MYCENAEAN POTTERY FROM THE ARGOLID AND ACHAIA—A MINERALOGICAL APPROACH WHERE CHEMISTRY LEAVES UNANSWERED QUESTIONS*

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The chemical classification of Mycenaean pottery from the Northern Peloponnese by neutron activation analysis (NAA) resulted in two groups with very similar chemical compositions assigned to production centres in the Argolid and in Achaia, respectively. The statistical separation of these two groups on the basis only of their chemical composition was difficult, and not clear-cut for all of the examined samples. A complementary mineralogical examination by X-ray diffraction (XRD) of some selected samples indicated differences in the mineralogical composition, which confirmed the determined chemical differences. Furthermore, a clay sample was examined, which showed a composition similar to that of the pottery.

KEYWORDS: MYCENAEAN POTTERY, CLAY PROSPECTING, PROVENANCE, NEUTRON ACTIVATION ANALYSIS, X-RAY DIFFRACTION

INTRODUCTION

Within a long-term project on Mycenaean pottery (Mommsen *et al.* 1995; Maran *et al.* 1997), a large number of samples from different regions in Greece have been analysed by NAA in Bonn: among these were also samples from the Argolid and Achaia. The predominant chemical pattern, which was found in the samples from the Argolid, was named Mycenae/Berbati (MB) (Mommsen *et al.* 1988), according to distribution arguments. Presumably this pottery group was produced in the northern Argolid (Fig. 1). In the following years, this pattern has been found in many sherds from all the Argolid sites sampled; for example, Mycenae, Berbati, Midea, Tiryns, Argos and others. Meanwhile, the provenance question of this pattern has been verified as a result of new evidence. Measurements of several wasters from the Mycenaean workshop at Berbati supplied to the Bonn laboratory by B. Wells (unpublished data) ascertained the assignment of the MB pattern to this workshop.

However, it has to be mentioned that chemical compositions similar to that of the MB group have also been found in pottery from other Greek sites, and even in pottery from the Troas (Knacke-Loy 1994; Mommsen *et al.* 2001). The archaeological classification contradicted the idea of a common origin for all of these products. Because of the similarity of the MB pattern to the patterns of other groups of pottery, the initial differentiation between them created problems. Nevertheless, after the application of the statistical data evaluation that is used in Bonn (Beier and Mommsen 1994), the patterns were always distinguishable.

Recent measurements of samples from Achaia (Mommsen *et al.* 1997) have revealed a new pattern named ACH-a, which proved to be very similar to the MB pattern. The two patterns

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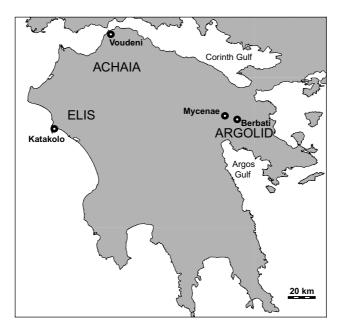


Figure 1 A map of the Peloponnese. The sites Mycenae/Berbati and Voudeni and the clay deposit of Katakolo are indicated.

show small but significant differences for only a few elements (Ca, Na, K, Rb and Cs). It turned out that the separation of the two patterns was not possible after the application of our usual statistical data evaluation procedure (Beier and Mommsen 1994). This procedure works like a filter, sorting out all samples of similar composition from a given data bank using 2σ confidence levels. The additional application of a best relative fit factor takes into account measurement errors as well as possible dilutions of the clay paste by, for example, quartz or calcite. As is usual, Ca and Na were disregarded during the statistical grouping procedure due to their commonly large spreads, since they are often related to the non-plastic tempering material. A separation was achieved only when the method of the 'core group' formation was applied, using 1σ confidence levels (Mommsen 2001). For a number of samples found in Achaia and in the Argolid, respectively, a clear-cut group assignment was still not possible, since the assignment probabilities were small and about the same when considering both patterns.

Such a close similarity between two patterns had previously been observed among the MB and MBKR groups (MBKR: slight deviation in K and Rb compared to MB); they were both assigned to the Argolid but in fact they comprised one group, the reference group for Mycenae/Berbati (Mommsen *et al.* 1996; Buxeda *et al.* 2002). For these two groups the differences in their chemical compositions were observed in the concentration of the alkaline elements, and after mineralogical examination of the samples these differences could be explained as a result of post-burial alteration (Buxeda *et al.* 2002).

For the specific case of the MB and the ACH-a groups, however, another aspect had to be borne in mind. From an archaeological point of view, an extensive unilateral exchange of pottery between the two regions was rather improbable. Furthermore, the bulk of the Achaian material chosen to be analysed dates to the Late Mycenaean period, more specifically to LHIIIC (Mommsen *et al.* 1997) whereas the MB pattern covers the whole range of the LH period. The pottery of this phase shows several stylistic peculiarities, which indicate a local Achaian manufacture (Papadopoulos 1979; Mountjoy 1990). In order to confirm the archaeological assumption, to ensure the purely chemical core-group separation into two regional groups and to exclude the possibility of dealing with a case of post-burial alteration, additional mineralogical examinations of some selected samples, as well as of a raw material, are presented below.

THE SAMPLE CHOICE AND METHOD FOR XRD MEASUREMENTS

For the mineralogical examination, four samples of pottery from Berbati (Argolid) and seven samples from Voudeni (Achaia) were selected (Fig. 1). The samples represented the chemical groups MB and ACH-a, respectively. They were chosen at random, mainly according to the amount of powder left after the NAA analysis. Additionally, a clay sample from a Pliocene marine deposit close to Katakolo on the west coast of the Peloponnese (Elis) was included (Fig. 1). According to the NAA results, this clay was found to chemically resemble the composition pattern of the pottery from Achaia. The average NAA compositions of the groups of the four samples from the Argolid and the seven from Achaia, respectively, and of two Katakolo clay samples are shown in Table 1 together with their spreads (σ = root mean square deviation).

Table 1 Element concentrations of the groups of four samples from Berbati (Argolid, MB), seven samples from Voudeni (Achaia, ACH-a) and two samples from the Katakolo clay (same bulk). The average values M are given in parts per million (ppm), if not indicated otherwise, and spreads (σ) in percentages of M. The values are corrected for dilution to the main group MB using 25 elements (all except As, Ba, Ca, Na and Cs)

Element	Berbati four sa		Voudeni (seven se	, · ·	Katakolo clay, two samples		
	М	σ(%)	М	σ(%)	М	σ(%)	
As	6.01	28	3.00	51	5.37	4.4	
Ва	545	39	376	17	239	28	
Ca (%)	9.95	13	3.94	51	11.2	2.3	
Ce	62.3	3.0	63.6	4.4	62.1	1.7	
Co	28.7	4.8	27.8	6.1	24.8	2.1	
Cr	198	5.2	215	3.2	237	0.7	
Cs	8.77	5.3	6.81	7.4	7.26	2.2	
Eu	1.34	2.6	1.17	3.3	1.23	2.5	
Fe (%)	5.25	1.9	5.05	5.0	4.64	1.3	
Ga	18.1	25	21.8	7.2	17.0	10	
Hf	3.49	5.0	3.78	8.8	3.82	3.4	
K (%)	2.69	6.8	2.53	3.8	2.45	1.5	
La	32.1	1.6	30.1	4.9	30.1	0.3	
Lu	0.44	6.3	0.42	15	0.42	4.0	
Na (%)	0.44	12	0.98	13	1.80	0.5	
Nd	27.8	4.8	25.7	9.2	27.6	14	
Ni	235	7.0	195	6.7	199	5.7	
Rb	151	7.0	142	6.6	144	1.9	
Sb	0.63	11	0.61	8.2	0.76	26	
Sc	21.1	0.9	21.4	5.1	19.9	1.4	
Sm	5.14	1.9	4.99	9.6	5.36	0.8	
Та	0.81	5.8	0.84	5.5	0.78	4.2	
Tb	0.72	5.7	0.72	5.8	0.79	6.8	
Th	11.0	1.5	11.1	5.6	10.4	2.8	
Ti (%)	0.47	16	0.45	9.4	0.62	62	
U	2.39	3.9	2.56	18	2.76	7.5	
W	2.45	9.5	2.06	15	2.13	7.8	
Yb	2.80	4.3	2.89	7.4	2.71	2.9	
Zn	103	5.6	125	14	102	2.5	
Zr	118	26	190	18	187	27	

 Table 2
 Element concentrations C in ppm, if not indicated otherwise, and average errors, in percentages of C, for the

 13
 individual samples. The division by the indicated factor gives the raw NAA data. The values in parentheses indicate outliers

Sample	Factor	As	Ba	Ca (%)	Ce	Со	Cr	Cs	Еи	Fe (%)	Ga	Hf	K (%)	La
				(/0)						(///			(,0)	
Berb 8	1.07	8.10	839	11.0	61.5	28.1	185	8.20	1.55	5.12	18	3.50	2.96	32.4
Berb 17	0.94	6.66	560	9.2	61.4	30.7	208	9.24	1.30	5.22		3.25	2.54	31.9
Berb 18	1.01	4.86	352	11.0	61.2	28.4	195	8.60	1.30	5.31		3.57	2.64	31.5
Berb 23	0.98	4.41	428	8.6	64.9	27.6	204	9.05	1.24	5.34	(66)	3.65	2.63	32.7
Voud 3	0.95	2.67	330	1.7	62.9	25.9	215	6.50	1.18	4.91	16	3.68	2.60	30.4
Voud 4	0.96	3.90	304	2.6	66.6	30.1	227	6.43	1.23	5.38	23	3.76	2.51	31.7
Voud 22	0.99	2.59	453	7.9	61.9	25.8	220	6.50	1.16	4.63	21	3.90	2.47	29.1
Voud 25	0.97	5.95	375	4.4	61.5	27.4	211	6.49	1.13	4.99	24	4.05	2.50	28.4
Voud 30	0.94	1.24	341	4.6	60.2	28.2	206	7.60	1.12	5.32	21	3.17	2.70	28.4
Voud 34	0.95	2.40	359	3.0	64.1	27.1	217	6.71	1.19	5.04	21	4.22	2.40	30.6
Voud 35	0.95	2.24	486	3.4	67.8	29.9	211	7.46	1.20	5.08	23	3.71	2.54	31.9
Kata cl	1.26	5.23	216	11.2	61.5	24.4	236	7.17	1.21	4.60	17	3.74	2.47	30.1
Kata cl r	1.25	5.57	265	10.7	63.0	25.2	238	7.40	1.25	4.69	(48)	3.93	2.41	30.2
Exp. error		0.11	37	0.4	0.6	0.2	1	0.10	0.03	0.02	2	0.07	0.04	0.2
In %		2.6	9	6.2	0.9	0.5	0.6	1.4	2	0.4	8.8	1.9	1.6	0.5

To facilitate the comparison, a best relative fit for the three data sets to the mean values of the respective groups was performed, and then the two patterns ACH-a and Katakolo were matched to the MB pattern. The raw data are listed in Table 2, corrected by the best relative fit factors.

The mineralogical analysis of the powdered samples by XRD was carried out at the Institute of Materials Science of the N.C.S.R. 'Demokritos'. In the case of the clay samples, besides the bulk material, the clay fraction (particle size < 6 μ m) was analysed as well, in three different preparations. The powdered samples of the pottery and the clay were measured with a Siemens D 500 spectrometer with a Cu–K_a source in an angular range of 4–70° 2 θ and with a step width of 0.03° 2 θ s⁻¹. The clay fractions from Katakolo were measured in an angular range of 2–40° 2 θ and 2–20° 2 θ , respectively. The orientated aggregates of the clay samples were prepared by pipetting a clay–water suspension on to a slightly roughened glass sample-holder and leaving it to dry afterwards. Apart from the simply air-dried sample, one orientated aggregate was heated at a temperature of 550°C for 1 h, and another one was saturated in an ethylene-glycol atmosphere. The latter two preparations affect specific phyllosilicate minerals in a different way and enable a distinction to be made between them (Brindley and Brown 1980, ch. 5).

THE XRD RESULTS OF THE POTTERY

The XRD spectra of the samples from Berbati are shown in Figure 2. In samples Berb 8, 18 and 23, peaks of illite/muscovite (I/M), quartz (Q), plagioclase (P), pyroxenes (D), gehlenite (G), hematite (H) and traces of K-feldspar (KF) can be found. Besides these minerals in Berb 8 and Berb 18, calcite (C) can be identified and apparently traces of analcime (An, the marked peak corresponds to a magnesian analcime). In view of the presence of pyroxenes and gehlenite, which indicate an equivalent firing temperature (EFT) of over 850°C, calcite is most probably

Lu	Na (%)	Nd	Ni	Rb	Sb	Sc	Sm	Та	Tb	Th	Ti (%)	U	W	Yb	Zn	Zr
0.47	0.50	25.8	231	134	0.73	21.0	5.15	0.77	0.75	10.9	0.38	2.38	2.41	2.87	(68)	
0.44	0.38	29.2	247	155	0.64	21.0	5.18	0.83	0.69	10.9	0.55	2.45	2.53	2.63	101	107
0.43	0.42	27.4	247	157	0.55	21.0	4.99	0.77	0.68	10.9	0.47	2.47	2.61	2.77	109	151
0.41	0.47	28.4	213	155	0.64	21.4	5.22	0.86	0.75	11.2	0.50	2.28	2.35	2.89	98	98
0.47	1.11	22.7	187	141	0.60	21.4	4.45	0.89	0.71	11.1	0.49	2.39	2.29	2.93	127	251
0.46	1.03	24.5	220	139	0.62	22.7	4.58	0.87	0.69	12.1	0.51	2.52	2.65	3.17	128	208
(1.29)	0.93	27.5	182	129	0.62	19.7	5.16	0.79	0.71	10.6	0.39	2.19	1.90	2.74	102	162
0.46	1.06	24.2	186	134	0.62	20.4	4.79	0.83	0.73	10.5	0.42	2.49	1.93	2.78	102	178
0.41	0.72	26.3	196	153	0.56	22.0	4.72	0.77	0.68	10.7	0.43	2.12	2.24	2.54	141	150
0.32	1.04	27.1	197	141	0.60	20.9	5.58	0.88	0.79	11.2	0.44	3.47	1.88	3.01	128	212
(0.79)	0.98	28.9	205	155	0.62	22.4	5.64	0.85	0.76	11.8	0.47	2.74	1.70	3.05	146	178
0.43	1.80	27.1	200	140	0.63	19.7	5.34	0.77	0.80	10.2	0.36	2.88	2.08	2.74	103	157
0.42	1.81	28.1	195	143	0.91	20.1	5.37	0.79	0.75	10.6	0.91	2.58	2.33	2.62	100	230
0.02	0.01	1.5	10	2.7	0.06	0.04	0.03	0.03	0.04	0.1	0.03	0.09	0.17	0.06	2	27
3.9	0.6	5.3	5.1	1.9	9.3	0.2	0.6	3	5.8	0.6	6.9	3.6	7.9	2	2	16

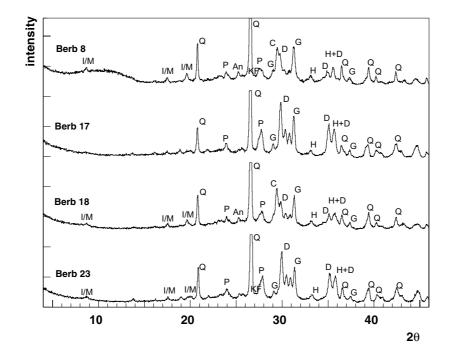


Figure 2 XRD spectra of four ceramic samples from Berbati (Argolid). The indicated mineral phases are illite/ muscovite (I/M), quartz (Q), plagioclase (P), analcime (An), K-feldspar (KF), gehlenite (G), pyroxene (D), calcite (C) and hematite (H).

present as a secondary phase (Maggeti 1981). In sample Berb 17, the illite/muscovite peak has disappeared, which indicates a higher firing temperature. According to the high-temperature XRD analysis of other MB samples at the University of Barcelona (Buxeda *et al.* 2002) the EFT for Berb 8, 18 and 23 can be estimated in the range of 850–950°C. In the case of Berb 17, the EFT is about 1000–1050°C.

In samples Voud 3, 4, 25, 30, 34 and 35 (Fig. 3), the mineral phases present are illite/ muscovite (I/M), quartz (Q), K-feldspars (KF), plagioclase (P), hematite (H) and pyroxenes (D). Apart from these phases, Voud 22 also presents calcite (C) and gehlenite (G), which is correlated to the higher Ca content in this sample (see Table 2). Considering the presence of hematite (H) and at the same time, in at least three samples, the presence of illite/muscovite (I/M), the EFT should be in the same range as in the examined MB samples.

The main difference between the diffractograms of the two groups is that the MB group comprises calcareous samples and the ACH-a group consists of samples with a low Ca content. The results of the chemical analysis indicated that the ACH-a group is characterized by a low Ca concentration (3.9%) compared to the respective concentration of the MB group (10.0%). The different evolution of the mineral phases during firing, which is due to the Ca content, is reflected in the XRD spectra of the two groups by the absence of gehlenite in the samples of the ACH-a group, except for Voud 22. Mineralogically, the two groups present differences in the intensity of the plagioclase peaks, which are more intense in the ACH-a group. This is correlated with the chemical results, which give a higher Na concentration for the samples from the ACH-a group. Another difference between the two groups is the presence of intense K-feldspar peaks in the ACH-a group, whereas they are only present in trace amounts in the MB group.

THE XRD RESULTS OF THE CLAY FROM KATAKOLO

The occurrence of the chemical pattern of the MB group—or, at least, of rather similar patterns in pottery from all over Greece and even from the Troas indicates the use of clays with very similar properties by ancient potters for the production of specific kinds of pottery. It can be assumed that, with regard to standard ware types and a constant quality of production, a specific type of clay deposit was preferably selected for exploitation (Jones 1984). Recent investigations of Neogene clay deposits in Crete (Hein *et al.* forthcoming) show that clay deposits that were formed during the same geological epoch—and thus, apparently, from the same sediments can present similar chemical and mineralogical compositions, even though they are not connected as far as today's topography is concerned.

Several earlier pottery studies have included the investigation of clay deposits in the Argolid and Corinthia (Jones 1986, 169–206; Newton *et al.* 1988; Bonn unpublished data). However, the analysis of the clay from Katakolo has indicated for the first time a deposit with a chemical clay composition similar to that of the Mycenaean pottery groups MB and ACH-a. The deposit at Katakolo was one of a number of apparent clay deposits that were sampled incidentally during a sampling campaign of Mycenaean pottery from the Peloponnese. Among the other clays from this series, including deposits in the vicinity of the relevant archaeological sites, no similarities with the respective ceramics could be found as far as their chemical compositions were concerned.

With regard to the tectonic development of the Peloponnese during the Pliocene, the Argolid was separated from the rest of the Peloponnese, and the Gulf of Corinth was connected to the Argolic Gulf. During the same period, the present-day west coast of the Peloponnese lay below sea level (Symeonidis and Anapliotis 1969; Jacobshagen *et al.* 1978). The sediments deposited

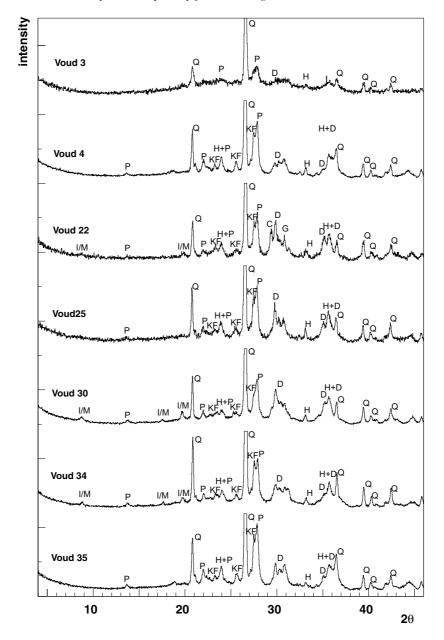


Figure 3 XRD spectra of seven ceramic samples from Voudeni (Achaia). The indicated mineral phases are illite/ muscovite (l/M), quartz (Q), plagioclase (P), K-feldspar (KF), gehlenite (G), pyroxene (D), calcite (C) and hematite (H). The poor counting statistics of sample Voudeni 3 are due to an insufficient amount of material.

during the same geological period in these areas might bear chemical and mineralogical similarities (Hein *et al.* forthcoming) that are reflected in the relevant clay deposits.

The XRD of the bulk sample presents quartz, calcite, plagioclase, K-feldspar, illite/muscovite and chlorite (Fig. 4). However, the most intense peak is at 2.82 Å ($2\theta = 31.7^{\circ}$), which corresponds

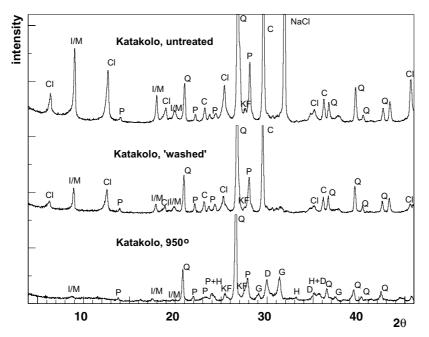


Figure 4 XRD spectra of the untreated bulk clay sample from Katakolo (Elis), the washed Katakolo clay sample and the washed sample fired at 950°C. The indicated mineral phases are chlorite (Cl), illite/muscovite (I/M), quartz (Q), plagioclase (P), K-feldspar (KF), calcite (C) and halite (NaCl). Furthermore, in the fired sample the formation of pyroxene (D) and gehlenite (G) can be observed.

to a high halite (NaCl) abundance, probably due to the close vicinity of the sea and/or the marine origin of the deposit. A second XRD spectrum, which was measured after 'washing' the sample (the clay was mixed repeatedly with water and the clay–water suspension was removed after settling), does not contain the 2.82 Å peak, as shown in Figure 4. Furthermore, the peaks of the clay minerals are less intense due to the removal of part of the small particle size material. The XRD of the clay fraction showed no changes in the spacings of the ethylene-glycolated sample. Hence there are apparently no smectite minerals present in the sample and the peak at 14 Å ($2\theta = 6.2^{\circ}$) corresponds to chlorite only. The apparent double peak at 3.57 Å/ 3.54 Å ($2\theta = 24.9^{\circ}/25.1^{\circ}$) indicates the presence of kaolinite in addition to the chlorite. This assumption is supported by the almost complete disappearance of the peak at 7 Å ($2\theta = 12.6^{\circ}$) in the heated sample, since kaolinite decomposes at a temperature below 550°C.

For a further examination of the mineralogical properties of the clay, a briquette was produced from the 'washed' clay sample and fired for 1 h at 950°C under oxidizing conditions. The XRD spectrum (Fig. 4) shows a development similar to that of the ceramic samples of the MB group, which is characteristic of highly calcareous samples.

CONCLUSIONS

The XRD measurements of the pottery samples from Achaia and the Argolid confirmed their separation into two regional groups. The presence of gehlenite in the MB fired pottery, which is missing in the ACH-a samples except for Voud 22, corresponds to the higher Ca concentration in the MB samples. The higher Ca content of the Voud 22 sample compared to the rest of

the ACH-a samples could be explained as a result of tempering or natural variation in the clay deposit. These assumptions cannot be verified, because the samples were only available in powder form.

As far as the Ca and the Na concentrations are concerned, they could be related to different environments of the sediment deposition. In a marine environment, a cation exchange emerges in river sediments between Ca^{2+} ions and Na^+ ions (Jasmund and Lagaly 1993, 173). Additionally, the differences between the Cs and Na concentrations are correlated with the higher amount of feldspars (Middelburg *et al.* 1988) in the ACH-a samples. The latter characteristic could also be the result of a different grade of chemical weathering of the parent rock or sediments, respectively. However, the regional distribution of the two groups suggests different deposits with a different state of weathering rather than a single deposit with a weathering profile. The raw materials used for pottery production probably came from deposits formed from very similar sediments. The differences in the chemical and mineralogical compositions are small, but they indicate different settling environments and weathering states of those deposits that were exploited for the production of pottery in Achaia and the Argolid.

The examined clay showed similarities in the chemical and, after firing, in the mineralogical composition as well. Although it is unlikely that the pottery workshops in the Argolid exploited a clay deposit at a distance of 120 km to the west, the Katakolo deposit might represent the preferred clay type. In order to confirm this assumption, a systematic programme of clay prospecting in Achaia and the Argolid is planned, focused on Pliocene clay deposits that are geologically related to the Katakolo deposit.

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