesis)

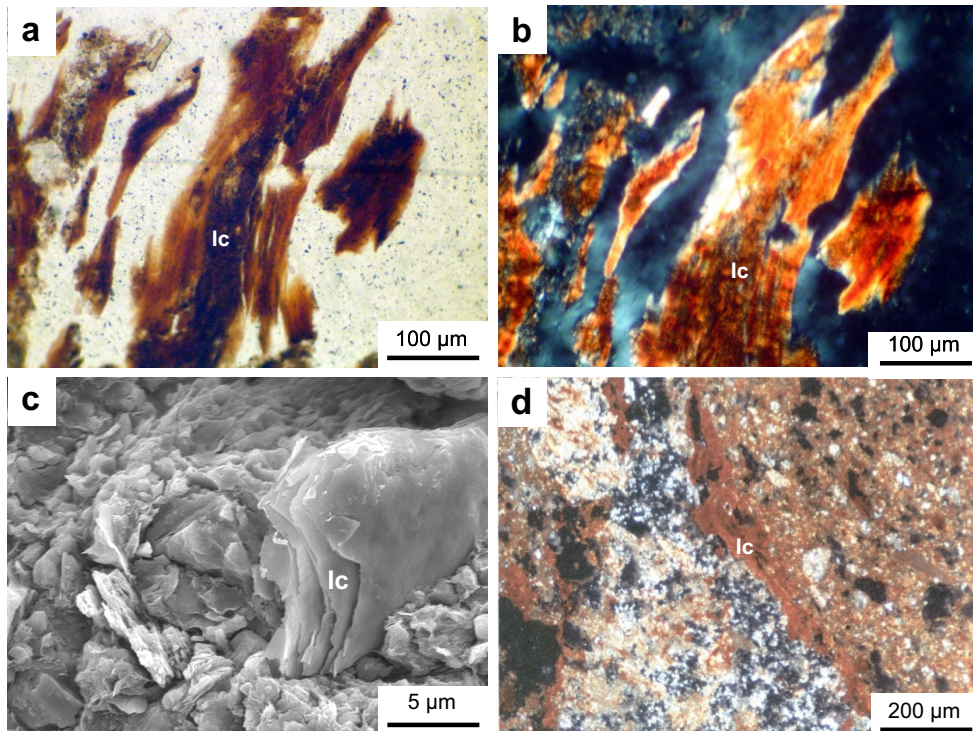
occurring in the vicinity of the buried horizons sheltering the ceramic shards. Clay coatings, formed from illuviated-leached clay size particles suspended in percolating soil solutions and deposited at depth on mineral surfaces or pore infills (FitzPatrick, 1993), most likely developed in ceramic shards immersed in the near surface soil-water suspension for elongated periods and/or located in subsurface soil horizons. These coatings are readily identified by their pleochroism in plane polarized light (Figure 12) and display reddish interference colours in cross-polarized light, together with linear clay coatings-anisotropic clay domains and rounded-banded SEM morphologies. In the present study, these features have been best observed in vessels from the Neolithic Çatalhöyük site, Konya, Turkey (5500 BCE) (Akça et al., 2009a) and also from the Late Bronze Age (Hittite) Gözlükule excavation, Tarsus, Turkey (2000 BCE (Kızılaslanoglu, 2013).

It has been argued previously (Erol, 1979; Mermut and Jongerius, 1980; Kapur et al., 1990; Fedoroff and Courty, 2013) that similar leaching-deposition features of clay particles (clay coatings) occurring in soils may document the climatic fluctuations that occurred during the Late Pleistocene and the early- to mid-Holocene wet-dry periods (Mélanie Roffet-Salque et al., 2018) in Çatalhöyük, central Turkey [8.2 kyr BP/ca. 6200 BCE: dates ranging from before 8325–8205 BP/6325–6205 BCE to 7925–7815 BP/5925–5815 BCE calibrated years (cal)]. Nevertheless, the presence of reed fragments (*Phragmites* spp.) in these shards suggest that the early- to mid-Holocene climatic fluctuations were most likely milder than the glacial-interglacials (pluvial-interpluvials) recorded in Pleistocene deposits across the Mediterranean region.



**Figure 11.** SEM image of glaze displaying extensive vitrification features and/or bloating pores, e.g., rounded pore spaces (Cp) (i.e. “cellular structure”), and stress coatings (Sc) in the Ottoman Coral Red İznik ceramics (1560–1610 CE) from Bursa, western Anatolia (Sakarya, 1999).





**Figure 12.** (a) PPL image of repeated anisotropic linear clay coatings-anisotropic clay domains in body of Neolithic shard from Çatalhöyük (Konya, Turkey; 5500 BCE); (b) XPL image of the same specimen; (c) SEM image of illuviation coating as pore-fill (lc); (d) PPL image of illuviation coating on Bronze Age ceramic (2000 BCE, Gözlükule, Tarsus, Anatolia) (Kızılaslanoğlu, 2013).

### 3.2.2. Faunal activity and calcification

Indications of faunal activity, such as abandoned earthworm channels may reflect on-going bioturbation during the process of soil formation (Figure 13). The rare secondary carbonate (micritic calcite) accumulations (Figure 13) found in the pores of the Kubadabad Emirates ceramic shards from Konya, Turkey (1200 CE) may also indicate earlier calcification-decalcification of accumulated aeolian material of ultimate Saharan origin and recrystallization of the carbonates in the soil hosting the ceramics as well as in the pores of the Kubadabad Emirate pots (Topaksu, 2004). On the other hand, this rare phenomenon probably results from local crystallization within the ceramic matrix of calcium leached from of a widely used type of mortar, employed in the construction of the Kubadabad palace (Tunçoku et al., 2004).

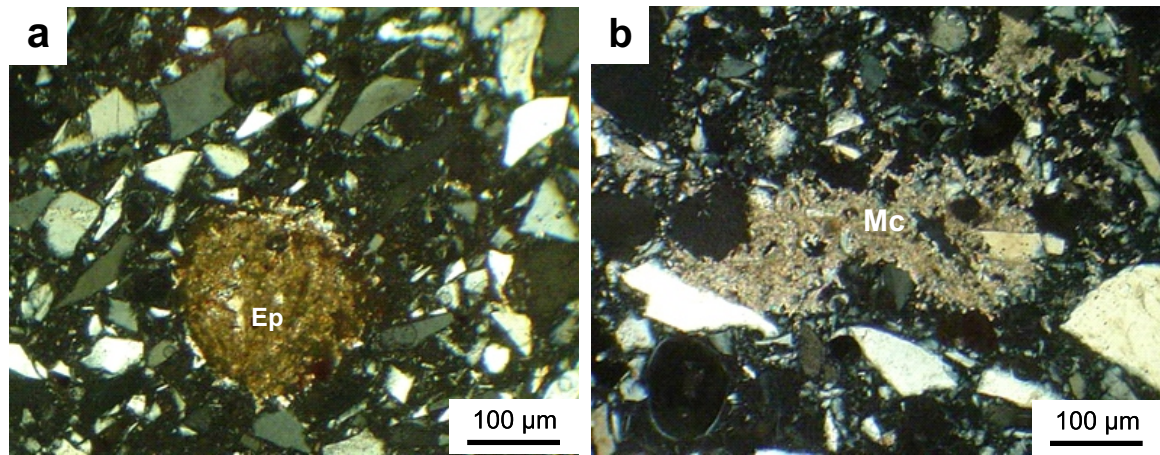
### 4. HTMs and pore microstructure developed in simulated Late Hittite ceramics

As demonstrated earlier, local basalt was used for tempering Early to Late Hittite (1450–1300 BCE to 1100–700 BCE) ceramics, fabricated mainly from local alluvial soil raw materials, at localities ranging from the Black Sea to the Mediterranean coasts of Anatolia, e.g., the İkiztepe (Samsun) and Karatepe (Osmaniye) sites (Kapur et al., 1992; Akça et al., 2009b).

To further test this conclusion, simulation studies have been conducted using mixtures of these probable local source materials, including some basalt clasts (later identified as containing neoformed kaolinite (Çambel et al., 1996; Kelling et al., 2020) and basaltic soils (from the Osmaniye palaeosol, buried soil underlying fluvial materials), all obtained from exposures in close proximity to the Karatepe excavation-open air museum site. Other test-mixtures for these simulations included materials derived from well-known Anatolian clay sources such as the Söğüt İnhisar materials from western Turkey.

Mixtures selected from this array of clay sources were fired in an electric furnace in an oxidising atmosphere at 1050 °C, using basalt tablets, and at 1200 °C using powdered pumice. Subsequently, a suite of HTMs, namely pseudowollastonite, wollastonite and forsterite was identified as present on vitrified surfaces and in the pores of the specimens. These HTM minerals are considered to be responsible for the durability of these simulated pots (Figures 14a–14c). Further tests conducted on the basaltic ceramic specimens of varying mixtures also revealed increased durability with increasing amount of irregular smaller pores compared to the rounded (Kelling et al., 2000).

Neoformed tabular crystals of pseudowollastonite (a high temperature modification of wollastonite forming



**Figure 13.** (a) XPL image of Kubadabad Emirates ceramic from Konya, Turkey (1200 CE), showing well-rounded earthworm pellet (Ep) or pore in-fill with secondary irregular calcite developed round the periphery (Topaksu, 2004); (b) XPL image of secondary carbonate (micritic calcite—Mc) accumulation in Kubadabad Emirates ceramic, Konya (Topaksu, 2004).

at an inversion temperature of 1120 °C) were identified within rounded to oval pores (in a criss-cross pattern-pore microstructure) (Figure 14c). These are attributed to the thermal transformation of pumice, silica-rich smectite, kaolinite, palygorskite, and calcite as well as small amounts of original wollastonite in the utilised raw materials (Tunçoku et al., 2004). Forsterite and wollastonite also formed in the 1200 °C fired ceramic specimen indicating the usual transformation sequence of forsterite to wollastonite and to pseudowollastonite at 1150 °C (Figure 14a; Kapur et al., 1995; Kelling et al., 2000).

## Conclusion

On the basis of the results described in the preceding sections of this review, the following conclusions are offered, concerning the mineralogical nature and micromorphological features determined in these ancient Anatolian ceramics with:

- The ceramic raw materials used in the prehistoric and classical period contained illite, kaolinite, and chlorite, as well as smectite, whereas kaolinite was the dominant mineral used in ceramics from the Seljuk Emirate and Ottoman periods.
- The main sources for raw materials used in the Anatolian ceramics were Fluvisols and Vertisols of alluvial origin, together with Cambisols formed on basaltic and mafic volcanic rocks and limestones. In addition, Luvisols developed on ophiolitic bed-rocks were likely utilized by the Urartu in eastern Anatolia.
- The clay coatings detected on the Çatalhöyük and Gözlükule shards probably reflect early- to mid-Holocene climatic fluctuations and/or local transportation-deposition of clay size particles within the pottery body.
- The organic chaff (*Phragmites* spp.) detected in the shards of the ancient pit-fired pottery from Çatalhöyük (5500 BCE) most likely reveals low temperatures achieved in their

furnaces and also suggests that the contemporary climate was similar to the present-day.

- The Neolithic Çatalhöyük and Early Hittite shards display elongated and irregular pores together with rare to occasional stress coatings. They contrast with the vessels produced by the more advanced societies of the Urartu and Ottoman epochs whose much improved furnaces and firing programs were responsible for the development of the oval to rounded pores in the body and slip.

- The red-black cores and hematite/magnetite crystals in the matrix of the Early Hittite İkiştepe ceramics further reflect the abrupt changes and fluctuations (oxidation-reduction) in their furnace firing temperatures, which varied from 550 °C to 1000 °C, as indicated by the preservation of illite and chlorite crystals in the shards.

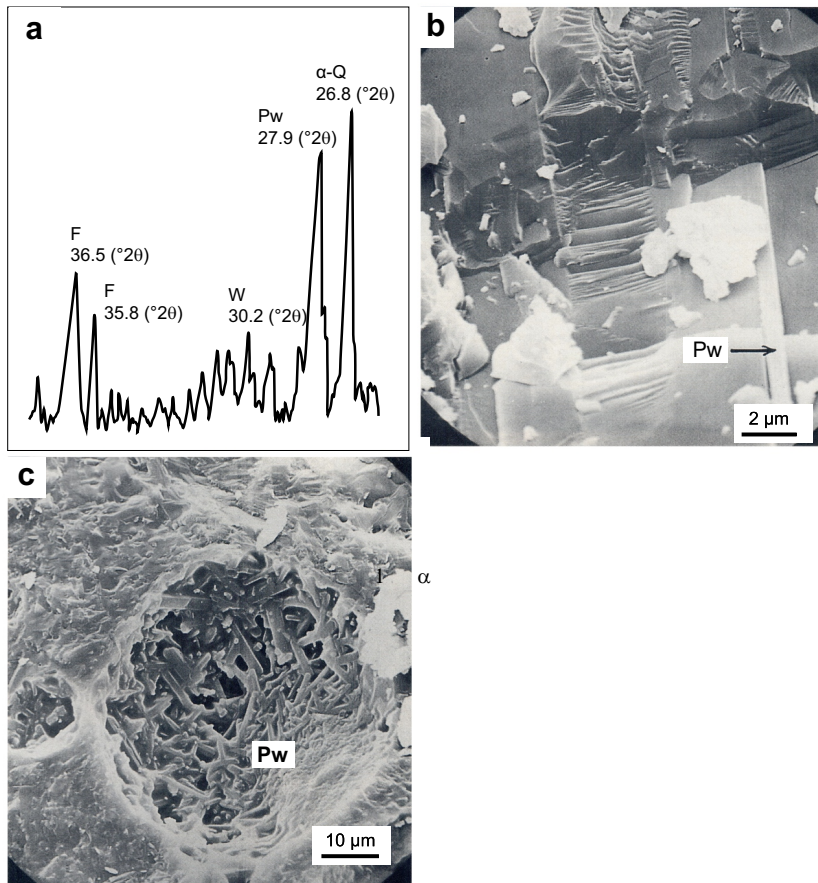
- Firing temperatures for most Late Hittite wares were around 900 °C, as indicated by the presence of smectite (partly collapsed to illite) and forsterite despite the presence of illite and kaolinite, as determined by X-ray powder diffraction.

- The low porosity and the uniform and intense vitrification of the Urartu pottery from eastern Anatolia also attest to the attainment of high and uniformly increasing firing temperatures (in excess of 1000 °C) in their kilns.

- Unusually high amounts of quartz were utilized as temper in the Seljuk Emirate and Ottoman pottery, likely enhancing their durability and resistance to wear.

- The presence of gehlenite and diopside in the matrix of Seljuk Emirate shards and of wollastonite in the Ottoman İznik ceramics attest to their manufacture under uniformly and slowly increasing firing temperatures to around 1000 °C.

- The angularity of the abundant quartz grains used as temper in the İznik ceramics also indicate firing temperatures below 1000 °C as stated by Tite (1989). Firing temperatures over 1000 °C would be indicated by the more rounded angular



**Figure 14.** Images relating to laboratory-manufactured ceramic specimens: (a) X-ray diffraction pattern showing the presence of forsterite, wollastonite, and pseudowollastonite; F: forsterite, W: f, Pw: pseudowollastonite,  $\alpha$ -Q:  $\alpha$ -quartz; (b) SEM images of pseudowollastonite on ladderlike and vitrified fracture surfaces; (c) well-developed tabular pseudowollastonite (Pw) in pore of fired specimen using basaltic pumice + Osmaniye paleosol (basaltic soils or Cambisols and/or Fluvisols) (IUSS Working Group WRB, 2022) mixture.

corners of the quartz crystals due to enhanced vitrification. The abundance of neoformed HTMs in the Seljuk Emirates and Ottoman ceramics are further indicators of the use of advanced furnaces with slow and uniform increasing temperature controls.

- The development of stress coatings (the porostriated b-fabrics) observed in the Ottoman İznik ceramics conferred to enhanced durability and resistance to everyday hazards and provides further evidence of advances in furnace technology that enabled higher, more uniform kiln-temperatures to be routinely achieved.

The ultimate goal of this brief review was to compile the mineralogical and pedological features of the Anatolian ceramics given herein, in order to create an interdisciplinary data repository in the future for use in research of simulated productions on a variety of ceramic specimens from around the world.

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