

Archaeometric comparison between the Neolithic pottery of different cultures at the archaeological site of Alba Iulia (Transylvania, Romania)

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ABSTRACT. A large quantity of pottery has been discovered at the Lumea Nouă Neolithic and Eneolithic settlement in Alba Iulia City (Romania). It belongs to different cultures, and in this paper a comparison among three of them is made: Vinča, Lumea Nouă and Foeni. An analogous number of samples has been selected for each of the three cultures, together with some clay samples collected in the Alba Iulia settlement area. The samples have been analysed by means of XRF, XRPD, and in thin section. Significant differences among the three cultures are only related to the alkalis contents. The Lumea Nouă pottery has higher potassium contents and contains microfossils, while the Foeni pottery differs to each other for the higher sodium contents. A clear correspondence between pottery and clays has not been found, so that we hypothesize that not all the pottery is local or, more probable, that a geological gradual variation in local clay deposits existed.

Key words: Romania, Alba Iulia, Neolithic pottery, Clays, Archaeometry

INTRODUCTION

Contrary to other European regions, in Romania only few investigations on prehistoric pottery have been made using modern techniques (Ghergari et al., 1999; Lazarovici et al., 2002; Goleanu et al., 2005; Fabbri et al., 2008; Varvara et al., 2008). In spite of the large quantity of ancient ceramic material collected from archaeological excavations, there are still many unknown aspects about the origin and the production techniques of the prehistoric pottery discovered on the actual Romanian territory.

In the last years, due to the rescue excavations made at the Lumea Nouă Neolithic and Eneolithic settlement, in the NE part of Alba Iulia city, which is located about 75 km south of Cluj Napoca in straight line (Fig. 1), a large quantity of ceramic fragments has been discovered, belonging to different cultures (Ciuta et al., 2007). The aim of this paper is to make a comparison among three of them: Vinča (5500-4700 B.C.), Lumea Nouă (5200-4900 B.C.) and Foeni (4800-4500 B.C.). The archaeological study led to the conclusion that Lumea Nouă painted pottery is associated with incised Vinča pottery, while Foeni pottery has always been recovered in a well distinct long-term habitation site.

The pottery of the old phase (A) of the Vinča culture (Fig. 2a) is characterized by barbotine pottery without chaff and well fired, a large quantity of fine black or greyish black pottery decorated with pleats and grooves, bowls with

prominent corner point and thicker walls. Black-topped pottery and the scarceness of incised decoration are also typical of this phase. During the second phase (B), the quantity of the black burnished pottery decreases, replaced by the greyish and yellowish pottery (Fig. 2b). The quality of the black-topped pottery lowers, while the decoration with incised lines and the incised dotted band become more frequent. The new wave of population leads, during the third phase (C), to the emergence of fine pottery from greyish to reddish in colour. It is also well burnished, having a metallic lustre and silvery shades.



Fig. 1. Location of Alba Iulia and Limba sites.

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The pottery of the Lumea Nouă culture is characterised by painted decorations applied before firing (Fig. 2c). They consist of sets of red or brown-chestnut lines, parallel, vertical, oblique or horizontal, applied over the slip background. Lumea Nouă painted pottery is made of a homogeneous paste that has no impurities, but contains fossil relics; the slip is mostly yellowish white and is made of carbonate clay with a high content of illite. Finally, it is

worth mentioning the particular category of the bitumen-painted pottery, where the bitumen is applied after firing.

Foeni communities produced very good quality pottery, by using fine clay without impurities. Significant quantity of well burnished vessels, some of them being black-topped fired, has been found (Fig. 2d).

Painted decorations applied on the vessels before firing, without any slip, are also typical.

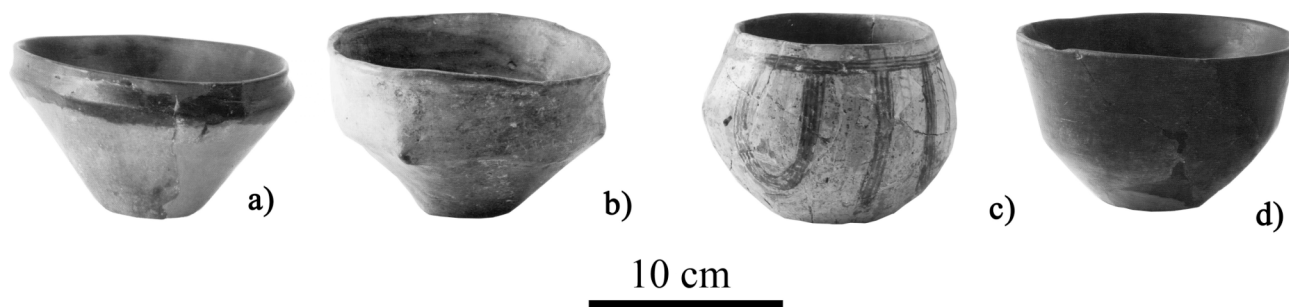


Fig. 2. Pottery examples: a) Vinča A; b) Vinča B; c) Lumea Nouă; d) Foeni.

ARCHAEOLOGICAL INVESTIGATION

In order to make the comparison as significant as possible, an analogous number of samples was selected for representing each of the three cultures. Because pottery attributed to Vinča culture is not so diffused in the archaeological site of Alba Iulia as that attributed to Lumea Nouă and Foeni cultures is, the number of shards representative of Vinča culture were increased by selecting several samples from the near Neolithic site of Limba, 3-4 km far from Alba Iulia. In addition, some clay samples have been collected in the Alba Iulia settlement area, in order to compare them with the pottery.

Then, all the selected ceramic fragments and clay samples were characterized by determining their chemical composition. This type of investigation was preferred because the majority part of the pottery has a fine paste and because it easily allows to compare pottery and clays. In addition to chemical analysis, some optical observations on thin sections and crystalline phase analysis by X-ray powder diffraction (XRPD) were also performed.

MATERIALS AND TECHNIQUES

In this paper 52 pottery samples have been taken into account, subdivided as follows:

- Vinča: 4 samples from Alba Iulia and 12 from Limba,
- Lumea Nouă: 18 samples, all from Alba Iulia,
- Foeni: 22 samples, all from Alba Iulia.

Four clay samples have been also considered, which were collected at different depth in the archaeological area of Alba Iulia. All the samples have been analysed from the chemical point of view, by determining the major and minor elements concentration, phosphorus included, by means of X-ray fluorescence spectrometry. To this purpose, a small piece of sample was detached from each shard, and its surface abraded in order to eliminate slip and painted decorations or eventual alteration material. The clean piece was powdered (<100 μm) and one gram of powder mixed with nine grams of lithium meta/tetraborate (flux). The

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mixture was then melted at about 1000°C and cooled in order to obtain a vitreous disk of 4 cm in diameter and about 5 mm thick.

Phosphorus was determined because it is often present in remarkable amount due to contamination, especially in the oldest pottery (Fabbri and Gualtieri, 2008; Maritan and Mazzoli, 2004). For the same reason, the weight loss of the dried sample powder, after ignition at 1000°C for two hours, was also measured. In order to make the comparison between the different samples and between pottery and clays possible, the chemical results were normalized excluding phosphorus and ignition loss values.

RESULTS

Vinča culture

The chemical compositions found for the samples representative of this culture are listed in Table 1, where the samples coming from Limba archaeological site (LV 1-12) are distinguished from those coming from Alba Iulia (ALNV 13-16). The first thing to be noticed is that there is a substantial homogeneity among the samples, and no difference can be evidenced between the samples from Alba Iulia and Limba. Only three small exceptions can be pointed out:

- the higher calcium content of the samples LV11 and LV12 (>6 wt.% CaO instead of an average of about 2-2.5 wt.% of the others),
- the relatively higher sodium content of the sample LV5 (1.48 wt.% Na₂O against an average less than 1 wt.%).

These values possibly are normal variations for a natural clay deposit, but they could also be the result of an alteration process, especially in the case of calcium oxide. Unfortunately, we did not made optical observations on thin sections for the samples LV11 and LV12, so that it was not possible to ascertain if the CaO is present as primary calcite or secondary calcite. For these reasons, we preferred to calculate the statistical parameters (arithmetical mean and standard deviation) representative of the Vinča pottery only by taking into account 13 samples (Table 2).

Table 1. Chemical composition of the Vinča pottery (wt.% oxides) recalculated to 100% after subtracting P₂O₅ and LOI (Loss On Ignition).

Sample	SiO ₂	Al ₂ O ₃	TiO ₂	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	LOI
LV1	65.96	17.67	0.84	6.90	0.12	2.65	1.99	0.88	3.00	0.55	6.56
LV2	67.47	17.40	0.89	6.14	0.11	1.64	2.66	0.79	2.90	1.20	6.17
LV3	63.79	18.81	0.91	7.60	0.14	2.55	2.25	0.86	3.09	0.56	6.93
LV4	67.70	18.58	0.96	5.44	0.06	1.91	1.89	1.00	2.46	0.57	3.76
LV5	62.40	20.63	0.99	6.40	0.08	2.97	1.92	1.48	3.13	0.25	2.36
LV6	66.40	17.49	0.83	6.62	0.17	2.24	2.14	0.92	3.20	0.70	4.14
LV7	65.54	17.95	0.89	6.69	0.12	2.37	2.64	0.96	2.83	0.18	5.41
LV8	67.66	17.29	0.84	6.39	0.11	2.23	1.54	1.03	2.91	0.17	3.23
LV9	68.52	16.94	0.90	6.03	0.11	1.95	1.86	0.97	2.72	0.63	5.11
LV10	67.47	16.85	0.82	5.33	0.07	2.27	3.24	0.96	2.98	0.67	5.77
LV11	62.87	17.73	0.87	6.11	0.06	2.55	6.02	0.99	2.80	0.73	4.24
LV12	63.64	17.16	0.81	6.13	0.13	2.34	6.36	0.85	2.59	0.24	6.87
ALNV13	64.36	18.94	0.90	7.49	0.18	2.44	2.16	0.99	2.53	0.50	8.49
ALNV14	65.71	18.84	0.92	7.24	0.05	2.08	1.58	0.99	2.58	0.33	1.95
ALNV15	64.83	18.67	0.91	7.25	0.08	2.40	2.14	0.88	2.85	0.59	5.37
ALNV16	68.89	16.84	0.92	5.43	0.06	2.09	2.11	0.91	2.76	0.26	2.16

Lumea Nouă culture

The chemical composition of this pottery has been already published (Varvara et al., 2008). It has pointed out that the data are very homogeneous, and only two anomalies have been registered, that is the lower quantity of potassium in the samples LN1 and LN9 (about 2 wt.% K₂O against more than 3 wt.% of all the others).

On the contrary, the thin section observations evidenced two groups of samples, of similar consistence, which are characterized by absence/presence of microfossils. The amount of microfossils, estimated by comparative method, varies from traces to some 10%, but their presence has no

practical influence on the chemical composition of the pottery samples because of their siliceous composition, as evidenced by some measurements made by scanning electron microscope equipped with energy dispersion spectrometer.

Due to the gradual variation of the microfossil content, it is possible to hypothesize that the clay sediments used by the potters were characterised by a gradual increase/decrease of microfossils content in depth. Therefore, we decided to calculate the arithmetical mean and standard deviation representative of the Lumea Nouă pottery taking into account 16 samples, only excluding samples LN1 and LN9 (Table 2).

Table 2. Chemical compositions (average, ± = standard deviation) of the pottery of the three cultures and clays (wt.% oxides).

Sample	SiO ₂	Al ₂ O ₃	TiO ₂	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	
Vinča	66.48	17.87	0.89	6.50	0.11	2.21	2.17	0.93	2.83	
	±1.61	±0.81	±0.04	±0.79	±0.04	±0.28	±0.46	±0.07	±0.22	
Lumea Nouă	64.65	18.24	0.82	6.72	0.11	2.55	3.09	0.78	3.33	
	±2.22	±1.10	±0.05	±0.71	±0.05	±0.27	±1.38	±0.20	±0.34	
Foeni	67.16	17.81	0.85	6.20	0.10	1.92	1.87	1.37	2.66	
	±1.65	±1.04	±0.03	±0.56	±0.03	±0.24	±0.40	±0.13	±0.26	
Clays	AB1	69.94	16.85	0.90	6.11	0.13	1.47	1.26	1.10	2.24
	AB2	69.16	17.10	0.92	6.43	0.13	1.77	1.27	1.14	2.08
	AB8	71.43	16.28	0.80	5.61	0.11	1.47	1.57	0.86	1.86
	AB9	66.42	16.50	0.77	5.60	0.10	1.76	5.66	1.00	2.20

Foeni culture

The chemical composition of this pottery has been already published (Fabbri et al., 2008). It has pointed out that a set of 17 very homogeneous samples exists, but five samples (AFO1, AFO3, AFO5, AFO6, AFO7) are significantly different, due to their high calcium content (5 to 13 wt.% CaO) in comparison with an average CaO percentage less than 2 wt.% of the main group. This subdivision is confirmed by the observations in thin section, which revealed that the anomalous samples have a very fine and homogeneous microstructure, except for AFO7 characterized by the presence of an abundant and well-sorted (150-200 µm) temper, made of quartz and feldspar grains. In conclusion, we retain that the most representative pottery for the Foeni culture is given by the statistical data of the main group (Table 2).

Local clays

The data of the four clay samples, already published (Fabbri et al., 2008), are again reported in Table 2. It is

interesting to note that only the clay sample AB9 shows some differences if compared with the other three, due to a lower silica content (about 66% against 70%) and a higher amount of calcium oxide (5.66 wt.% instead of 1.3-1.6 wt.%).

DISCUSSION

As it has been said before, the pottery of each culture has sufficiently homogeneous chemical composition, whose statistical parameters are listed in Table 2. So, they identify 'chemical groups' which could be easily transformed into true 'chemical reference groups' by increasing the number of samples analyzed for each culture. These data reveal that the differences among the cultures are generally not significant, as it can be seen, for example, in the binary diagram Al₂O₃/SiO₂ shown in Fig. 3. Only the contents of sodium and potassium give interesting differences, as demonstrated by the correspondent binary diagram shown in Fig. 3.

The Lumea Nouă pottery has higher potassium content with reference to Vinča and Foeni ones, while Foeni pottery has higher sodium content than the other two. Therefore, the

composition of the oldest culture (Vinča) is located between the other two, so that Lumea Nouă pottery and Foeni pottery are clearly separated between them. If we also take into consideration that most of the Lumea Nouă pastes also contain microfossils, it should be deduced that the source of their clay raw materials is different from the others.

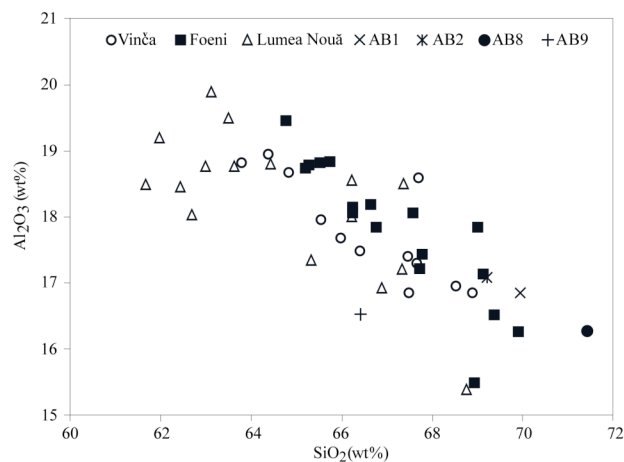


Fig. 3. Binary correlation diagrams of the different cultures and collected clays.

It does not mean that all the pottery cannot be local. A geological vertical variation in the clayey deposits could justify this situation, according with a limited variation of the supply areas. This hypothesis seems to be more likely than the possibility of a different processing of a same raw material. The hypothesis of a differentiation due to mobility of sodium and potassium during the ceramic firing is to be rejected, because it would imply a decrease of the contents of sodium and potassium in the ceramic paste in comparison with the clays. On the contrary, all three cultures show a noticeable higher content of potassium in the ceramic paste, while sodium is higher for the Foeni culture and similar or slightly lower for the other two cultures.

Therefore a deep investigation on the geological characteristics of the local clay deposits is necessary before concluding that all the pottery productions are local, or if some of them is not.

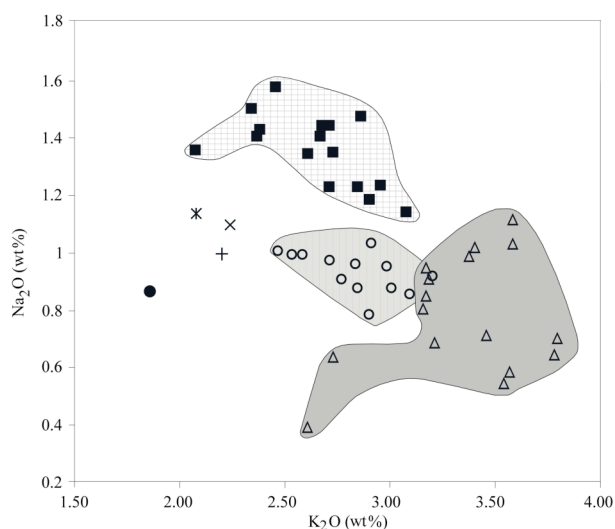
CONCLUSION

The main results of this study on Neolithic Romanian cultures from Alba Iulia settlement are the follows:

- 1) Significant differences among the three cultures are only related to the alkalis contents;
- 2) The Lumea Nouă pottery differs for the higher potassium contents and for the microfossils presence in the paste of many samples;
- 3) The Foeni pottery differs to each other for the higher sodium contents;
- 4) There is not a clear correspondence between pottery and clays. Therefore it is possible to hypothesize that not all the pottery is local or, more probable, that a geological gradual variation, probably vertically, in clay deposits existed.

Rest assured that a deep geological study of the area of the settlement and there around is necessary to confirm the last hypothesis.

In the same diagrams shown in Fig. 3, the representative points of the local clay samples are also plotted. They are not too much far away from the composition intervals defined for Vinča and Foeni pottery, but very far from Lumea Nouă pottery. This observation appears to be a support to the above statement referred to the source of the clays.



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