Acta Mineralogica-Petrographica, Szeged XXI/1, 41—47, 1973

MINERALOGICAL-PETROGRAPHICAL AND CHEMICAL INVESTIGATION ON ARCHAEOLOGICAL CERAMIC GRAVEGOODS

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As well known, an important part of the archaeological finds consists of utensils, vessels or building material made of fired clay. It can be easily understood that the claim to study this material, often very important from an archaeological point of view, with modern scientific methods, necessarily arose recently all over the world.

With the spreading of the scientific methods of approach, it was natural to realize the fact that the results of the modern testing may be a great help in archaeological identification of finds. It becomes clear, however, that in the investigations on archaeological material numerous particularities have to be taken into consideration, which cause the methods developed and generally used to give results to insufficient in some cases.

The difficulties in determination of ceramic products arrive partly from the processing, partly from the physical and chemical effects of being buried in the earth, and which may alter the original state, the natural composition and structure of the clay, in a not negligible measure. The sluicing of the clays used for pottery, their mixing with further mineral substances — lean material — leads to changes in the natural ratio of the mineral constituents; their forming and throwing (working on the potter's wheel) causes of the texture, finally the firing produces chemical and physical changes, considerably modify the mineral composition. As in the most cases it is impossible to distinguish the changes caused by human activity from those due to natural circumstances, in our investigations to be described, we used the antropogene effect, too, as characteristics and criteria for determining identities or differences.

Investigations for archaeological purposes were made on ceramics material from the beginning of our century and even earlier. As in those times chemistry was in the foreground among of natural sciences, it is easy to understand that, chemical methods were employed for determining ceramics. However, as in consequence of their structure, both the raw material of ceramics and, the end-products can be characterized, only by the chemical components, the determinations based solely on the results of chemical analysis cannot be satisfactory, except special cases.

It follows from the character of the ceramic material, that the scientific determination of both the raw material, and the end-products can be based, in the first line, on their mineral constituents. It can be understood, that in our days the methods — for mineralogical-petrographical determinations — chiefly thin slides — are often used for characterizing ceramic finds. Thus for studying ceramics made from potter's clay it was necessary to elaborate methods, in which spite of difficulties the results of chemical analyses and petrographical determinations assert themselves, and their simultaneous evaluation becomes possible.

Therefore it has been tried to draw inferencies, concerning the minerals constituting the raw materials, calculations based on the analytical data of the chemical components, in order to divide into groups the material of ceramic finds.

The calculation generally used in silicate-chemistry, do not give, however, satisfactory results in the case of potter's clays, due to the presence of different clay minerals pertaining to the montmorillonite and mica group, which are hardly to be characterized chemically. As the silicates (except some alkali-silicates) can be decomposed only by fusion, the result of chemical investigations will not be able to answer the questions concerning the original connections between the components, the composition of minerals. This logically leads to our earlier statement, that the pottery, row, and fired products alike, the chemical composition in itself cannot be characteristic.

TABLE 1

Chemical analyses (weight percent) of the ceramics Szekszárd—Palánk find

Sample	Group	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	Ing. loss.
39	A	75,1	9,8	7,1	1,5	0,8	1,5	0,5	3,2
40		73,8	9,9	7,0	1,9	0,6	1,5	0,5	4,5
28		46,1	13,9	8,9	11,2	3,0	2,9	0,4	13,2
29		48,4	15,5	9,2	11,5	3,5	2,9	0,6	8,0
30		58,8	18,0	9,5	3,8	3,0	2,4	0,3	4,0
31		54,6	16,6	9,5	7,2	4,9	2,4	0,4	4,1
34		46,1	13,0	9,7	9,8	4,2	2,8	0,3	13,8
38		53,8	17,2	10,3	2,6	3,6	2,9	0,4	8,8
41	B	48,4	15,8	8,9	9,9	4,4	3,1	0,5	8,6
44		47,0	13,6	7,8	12,6	2,8	3,1	0,4	12,5
45		54,7	16,7	8,7	7,2	3,5	3,7	0,5	4,7
47		50,6	15,8	9,1	10,1	3,3	3,6	0,3	6,8
50		51,4	15,1	12,0	7,0	3,5	4,0	0,4	6,4
51		53,3	15,9	9,3	8,6	4,0	3,9	0,5	4,0
52		45,9	14,6	8,2	10,2	4,0	3,7	0,2	13,0
54		49,6	14,8	10,4	6,6	2,0	2,9	0,3	11,8
55		44,9	13,5	7,7	13,1	3,1	3,9	0,4	12,9
37		65,8	12,6	8,5	2,0	1,6	1,3	0,6	7,2
46	C	73,3	14,7	5,9	1,1	0,9	0,9	0,4	2,6
49		66,6	11,9	8,9	2,3	1,6	2,0	1,0	5,1
53		69,0	15,3	6,7	1,6	0,9	0,8	0,3	5,1
27		67,1	16,0	10,1	1,4	1,7	1,4	0,4	1,6
33		38,4	8,2	5,4	21,6	2,4	1,6	0,4	21,5
35	D	56,2	13,7	8,9	5,0	4,2	2,0	0,3	9,4
36		51,2	11,9	10,6	7,9	4,2	3,8	0,4	9,6
42		54,5	13,2	9,2	9,2	3,5	3,1	0,5	6,5
43		59,4	15,0	9,7	6,3	2,8	2,4	0,4	3,5
32	B/D	51,9	13,5	8,7	10,6	3,6	3,1	0,5	7,5
48		52,4	14,4	9,0	5,8	4,7	2,4	0,2	10,6

As known, in the clays processes connected with the loss of weight occur during the firing. This loss of weight manifests in the loss an ignition, and therefore it is always an important and characteristic factor among the components, resulting from chemical analysis. The fired clay — pottery — will show further ignition losses only in the case, if the thermal modifications, connected with loss of weight, have not been terminated during the firing, or the material of the pottery suffered changes after the firing, which subsequently may produce such processes. We found that the refiring of the material of unglazed pottery, produced with primitive technology, and buried in the earth mostly results in significant loss of weight. In the fragments exposed during the excavations we often could observed the enrichment in organic and various inorganic material; in several cases also some transformation, rehydration of the clay minerals could be determined. As the value of the ignition losses of the pottery and end-product is significantly modified by all these processes, therefore the losses in weight of the ceramic material of excavation, caused by firing cannot be considered as characteristic, neither for the original pottery, nor for its raw material.

It follows from the above that ceramics can be compared with mutually on the base of their components if they do not show any loss on ignition, or of the losses found represent their full value. The first condition practically applies only to compact, non porous pottery, the second only on the state preceding firing. The ignition losses found in all intermediary states introduces uncertainties in the evaluation of the comparative studies. In the material investigated by us the ignition losses varied between 1,6 and 21,5 weight percent (Table 1).

Owing to the above, it is not possible to compare the ceramics, brought to light during the excavations, directly on the base of quantity of their chemical components. Therefore our calculations were made on the basis of the relative distribution of components can be taken into consideration, which may be characteristic both for the raw material processed and for the ceramic product. Generally three components are chosen; for comparing the relative quantity of the latter, the method generally used for the representation of three-component systems, proved to be very suitable.

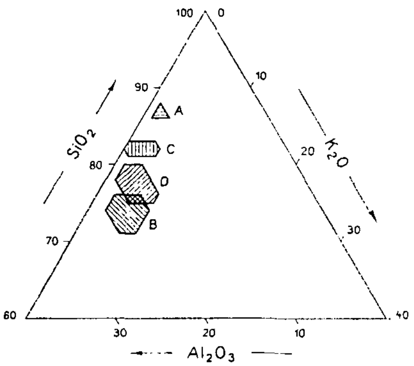


Fig. 1. Arrangement of the main sample-groups formed by the comparison of the chemical and petrographical data, in the SiO_2-Al_2O_3—K_2O system.

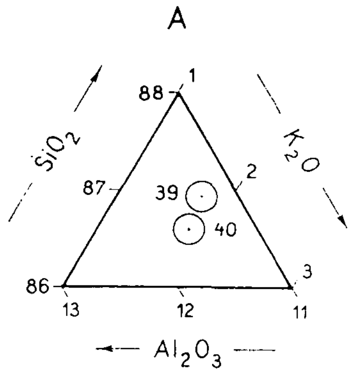


Fig. 2. The samples of the A-group plotted in the SiO_2—Al_2O_3—K_2O ternary system.

We found in many cases, that — similarly to example given in *Fig. 1* — the ceramics could be characterized by the percentual distribution of three components Al_2O_3—SiO_2—K_2O.

This choise cannot be considered as generally valid, because it is characteristic only for certain type of clay. It is, however, in good agreement with our finding, that in the Pannonian pottery's clays the dominant clay minerals are mostly hydromica and illite. In some cases we used the K_2O+Na_2O content; or the some of several components to calculate the relative value.

The mineralogical and petrographical studies on the ceramics were made with the petrographical thin slide methods generally used. Similarly as for rocks, this method permits to determine the mineral composition, the quantity of the constituents of the sample, as well as of their shape, dimensions, and mutual spatial arrangement (texture). As the our methods used in those investigations are of petrographic character: in our opinion the ceramics are to be considered as rock, despite of the antropogene effects mentioned above. Therefore in our descriptions we used the petrographical nomenclature in all cases.

The comparison and grouping of the data of macroscopic and microscopic examinations, based on similar features is not necessarily limited to statistical identification of the characteristics mentioned, as the raw material of the samples studied originates from fragmentation of earlier rocks. One of our further target may be the determination or reconstruction of the original locality of these rocks. In our work, depending on the number and character of available data, we tried to give an answer to these questions of genetic character, too.

In the following, we wish to exemplify the method described above, which is one of the possibilities evaluating the combined result of quantitive chemical analyses and mineralogical-petrographical studies by our investigations, on the material of the VI—VII Century Avar cemetery in Szekszárd-Palánk (SW-Hungary) excavated between 1958—1962.

The raw material of the samples, in its geological sense, was clay, sandy clay and calcareous sandy clay. The separation of the main groups was made on the basis of the different ratio of the contents in calcareous material, SiO_2, and clastic minerals.

I. Sandy clay: sedimentary rock formed in seawater. Detailed evidence concerning its geological age is not available. The mafrix consists mostly of recrystallized — partly of amorphous — SiO_2 network, containing sericite of laminar texture, altered biotite, more rarely of carbonate patches.

The material of the clastic minerals is: quartz (chiefly of magmatic origin) potash-feldspar, biotite, limonite, some plagioclase. In some samples haematite can be recognized.

Fossil debris of limy material are seldom. The samples No. 28, 32, 34, 47, 48, and 51 can be classed in the group.

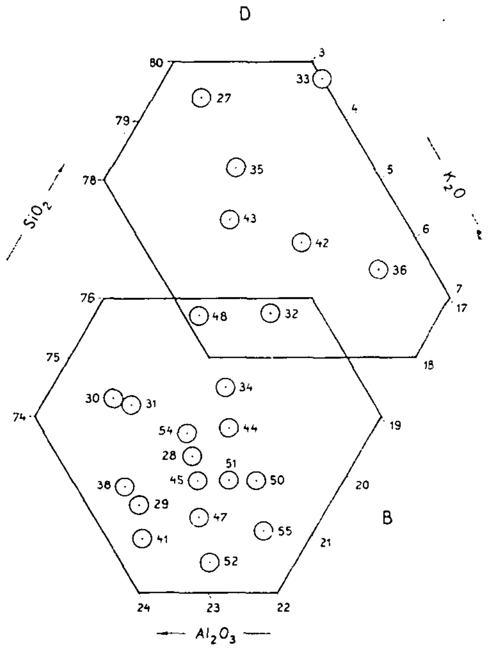
II. Silty clay, with patches of calcareous material. The material of this group originates from overwhelmed volcanic rocks, the mineral composition points to Miocene volcanic products. The samples can be divided into several subgroups, based on qualitative and quantitative differences in clastic minerals.

a) Much amorphous or recrystallized SiO_2, and much biotite as matrix with very little fine-sand. The latter consists of quartz, limonite, some plagioclase, and a few tiny zircon and apatite crystals. Samples No. 36, 41, 42, 45, and 50 belong to the subgroup.

b) In a matrix of SiO_2, biotite and little calcareous material, detritus of silt and sand sized. The SiO_2 in the matrix is amorphous or recrystallized: as a clastic component appears in the form of quartz. Greater quantities of muscovite and feldspar are to be mentioned, too. Heavy minerals: magnetite, limonite, garnet, apatite, sphene, some hypersthene crystal fragments. This subgroup includes the samples No. 29, 35, 38, 44, 52, 54 and 55.

c) More clayey, of lesser siliceous character than the precedent subgroup. The bulk of the clastic minerals consits of quartz, plagioclase, potash-feldspar, biotite, limonite, chlorite, and muscovite. Heavy minerals: zircon, rounded rutile and sphene. The samples No. 30, 31, 33, and 44 are to be reckoned to the subgroup.

III. This group consists of the most heterogeneous material: the clastic material originating from erosion of volcanic, plutonic and metamorphic rocks, enclosed in a clayey matrix. Oligocene age of formation is rendered probably by mineralogical, petrographical analogy. The subgroups are based chiefly on differences in the texture of the rock or caused by firing.



Fig, 3. The samples of the B and D-groups in the SiO_2—Al_2O_3—K_2O ternary system.

a) Siliceous-clayey matrix with much silt and fine grained sand. The material of the clastic components is quartz, orthoclase, muscovite. Heavy minerals: ironhydroxides, and a some epidote, apatite, hypersthene, rutile, sphene, tourmaline, zircon, magnetite, kyanite. Samples No. 46 and 47 can be ranged with this subgroup.

b) Red, clayley matrix coloured by ironhydroxide, much silt, with clastic material of fine and medium grained sand. Clastic minerals: quartz in the bulk (both magmatic and metamorphic), partly altered feldspar (potash-feldspar and acidic plagioclase), muscovite, chlorite. Heavy minerals: zircon, apatite, biotite, tourmaline, epidote, magnetite, hypersthene, kyanite and garnet. As rock fragments some quartzite, and limestone of fine crystalline texture containing magmatic quartz, occur sporadically. Samples No. 37, 39, 40, 49 belong to the subgroup.

c) Sandy clay. Only one sample, the coarser, the medium grained clastic material and the angular grain contours of which are different from all other samples. The most of the clastic minerals are quartz, the major part of which is of magmatic origin, a minor part being metamorphic. Heavy minerals :sphene, chlorite,hypersthene, apatite, zircon, rutile. As rock fragment a single grain of sandy marl can be recognized. This subgroup is represented by sample No. 53.

As can be seen from the above, it is possible to make comparatively very fine distinctions on the basis of mineralogical-petrographical macroscopical and microscopical investigations within the ceramic material of an archaeological excavation. Further refinements are possible with the aid of micromineralogical studies, and, in the first line by X-ray investigations for the qualitative determination of the clay-minerals and SiO_2 varieties. This possibility was already used by the authors in investigations on the material of an other find, result of which will be presented elsewhere. The authors come to the conclusion that, though further differentiations are possible compared with the results of study based only on thin slides and the conclusions can be supported by further data; but the identities of differences cannot be easily recognized by the archaeologist on the ceramics itself, these studies do not help immediately in approaching the purpose. It should be mentioned, further, that the petrographical features cannot be served as primary, determinent characteristics for the archaeologist, because other points of view, discussed in the introduction have also to be taken into consideration.

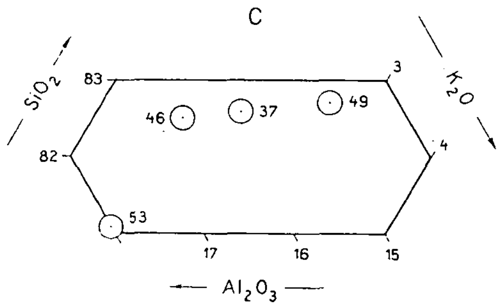


Fig. 4. The samples of the C-group in the SiO_2—Al_2O_3—K_2O ternary system.

The three-component representation based on chemical analysis by generalizing the petrographical features in a certain degree, proves to be very usefull in presenting or confirming the similar results of chemical and petrographical investigations *(Fig. 1, 2, 3, 4)*. This principle and method of grouping is proved to be very suitable in our practice for drawing conclusions, which are in accordance with the results of archaeologist. It is obvious that the number of the samples exerts a substantial influence on the value of scientific investigation. Reliable result based on few samples can be expected only in exceptionally cases.

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