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TECHNOLOGICAL FEATURES OF THE CHALCOLITHIC POTTERY FROM TÂRPEȘTI (NEAMȚ COUNTY, EASTERN ROMANIA)

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ABSTRACT

The technological parameters of representative pottery samples attributed to Precucuteni (5050-4600 cal BC) and Cucuteni (4600-3500 cal BC) cultures identified at Târpești (Neamț County, Eastern Romania) were determined using a complex archaeometric approach. The site is located in the north-eastern part of the present-day Romania occupying a small plateau situated in a hilly region. In order to evaluate the raw materials and the firing process we have used optical microscopy (OM), X-ray powder diffraction (XRPD) and magnetic measurements. Further on, the XRPD data were statistically treated using hierarchical cluster analysis (HCA) taking into account position and peak intensity, the Euclidian distance as metric and the average linkage method as a linkage basis for gaining a more refined estimation of the mineralogical transformations induced by the firing process and for defining homogenous group of samples. Based on the multivariate statistical analysis we could determine the most representative sample from each group, which was disccussed separately in order to identify the specific mineral phases responsible for the separate grouping. In addition, the mineralogical transformations were investigated by measuring the magnetic properties, especially the major hysteresis loop (MHL) of the pottery samples. All the Precucuteni pottery falls within the same group with some of the Cucuteni pottery samples due to the similar firing temperature range, while the other group containes only Cucuteni A pottery which were exposed to a higher temperature. Besides the similarities in the firing regime, our results revealed afinities in the raw material selection and processing between the Precucuteni pottery and some of the Cucuteni samples.

KEYWORDS: pottery, Cucuteni, Precucuteni, firing technology, optical, XRPD, magnetic properties

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1. INTRODUCTION

The technological choices made by the potters are not only reflecting the various sequences involved in the vessel manufacture, but also helps to further assess the cultural and social milieus. Therefore, the material qualities of potteries like color, texture, clay processing, and the addition of various inclusions or the estimation of firing temperature can be used to infer the technological abilities of the prehistoric artisans.

For determining the technological features like porous, mineral and amorphous phases, texture and the orientation of grains and aggregates various combinations of analytical techniques such as optical microscopy (OM), Scanning Electron Microscopy (SEM), Raman Spectroscopy, X-ray Powder Diffraction (XRPD), thermal analysis (TGA), Fourier transformed infrared sspectroscopy (Ibrahim, Mohamed, 2019; Antonelli et al., 2018; Panagopoulou et al., 2018; Javanshah, 2018; Abdel Rahim, 2016) or the use of magnetic methods (Chudin et al., 2019) are available.

The investigation of the pottery assemblages represents a focal point in the archaeological debate regarding the social, economic and symbolic transformations specific to the Chalcolithic period in South-Eastern Europe. One of the most intriguing subjects when analysing European prehistory is perhaps the Cucuteni-Trypillia civilization (5th – 4th millennia BC) which extended over the present-day eastern part of Romania, Republic of Moldavia, to the south of Ukraine. It displays features common to Chalcolithic period in South-Eastern Europe, such as layered sites, long-term settlements, large dwellings with abundant inventories, as well as richly and specifically decorated pottery.

The present study aims to identify the main technological parameters of the Precucuteni and Cucuteni A pottery identified at Târpești-*Râpa lui Bodai* in order to assess the degree of variability manifested between Precucuteni and Cucuteni ware and among various production sites. We infer the main characteristics of the raw materials used, and show a correlation between pottery composition and the estimated equivalent firing temperature, combining macroscopic observations with mineralogical and petrographic analysis using polarized light microscopy (OM), X-ray powder diffraction (XRPD) and magnetic properties analysis. This research is part of a more systematic archaeometric study focused on identifying the technological features of the Cucuteni pottery (Matau et al., 2013a; 2013b; Salaoru et al., 2013; Matau et al., 2014; Tencariu et al., 2018).

OM is very useful for evaluating the type of raw materials used and for differentiating between various technical recipes followed to make the ceramics (from shaping to firing) (Degryse, Braekmans, 2017). Among the numerous techniques used to determine the firing temperature, X-ray powder diffraction (XRPD) occupies a forefront position due to its ability to identify the newly formed crystalline phases in the ceramics (Heimann, 2017). Although not yet extensively used for evaluating the mineralogical transformations induced by the firing process, the hysteresis measurements are very suitable for inferring the magnetic domain state and related grain size of the magnetic minerals (Jordanova et al., 2019 with references therein).

When trying to reveal the firing process of the prehistoric pottery the most important parameters are the heating rate and the length of exposure time to heat (soaking time). Accordingly, the evaluation of the firing temperature is considered as the *equivalent firing temperature*, which may not correspond exactly with the firing temperature set initially (Gosselain, 1992; Livingstone Smith, 2001).

2. ARCHAEOLOGICAL BACKGROUND

The terminology used to designate this civilization is influenced by different research traditions which tends to follow modern regional traditions and political divisions – *Ariușd* for the Transylvanian variant (in Hungarian, *Erösd*), *Cucuteni* for the present-day eastern part of Romania, *Trypillia* for the sites located in Ukraine (in Russian, *Tripolye*). Russian and Ukrainian archaeologists considers Trypillia as one culture with several phases, while Romanian archaeologists describe Precucuteni, Cucuteni, and Horodiștea-Erbiceni as three different civilizations with various phases: Precucuteni is equivalent to Trypillia A, Cucuteni (A, A-B, and B) to Trypillia BI-CI and Horodiștea-Erbiceni to Trypillia CII (Lazarovici, 2010 with references therein). Radiocarbon dates for settlements attributed to Precucuteni and Cucuteni communities located in the eastern part of present-day Romania covers a large timespan from c. 5050-3500 cal BC (Mantu, 1998; Lazarovici, 2014).

Most of our knowledge concerning the Precucuteni and Cucuteni ware is based on stylistic and typological analysis, while the pottery function (Bodi, Solcan, 2010) was only recently addressed. The technological characteristics of the Cucuteni pottery were evaluated for inferring the firing process and the related mineralogical transformations (Ionescu, Hoeck, 2011; 2012; Matau et al., 2013a; 2013b; Matau et al., 2014; Oancea et al., 2017), the type of pigments (Stos-Gale, Rook, 1981; Niculescu et al., 1982; Pântea, 1984; Burghelea et al., 2003; Constantinescu et al., 2005; 2007; Bugoi et al. 2008; Olaru, 2008; Buzgar et al. 2010a; 2010b; Grămăticu et al., 2010; Boldea et al.

2013; Buzgar et al., 2013), or the composition of the raw materials (Chiribuță, 1979; Sandu et al., 2010; Gâță, 2000; Boghian et al., 2015a; 2015b; Bodi et al., 2015). The *chaîne opératoire* of the Cucuteni pottery and the relationship between pottery production and the development of social complexity was investigated in the seminal work of L. Ellis (1980; 1984). In comparison, only one study was dedicated to the archaeometrical analysis of the Precucuteni pottery (Ellis, 2005).

Târpești is the only site comprising successive habitation layers of Precucuteni and Cucuteni A phases that was entirely investigated (Marinescu-Bîlcu, 1981). The site is located in north-eastern Romania, in a hilly region, on the middle terrace of the left shore of Topolița River (Fig. 1). The small plateau (about 5000 sqm) inhabited in prehistory overlooks the valley above the southern stepped slope and has a slow descent towards north (Marinescu-Bîlcu 1962, p. 235).

Figure 1. Map showing the spatial distribution of the Cucuteni-Trypillia civilization with location of the Târpeşti site (Map image is the intellectual property of Esri and is used herein under license. Copyright © 2019 Esri and its licensors. All rights reserved)

Systematic researches, conducted in 1959-1965 and in 1968, were initiated by Vladimir Dumitrescu and were continued by Silvia Marinescu-Bîlcu. The later processed the field data and published a consistent monograph of the site (Marinescu-Bîlcu, 1981). With the 3200 sqm excavated during the seven years of systematic investigations, Târpeşti stands out as one of the largest and best-documented archaeological project on a Chalcolithic site of the area (Marinescu-Bîlcu, 1981). The archaeological data is complemented by the interdisciplinary approach offering valuable paleo-environmental insights (Marinescu-Bîlcu et al., 1981).

The span of time documented by the archaeological deposit is rather impressive: the oldest horizon is attributed to the Linear pottery culture, followed by two Precucuteni occupation layers, one Cucuteni A, scarce Cucuteni A-B, and Cucuteni B remains. Later on, the site was used as a cemetery, several Early Bronze Age tombs being unearthed during the systematic research; another settlement existed here during the 2nd– 3rd centuries AD, while the latest materials dates to the 6th -7th centuries AD (Marinescu-Bîlcu, 1974). For one of the Precucuteni occupational layers (Precucuteni III) one radiocarbon date is available which falls within 4376 ±80 cal BC (Mantu, 1998).

The cultural layers of interest - Precucuteni and Cucuteni - constitute the most consistent deposits. In the first Chalcolithic layer, several hearths and pits were discovered, while in the second layer (also Precucuteni) ten wattle-and-daub structures and a defence ditch were outlined. Most of the ceramics from the Precucuteni layers is in fragmentary state, collected usually from the outer area of the structures. Among the identified shapes there are globular vessels, stemmed bowls, standing pots, ornamented with a wide variety of techniques: notches, impressions, grooves, projections, incision, excision and rarely the red paint applied before burning (Marinescu-Bîlcu, 1974).

As regards the Cucuteni A layer, another 17 dwellings and a ditch delimiting the settlement were investigated. The ceramic inventory is quite impressive, containing the usual earthenware characteristic to this period: from the simplicity of cups, pedestaled bowls, lids, biconical vessels, to the complex form of wide opened stemmed bowls, supply vessels or clay spoons. A wide variety of decoration techniques were in use: by incision, painting, projections, impressions, and sometimes combinations between these decorative styles (Marinescu-Bîlcu, 1981).

The impressive stratigraphic sequence with several layers dating from the Neolithic to the Early Medieval Period, as well as the wide area of the excavations and the thorough publication transformed this site in an important marker for evaluating the cultural interactions and technological abilities of various communities, which settled in the eastern part of Romania throughout this timespan.

3. MATERIALS AND METHODS

For the present study, we have selected 17 pottery samples (Figure 2) which are typologically and stylistically representative for the Precucuteni (11 samples labelled TRP_PREC) and Cucuteni A ware (6 samples labelled TRP_CUC-A) identified within the site. All the pottery fragments were chosen from known stratigraphic units.

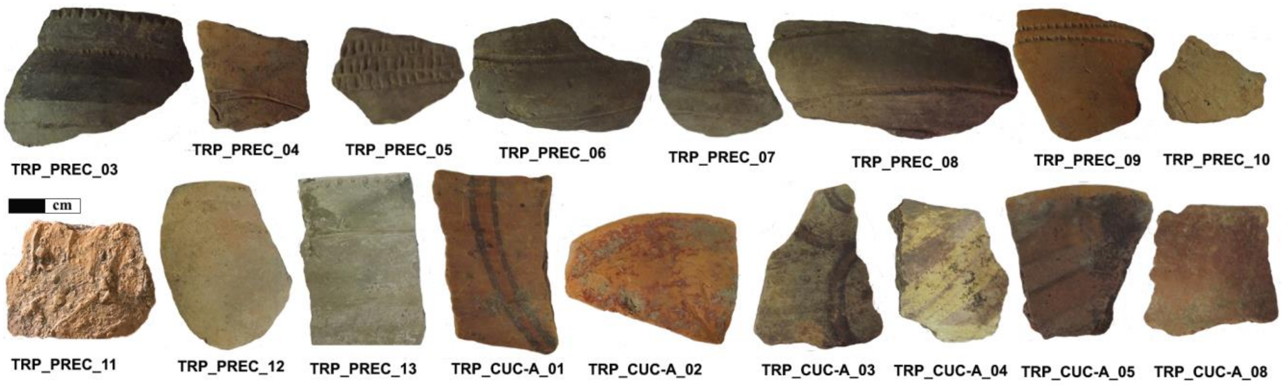

Figure 2. Precucuteni and Cucuteni pottery samples selected from Târpeşti archaeological site

The pottery samples were cleaned with distilled water in an ultrasonic bath, and then we have cut small slices across the ceramic wall for petrographic thin-section analysis using a Meiji ML9430 microscope and following the description system suggested by I. Whitbread (Whitbread, 2017 with references therein) and modified by P.S. Quinn (Quinn, 2013).

The phase composition was determined using a Shimadzu XRPD 6000 diffractometer using CuKα radiation (λ=1.54059 Å) in reflection mode. A small quantity of each pottery sample (2 g) was powdered using an agate mortar and then side-pressed into a top-loaded holder in order to minimize the preferred orientation and analysed in the range of $2\theta=4^{o}$-70^{o} with a scan rate of 0.02^{o} and 1s/step. For all the samples, we have carefully avoided the temper and sampled only the clay matrix, while for sample TRP_CUC-A_08 we have prepared one sample from the matrix (TRP_CUC-A_08a) and one from a fragment of grog temper (TRP-CUC-A_08b). Two samples were prepared, also, for samples TRP_PREC_09 (TRP_PREC_09a from the darker region and TRP_PREC_09b from the more reddish one) and TRP_PREC_13 (TRP_PREC_13a from the darker region and TRP_PREC_13b from the more reddish one). For the identification of the mineral phases, we have compared the samples with the reference powder patterns included in ICDD Powder Diffraction Files (PDF-4). The XRPD data were then statistically treated with the hierarchical cluster analysis (HCA) function of the X'Pert HighScore Plus® software (PANalytical) using the protocol advanced by Maritan *et al.* (2015). For the HCA analysis, we have used the average linkage method and, then, we have separately evaluated the most representative sample of each cluster.

The magnetic properties of the pottery samples were determined with a vibrating sample magnetometer (Princeton Measurements Co. MicroMag VSM&AGM 2900-3900). The maximum field applied was 1.2 T. Measurements were carried out at room temperatures with less than 1 s averaging time per point.

4. RESULTS AND DISCUSSION

Eight of the analysed Precucuteni pottery samples listed in Fig. 2 presents various patterns of incised decoration (TRP_PREC_03, TRP_PREC_04, TRP_PREC_05, TRP_PREC_06, TRP_PREC_07, TRP_PREC_08, TRP_PREC_09 and TRP_PREC_13), one of them being slightly burnished (TRP_PREC_03). Samples TRP_PREC_10 and TRP_PREC_11 shows a rough treatment of the outer surface, while one sample show no traces of decoration (TRP_PREC_12). Five of the selected Cucuteni pottery samples presents various combinations of painted decoration, while one sample show no traces of decoration (TRP_CUC-A_08).

Two of the Precucuteni pottery samples (TRP_PREC_09 and TRP_PREC_13) exhibits a diffused *sandwich* structure ranging from greyish hues on the outer surface to darker hues in the core in sample TRP_PREC_13, while sample TRP_PREC_09 displays reddish hues on the outer surface and darker hues in the core.

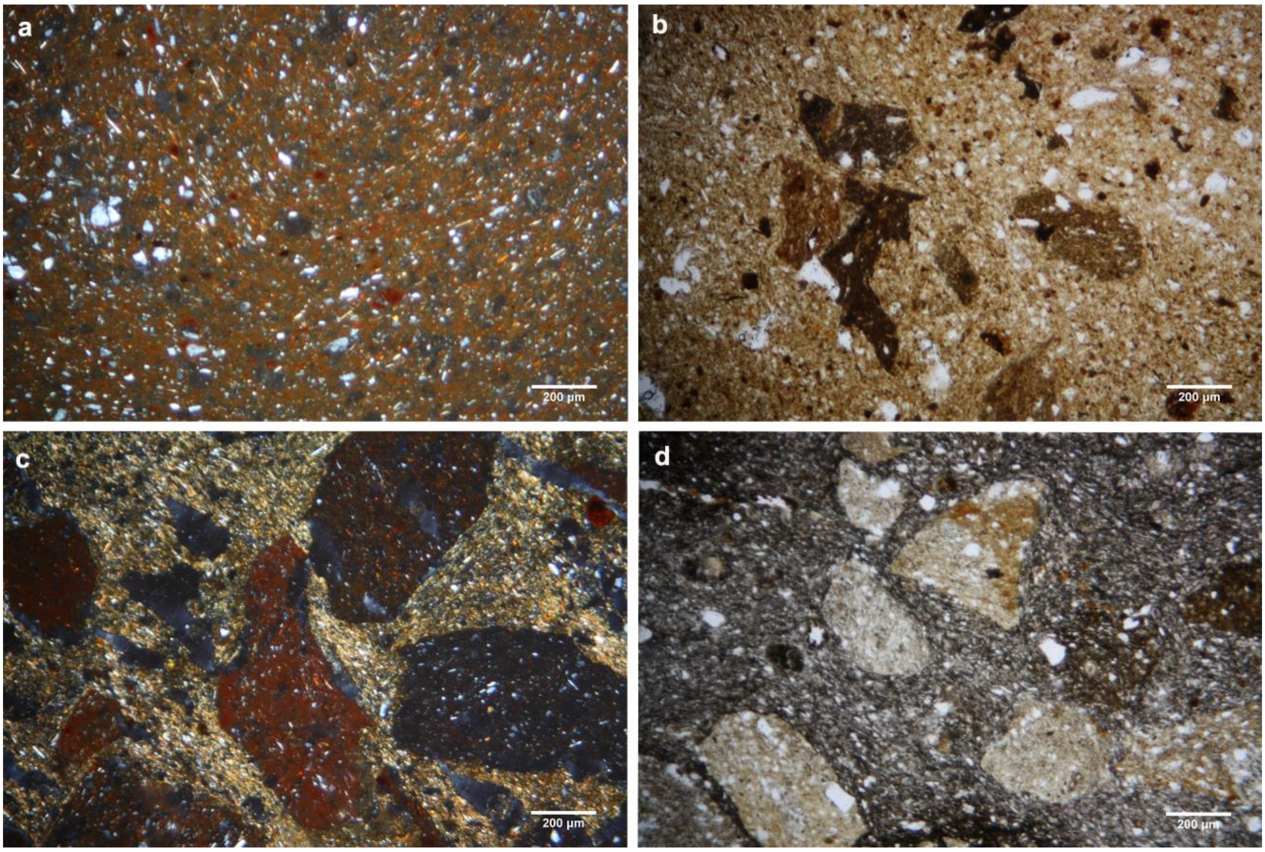

Figure 3. Figure 4: Polarized light microphotos of representative Precucuteni and Cucuteni A ware from Târpești (a – TRP_CUC-A_03, b – TRP_PREC_10, c – TRP_CUC-A_04, d – TRP_PREC_13; Images a and c with crossed polarizers at 4× and images b and d with plane polarizers at 4×)

The Precucuteni and Cucuteni pottery samples analysed in this study were made from a micaceous clay. All the Precucuteni sherds have various amounts and sizes of grog as added temper (Fig. 3). Four of the Cucuteni A ware samples (TRP_CUC-A_02, TRP_CUC-A_04, TRP_CUC-A_05, and TRP_CUC-A_08) were also grog-tempered, while the other two samples (TRP_CUC-A_01, TRP_CUC-A_03,) have no temper at all. The matrix of the Precucuteni pottery samples is homogenous with various colours ranging from greyish hues (TRP_PREC_03, TRP_PREC_13, TRP_PREC_12, TRP_PREC_13), light brown (TRP_PREC_04, TRP_PREC_05, TRP_PREC_06, TRP_PREC_07, TRP_PREC_08, TRP_PREC_10, TRP_PREC_11) to dark brown in sample TRP_PREC_09. The colours of the Cucuteni pottery matrix displays more reddish hues in samples TRP_CUC-A_01, TRP_CUC-A_02, TRP_CUC-A_03, TRP_CUC-A_05 and TRP_CUC-A_08 while sample TRP_CUC-A_04 has a light brown matrix.

Most of the grog fragments identified in the Precuteni and Cucuteni pottery fragments fall into the medium-sand category with sizes no larger than 0.6 mm and with sub-rounded to rounded morphology (Druc, 2015). The percentage of the grog temper in the Cucuteni and most of the Precucuteni samples is around 10%, while for sample TRP_PREC_07 is less than 5%. Mainly sub-angular and sub-rounded quartz grains represent the main mineral inclusions present in the matrix and in the added temper. In addition, all the samples contain feldspars and mica-flakes grains. Different amounts of small red grains, evenly distributed throughout the matrix and the grog temper, are representative for the amorphous iron oxide content. Small grains of calcite showing traces of intense thermal transformation were identified in samples TRP_PREC_04, TREP_PREC_05, TRP_CUC-A_01 and TRP_CUC-A_02, while sample TRP_CUC-A_05 exhibits recarbonatation around the rims of some of the largest grog fragments caused by soil contamination (Fabbri et al., 2014).

Cluster analysis performed on the XRPD data (Fig. 4) showed that the samples grouped into two main clusters: one containing only Cucuteni A samples (TRP_CUC-A_03, TRP_CUC-A_05 and TRP_CUC-A_08) and the other comprising all the Precucuteni samples and three samples attributed to the Cucuteni A chronological sequence (TRP_CUC-A_01, TRP_CUC-A_02 and TRP_CUC-A_04).

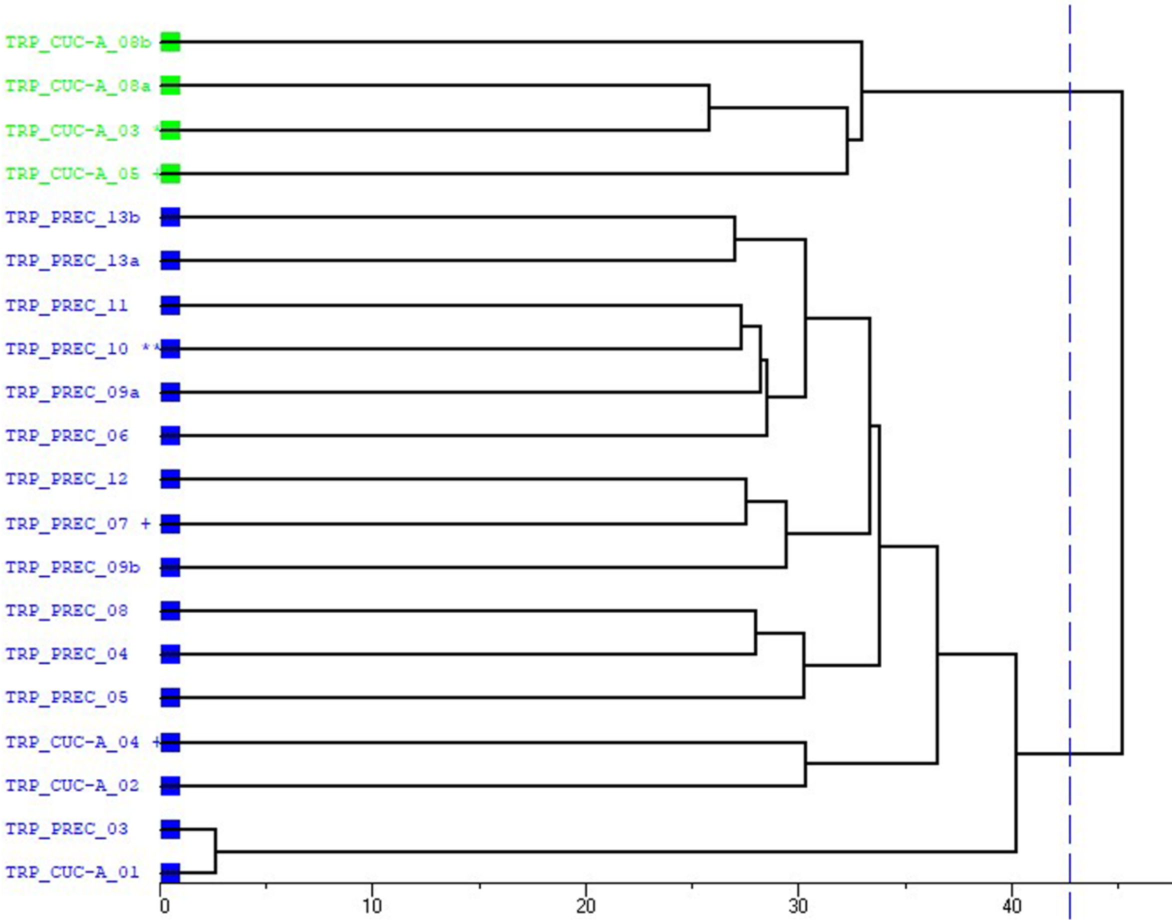

Figure 4. Dendrogram from the HCA of XRPD patterns of Precucuteni and Cucuteni ware from Târpești (TRP_CUC-A_03 and TRP_PREC_10** are the most representative sample within each cluster)*

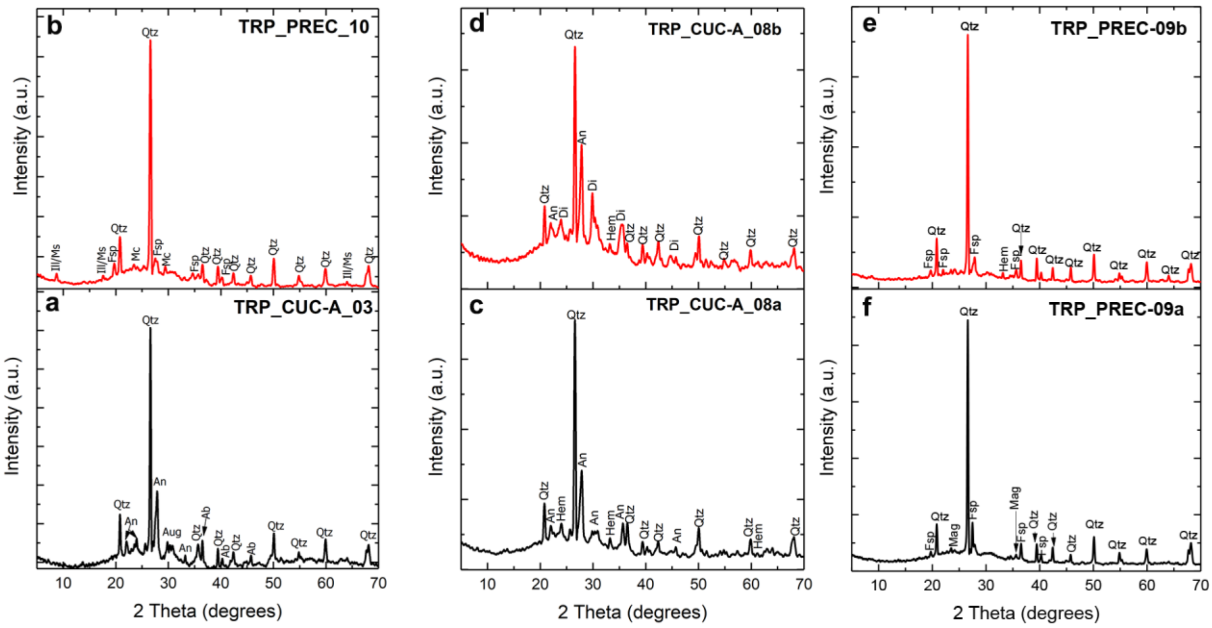

Figure 5. XRPD patterns of the most representative pottery samples for the HCA groups (a, b), for the matrix (c) and grog temper (d) identified in sample TRP_CUC-A_08, and for the sandwich structure identified in sample TRP_PREC_09 (e-black core, f-reddish outer surface) (Abbreviations: Qtz-Quartz, Ill/Ms-Illite/Muscovite, Fsp-Feldpars, An-Anorthite, Di-Diopside, Aug-Augite, Mc-Microcline, Hem-Hematite, Mag-Magnetite, Ab-Albite (Whitney and Evans, 2010)

Detailed analysis of the most representative sample from each group (Fig. 5/a, b) showed substantial mineralogical variation between samples forming the two clusters. The Cucuteni A pottery samples, which forms a separate cluster, exhibits the presence of high-temperature newly formed phases like anorthite and augite, besides albite and quartz, which rests as the dominant mineral phase for all the samples (Fig. 5a). The other Cucuteni and Precucuteni sherds, which fall into several separate subgroups, have lesser amount of newly formed mineral phases. The most representative sample, presented in Fig. 5b, displays the persistence of illite/muscovite diffraction peak in association with feldspar and microcline. The different types of iron bearing minerals determine the several subgroups that are visible for this group (Fig. 5 /e, f).

The structural transformation of the illite clay minerals are almost absent up to temperature of 800ºC (Murad, Wagner, 1996). Under reducing conditions, its decomposition starts at 820ºC (Heimann, 2017), while in an oxidizing atmosphere its decomposition evolves between 850-1000ºC (Nodari et al., 2007). The presence of albite in the pottery samples indicates a temperature around 900ºC (Javanshah, 2019). Anorthite appears due to the Ca incorporation in the newly formed silicate phases at a temperature higher than 850ºC (Cultrone et al., 2001). Another product of the thermally induced reactions is augite, a member of the pyroxenes mineral group with diopside as one of the endmembers (Grammatikakis et al., 2019). The increase of the augite content enhances the vitrification process and is responsible for the buff colour of the pottery sample (TRP_CUC-A_03) caused by the trapping of iron in the augite lattice (Moropoulou et al., 1995). Diopside is a member of the single chain silicate minerals (clinopyroxenes) (Grammatikakis et al., 2019) which forms at temperatures above 900ºC (Trojsi et al., 2002).

Hematite nucleates between 700-750ºC in oxidizing firing conditions, mainly in relation to the massive chlorite and illite decomposition, and is the main pigmentation mineral present in the ceramic body (Nodari et al., 2007). Magnetite appears due to the Fe^{2+} release caused by the breakdown of chlorite (Rathossi, Pontikes, 2010), its presence being a reliable indicator for the reduced firing conditions. When firing under reducing atmosphere, the reduction of Fe^{3+} in the clayey material starts at 400ºC and the Fe^{2+} content continue to raise until reaching 800ºC (Manoharan et al., 2015). Due to the reduced possibility of oxidizing gases to penetrate the ceramic body, the maintenance of Fe^{2+} in oxidative conditions was noticed for temperature above 800ºC (Rathossi, Pontikes, 2010). Also, the presence of small amounts of Fe^{3+} at temperatures between 600-650ºC (Manoharan et al., 2015), or even above 850ºC (Rathossi, Pontikes, 2010) were determined, caused by the reverse phenomena.

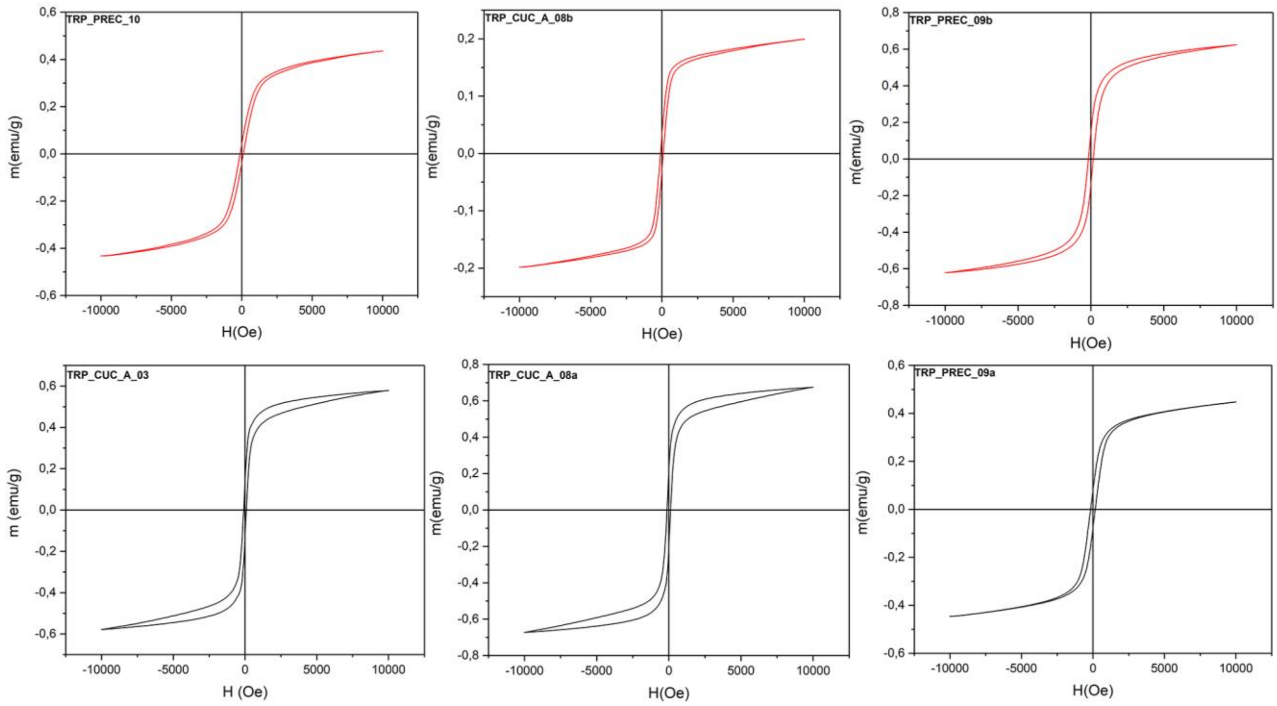

Figure 6. Typical hysteresis curves for representative pottery samples from Târpești

The investigation of the hysteresis parameters provides useful additional information for a more accurate evaluation of the mineralogical transformations induced by the firing process (Beatrice et al., 2008; Jordanova et al., 2019). Hysteresis curve shape is used to evaluate the magnetic domain state and related grain size of the magnetic minerals, mainly in environmental and rock-magnetic studies (Thompson, Oldfield, 1986; Henry et al., 2005). Several examples of major hysteresis loops are shown in Fig. 6. Samples TRP_CUC-A_03, TRP_CUC-A_08a and TRP_PREC_09b presents a distorted loop suggesting a significant addition to magnetization due to the presence of hematite. For two of the samples (TRP_CUC-A_08a and TRP_PREC_09b) the hematite content was determined, also, by the XRPD measurements, while in sample TRP_PREC_03 although iron bearing minerals were not detected, the Fe^{3+} and Fe^{2+} cations are included in the *M1* site of augite (Buzatu, Buzgar, 2010).

More tight loops are displayed by samples TRP_PREC_10, TRP_PREC_09a and TRP_CUC-A_08b caused, most probably, by the single domain contribution of magnetite. The presence of magnetite was identified by the XRPD measurements only in sample TRP_PREC_09a that was taken from the darkest region of the *sandwich structure* displayed by the pottery fragments in cross-section. The presence of magnetite suggests poor oxygen diffusion in the potsherds during firing (Nodari et al., 2004; Maritan et al., 2006). The poor diffusion of oxygen in the pottery matrix may be caused by the general reducing condition in the kiln or to the formation of amorphous phase and prevention of oxygen penetration. During the formation of amorphous phase and its vitrifying as a result of destruction of phyllosilicates, pores were filled by the viscous product of sintering and can further cause poor diffusion of oxygen in the pottery matrix. At higher temperature exposure, Fe released ions enter into the structure of newly formed silicates such as pyroxenes (Noghani, Mohammadamin, 2014). The diffraction pattern of sample TRP_CUC-A_08b (Fig. 5/d) revealed the formation of high temperature pyroxenes like diopside, which caused the blocking of oxygen. In sample TRP_PREC_10 illite/muscovite diffraction peaks are still present, indicating that the firing temperature was below 850ºC. Although the lighter colour of the pottery indicates oxidative firing conditions, the tighter shape of the hysteresis loop indicates that the firing conditions varied from oxidized to reduced conditions (Manoharan et al., 2014).

Based on the optical microscopy, statistical evaluation of the XRPD data and the differences observed in the shapes of the MHL we can estimate that the largest ceramic assemblage grouping all the Precucuteni and three Cucuteni A potsherds were fired in the 750-800ºC temperature range. The use of micaceous clay combined with various quantities of crushed sherds were previously reported for the Precucuteni pottery identified at Târgu Frumos-*Baza Pătule* (Iași County, eastern Romania) (Ellis, 2005). Another type of raw material containing oolithic clay with a sand/quartz fraction and the addition of limestone and some organic matter was, also, reported for Târgu Frumos (Ellis, 2005).

The firing temperature attained for the group containing only Cucuteni A samples falls between 850-900ºC. The results obtained for the group consisting of Cucuteni A samples are in agreement with our previous results, which reported that the majority of the potsherds were fired between 800-900ºC (Matau et al., 2013a; Matau et al., 2014).

5. CONCLUSIONS

Through minero-petrographic observations combined with magnetic properties, we were able to identify the technological parameters of the Precucuteni and Cucuteni pottery selected from Târpești in terms of raw materials selection, preparation and firing process. Although our database was not an extended one, we could notice the use of a similar type of micaceous clay for all the investigated pottery samples. All the Precucuteni samples contains various amounts and sizes of grog, a recipes used also by the Precucuteni communities located at Târgu Frumos. The use of grog as tempering agent was noticed, also, for Cucuteni A painted ware. Only two Cucuteni A samples were produced without the addition of any tempering material. Although displays a similar pattern in terms of raw material selection, the Precucuteni communities experienced a wide range of firing conditions, from exclusively reducing to oxidative ones. The wide spectrum of firing conditions seems to be the result of intentional experimentation as the firing temperature falls within the same range.

Two distinct groups were noticed based on the statistical treatment of the XRPD data associated to different firing temperatures. As previously reported by Maritan et al. (2015), the two groups tends to be more related to the firing temperature, than to the petrographic grouping of the pottery. The addition of magnetic properties allowed the refinement of the evaluation of the mineralogical transformations induced by the firing process.

In the next stage of our research, we intend to extend the database in order to be able to relate more explicitly the technological parameters identified for the pottery to the more general social transformations specific to the Early Chalcolithic timespan.

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