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Clays of Georgia for Ceramic Applications

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ABSTRACT

The paper describes the rich traditions of Georgian ceramic manufacture and includes a detailed survey of the locations and compositions of the principal clay deposits in the country. The most important deposits are in the Imereti, Ajara and Guria regions. In this study, samples were taken of the clays on the south slope of a hill near the village of Darbazi in Southwest Georgia. Tests involved measurement of residue >45 µm, plasticity using the Pfefferkorn test, quantitative chemical analysis (XRF), mineralogical analysis (XRD), DTA/TG, and firing in a laboratory muffle kiln. We concluded that the Darbazi deposit has significant potential as a raw material for ceramic applications. Additional study is recommended for other deposits.

KEYWORDS

Georgia, clay, kaolin, ceramics, Darbazi deposit
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1 Introduction

Clay is one of the most common sedimentary rocks. It consists of one or more kaolinite and montmorillonite group minerals or other bedding silicates.

Kaolin makes up a major proportion of the clay that is extracted in the world. It is used in the wood pulp and paper industry and in manufacturing china, faience and refractory products. Georgia is no exception. Numerous ceramics-making hotspots have been known here since ancient days and each of them was directly connected with a clay deposit.

Fig. 1 • Schematic map of regions of Georgia with the main clay deposits for ceramic applications

Earthenware or its remains are found in practically all excavations of living sites of early man. Of particular interest among ancient earthenware products are black ceramics, known since about the third millennium B.C. The tradition of making this type of ceramics originated with the Etruscan culture (ancient Etruria) and the culture of ancient Georgia as well [1].

Throughout its history Georgia has had rich traditions in the use of clays for medicine and cosmetology, in manufacturing earthenware, china, faience crockery and other articles. Georgia is also an abundant source of different types of refractory raw materials, including alumosilicates, and magnesial and siliceous clays (Fig. 1).

Georgian clays often have impurities: feldspars, mica quartz, pyroxenes, magnetite, limonite and other accessory minerals. The colour of the clay depends on the presence of iron oxides and black carbon. Grayish-white, light-grey, dark-grey and yellow-red clays are mainly encountered in Georgia. The white and grayish-white variety is called kaolin. Deposits and evidence of such clays are usually connected with Jurassic cretaceous and tertiary rocks. Jurassic fire clays are mainly associated with continental sediments and are situated on the Georgian Block.

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2 Clay deposits of Western Georgia

The primary clay deposits are at the following locations:
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- Shrosha, Moliti, Jvari, Tkibuli and others (Imereti Region), and
- Tsetshklauri, Ochkhamuri, Jihanjuri, Makvaneti, Uchhubi, Didkviani, Shemokmedi, Tsatkini (Ajara and Guria Regions). Other sources (process products of trachytes, phonolites and others) are situated in tertiary deposits and are most commonly linked to crust weathering actions.

3 Clay deposits of Eastern Georgia

The main locations are the following areas: Darbazi, Kulazi, Pitareti (Kvemo-Kartli Region). The last two places are located to the East of points 6 and 7 of the map. Their origin is connected with hydrothermal alteration of Upper Cretaceous acid volcanogenic rocks.

The chemical compositions of the most important clay deposits described in the article are listed in Table 1.

The most interesting Georgian clay deposit is located near Dzirula (Imereti Region). It is known in the literature as the Shrosha deposit. The deposit is exploited as a raw material for refractory bricks. The region is rich in evidences of this kind of clay. Besides Shrosha, there are similar outcrops near Kandara, Macharula and in other places.

These clays are linked to two stratigraphic and lithologic formations of Lias-tuffites and sandstone. They occur in a lens-shape and have continental and epicontinental origin. Their industrial importance is typical of clays that occur in quartz sandstones.

Macharula clay contains a fairly high quantity of Fe_2O_3 (in some instances it may be as great as 8.3 %) and SO_3 (from 0.90–3.04 %). Mineralogical investigations show that SO_3 is likely to be associated with oxidation of pyrite. Their alumina content classifies these clays as basic raw materials. Stony varieties of the clay are richer in alumina than lumpy samples. The specific gravity of this clay varies from 2.50–2.67 kg/m^3.

The thickness of the clay layer fluctuates greatly and a rule quantifying the variation has not yet been found. The roofs and floors of the beds have wavy surfaces with rather deep hollows. The roof of a bed is commonly made of quartz sandstone about 4.5 m thick. Geological study shows that the layer is formed by washout and re-sedimentation of clay from an ancient crust of weathered Dzirula crystalline rock mass.

The clays of this deposit can be classified as low-plastic and meagre. Low-plastic clays have limited porosity and their water absorption decreases with higher temperature. Technological investigations of Macharula clay show that it has value as a raw material for fabrication of refractory articles. Current reserves of Macharula clay amount to about 1 million tons.

The Macharula deposit extends into the Kandara section. It is formed into a sheet-like deposit.

Other clay outcrops in the same Imereti region which are appropriate for further study are:

Table 1 • Chemical compositions of the most important clay deposits / mass-%

	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	Na_2O	K_2O	SO_3	P_2O_5	LOI
Kandara	64.22	**29.51**	2.15	0.12	0.51	0.58	2.00	1.03	1.75	–	1.80
Macharula	51.89	31.98	2.46	1.15	0.81	0.18	1.44	0.25	0.88	0.05	9.69
Gelati	50.04	36.61	1.00	0.04	0.26	0.34	–	–	1.87	–	12.65
Djvari	49.12	35.01	2.93	0.10	0.32	0.60	–	–	–	–	11.93
Chartali	53.20	28.42	0.49	0.90	0.18	0.36	1.34	5.69	0.1	–	4.28
Tsetkhlauri – 1	47.06	28.26	2.60	–	0.49	0.35	1.1		0	–	9.90
Tsetkhlauri – 2	47.58	26.98	1.42	–	0.55	1.99	2.44		trace	–	7.11
Tsetkhlauri – 3	44.60	31.60	1.80	–	0.31	0.22	3.28		trace	–	10.22
Djikhadjvari	65.45	22.32	1.74	0.90	0.44	0.96	0.36	3.25	2.46	–	4.10
Ochkhamuri	52.50	27.72	4.43	0.12	1.90	0.65	1.53	1.63	0.71	–	10.40
Makvaneti	53.3–62.2	23.3–32.3	0.49–1.79	0.38–0.50	0.42–0.60	0.80–1.05	1.50–2.59	1.41–2.59	0.7–0.77	0	6.13–10.15
Gogolauri	47.30	32.96	2.04	–	–	–	–	–	–	–	7.84
Eliatsminda	50.7–64.73	22.4–33.63	0.60–3.00	0.61–1.08	0.52–2.21	0.13–1.35	0.06–0.76	0.15–1.25	0.03–0.42	–	5.66–10.84
Cikarauli	60.1–68.55	20.0–27.54	0.75–2.26	0.74–1.37	0.39–0.55	0.12–0.29	0.14–0.37	0.26–0.72	–	–	6.59–9.79
Sormoni	52.5–69.21	20.9–30.81	1.08–5.27	–	0.61–2.04	0.23–1.65	0.29–1.08		0.65	–	7.59–10.88
Rioni	51.7–61.61	20.2–34.30	1.10–4.85	–	0.44–3.31	0.24–0.94	0.15–1.17		0.13–1.59	–	7.00–10.95
Zarati	55.3–61.76	14.2–30.18	2.5–7.40	–	0.85–2.13	0.16–0.57	0.05–2.05		0.22–1.88	–	7.74–9.20
Dambludi	64.64	23.26	10.30	0.61	0.63	0.43	1.10	3.88	0.14	–	–

Fig. 2 • Clay mining near Askana village, Black Sea, Western Georgia

- near the villages of Satseble, Khoriti, Basaleti, Kroli and Tsevi,
- shales near the villages of Ubiza, Tsiplovana, Sanakshire, Gelati, Jvary, Tkribuli and others,
- refractory clays in the Aklemic-Keli country near the village of Bazaleti.

The deposits and outcrops of Ajara clays are of special interest. Layers of Tsetskhlauri clay deposit are situated about 15 km from Kobuleti, the Black Sea health resort. The surrounding area is covered by thick alluvial and diluvial sediments of brown clays and Quaternary alluviums.

Clay outcrops are found throughout the area. The southern boundary of the deposit is the Vashati River canyon, and the northern border is the Choloki River. Tsetskhlauri clays may be divided into two categories: light-gray and dark-grey, which appear together in the majority of cases, as light-grey clays almost always lie under dark-grey clays. Sharp change of facies over small areas is a characteristic of the deposit. It should also be noted that there are different clays in the formation, and a separate layer of carbonized trees that sporadically overlaps the clays and colours them in various shades of grey. The variation in thickness of the deposit is considerable – from 0.3–7 m. Calculated reserves constitute 1.7 million tons. The melting temperature of clay through the different sections varies from 1460–1640 °C. Judging from available data, the shape of the clay formation is close to lenticular. The layer lies almost horizontal and is very convenient for exploitation (Fig 2).

Tsetskhlauri clays have the following mineralogical composition: kaolinite, quartz feldspar, pyroxene (augite), iron oxide, and magnetite. The clay is composed of flakes and plates; admixtures are fine-grained with the size of the grains varying from 0.08–0.1 mm. The chemical composition of Tsetskhlauri clay is shown in Table 1. The gray clay of the deposit is an extremely interesting example of clay with extraordinarily high binding capacity. Rich with well-expressed plastic properties, it is easy to shape, is fine-grained, and has high purity combined with sufficient fireproof properties.

This clay is a rare sample of an analogous type of clay, called "Bindetone" in German nomenclature. Similar clays are very valuable in ceramic manufacture. In semifactory conditions, high-quality pipes have been obtained from Tsetskhlauri clay [2].

Situated on the outskirts of Tsetskhlauri are the Dikhandgora and Ochkhamuri clay deposits. They are similar to the Tsetskhlauri deposit from a genetic point of view, but show different varieties. The basic average ceramic indices are:

- moisture: 35–37.6 mass-%
- water absorption at 950 °C: 22.6–22.7 mass-%
- bending strength after firing: 125–128 kg/cm^2.

A fired sample assumes colours from light to bright pink. Ochkhamuri clay is predominantly infusible and can serve as a raw material for ceramic production.

On the northwest and western outskirts of the Adjara-Imerety mountain ridge a number of deposits and outcrops of "kaolin" type clays are present. Their names are Makvaneti, Uchhubi, Gogolouri, Gogieti, Mtispireti and others.

Sediments from the Paleocene period occur in the geological structure of the Makvaneti deposit. They appear as thin-bedded dark-grey marly argillaceous sandstone under a set of Eocene era volcanogenic rocks. These rocks are represented lithologically by tuffs, tuff breccias, augite-andesites and trachyte and trachyte tuffs. Geological-petrographic study of the deposit of kaolin and country rocks has shown that Makvaneti kaolin was formed by decomposition and leaching of trachyte [3]. The clays of the deposit are of alluvial type. The shape of kaolin bedding is anomalous, due to the intensity of the kaolinization process that formed primary rock. The deposit is divided into two sections, which display the same geological conditions. Kaolin clay of the second section is characterized by considerable lithologic variation, colour of rock and contamination. Kaolin of the first section also shows considerable lithologic variation and contamination, which affects the colour of the rock. Kaolin clay of the second section is highly ochreous, but lenses of white kaolin are encountered there as well [3].

The clay is mechanically contaminated by admixtures, mainly of mother-rock, feldspars, quartz and other substances. Kaolin clays show coarse granulometric size and are low-plastic in character. Their tensional strength is about 1.35–4.90 kg/cm^2. Temperature of fusion is 1670–1710 °C. The clays are divided into white and coloured categories. The clays suitable for manufacturing china articles belong to the first group. The second group contains clays with elevated content of iron oxide and have properties suitable as raw material for ceramic manufacturing.

Reserves of fire clay in the Makvaneti deposit are only 350,000 m^3 (white: 250,000 m^3, coloured: 100,000 m^3), which is not enough for industrial exploitation.

Some evidences of clay in the Guria Region (Western Georgia) have been examined. The Shemokmedi section is situated on the left slope of the Bzhuzha River. The thickness (or more exactly the depth) of argillized rocks varies from 2.8–8.0 m. An outcrop of clay of width 30–40 m is traced on the side of the river for a distance of 700 m. The clays, in the main, are highly ochreized and only in some explored openings are they white in colour. When the content of painting oxides (Fe_2O_3 and TiO_2) is measured, almost all clays of Shemokmedi section belong to a group with high content of iron (more than 3 mass-%). Only a few samples of clay show medium painting oxide content.

The content of Al_2O_3+TiO_2 in all samples fluctuates from 15–40 %, which classifies them as basic and semi-acid raw material. Regarding refractory behaviour, these clays reach a melting point at temperatures not lower than 1610 °C.

Direct prolongation of the Shemokmedi section is evident in the Gomi area. The depth of affected rocks occurring here is as deep as 7 m. Argillized tuffs and trachytes are mainly observed. The productive formation is located in the dividing crest and ex-

tends for 500 m with a width of 30 m. Painting oxide content is high in nearly all samples (as in Shemokmedi).
These are refractory clays by nature. They are inherently friable and do not get wet when immersed in water.
The Goglauri deposit is situated in Guria [4–5] to the southwest of Uchkhubi village. A relief map of the countryside is highly indented by mountainous rivers and the left-side tributaries of the Natanebi River. By mineral content as well as ceramic properties, the clay is similar to that of Makvaneti [5]. The clay of the Goglauri deposit has high iron oxide content so its tone after firing is highly coloured. Because of this, Goglauri clay cannot be used in china manufacture.
In our opinion future investigations should apply critical attention to the deposits of kaolin clays of the Tkibuli and Tskaltubo districts of Georgia as well as the deposits of southeastern Georgia.
Kaolin clays of the Tskaltubo and Tkibuli districts exist in the same geologic conditions and are linked to the formation of foliated shales during the Bathonian period. A clay deposit is located to the north of the village of Orpiri Jvarisi and is divided into two sections – Eliatsminda and Tsikarauli.
The Eliatsminda section exhibits dark-grey clay, foliated shales and kaolinized tuffogenic rocks. This alternates with shales and a horizon of kaolin argillous rocks. Outcrops of kaolin clays are all situated at nearly the same hypsometric height.
The deposit has a sheet-like shape. The length of the sheet is about 1 km and its width varies from 200 to 400 m. Kaolin is dense, sometimes loose and earthy, and is white in colour. The clay usually shows jointing patterns. Microscopic study reveals that the rock is a clay mass of chiefly kaolinite and montmorillonite with dispersed granules of plagioclase and occasionally rutile and quartz. The chemical composition of individual samples of Kvarisi kaolin is greatly variable (Table 1). The composition of the clays and their painting oxide proportions appear to qualify them as good raw material even for china.
The lithologic structure of the Tsikarauli sector of the Kvarisi deposit is the same as Eliatsminda with only one difference – here thicker layers of clay shales occur on the bed of kaolin clay. The clay is white-grey and yellow, mostly loose and occasionally sandy. The mechanical composition of the clay is not homogeneous – outcrops of clayey mass make up 30–37 % of samples. The clay is refractory up to 1630–1670 °C. Samples fired at 1300 °C show white colour.

Fig. 3 • Southern slope of the hill near the village of Darbazi

The Sormoni, Rioni and Zarati deposits are located to the north of Kutaisi town. In the geological structure of the district, sediments of the Lower Bathonian period are found; they are represented by foliated shales, dark grey dense clays and clay shales. Among the clay slates a horizon of kaolinized feldspar sandstones occurs. Alternating beds of kaolin clays have thickness of 1 m each. The clay is mostly loose, low-plastic and can be scrambled easily if in a dry state. It is composed of the following minerals: kaolinite, montmorillonite, feldspar and (rarely) sheets of mica. Typical ceramic properties of these clays are:

- moisture: 20–37 mass-%
- loss of weight after firing: 9.1–16.4 mass-%
- shrinkage after firing: 2.34–11.14 %
- water absorption after firing: 6.8–20.7 mass-%
- bending strength after firing: 42.8–87.7 kg/cm^2.

The Dambludi deposit of stone-like clays is situated in the ravine of the Dambludi River in Southwest Georgia. The clays occur as sheets on old granite and alternate with quartz sandstones and conglomerates of Lower Lias. There are seven sheets of clay with thickness from 0.5–2 m.
The rock is light-grey in colour with rusty traces along the shell-like fracture surfaces. It is hard and does not fall to pieces in water. Under a microscope the texture of the clay is aleuritic. The rock has a finely sealed mass in which angular grains of quartz and feldspar are scattered. Clay masses are composed of kaolinite. The melting temperature of Dambludi clay varies from 1580–1650 °C. The clay is good for manufacturing decorative glazed tiles.
Besides those mentioned above, numerous smaller clay deposits are described in the literature. We think that all the deposits, large and small, must be purposefully studied by modern methods. They may have economic potential when modern technology is employed in up-to-date manufacturing.
Clay evidence must be studied deeply, including investigating composition and drilling for the purpose of estimating reserves [6]. For this purpose we have taken samples of the clays on the south slope of a hill near the village of Darbazi (in Southwest Georgia). The hill is composed of kaolinizated tuffs and trachytes.
The clays at this location are acid vulcanite products of the Late Cretaceous era. The clay is in contact with acid tuffs, trachytes and subvolcanic bodies of rhyolites subjected to high hydrothermal alteration.
Argillization appears in the vertical section of the hill. The altered zone has a length of 100 m (further on it is covered by alluviums and vegetation) with thickness as great as 30–40 m (Fig. 3). Outcrops of similar kaolinitic clays may be traced in other places and gorges in the vicinity of Darbazi, which shows that the common zone extends for a considerable distance.

4 Experimental

To have a preliminary idea of the potential for industrial ceramic applications of the Darbazi clay, we examined two samples representative of the deposit: Sample I (from the altered part), and Sample II (from the part containing harder material).
We conducted the following laboratory analyses of these samples in the R.M. Ricerche Minerarie Srl laboratory (Lozzolo, Italy):

- measure residue >45 µm (only on Sample I)
- Pfefferkorn Plasticity test (only on Sample I)
- quantitative chemical analysis (XRF) with loss on ignition at 1050 °C

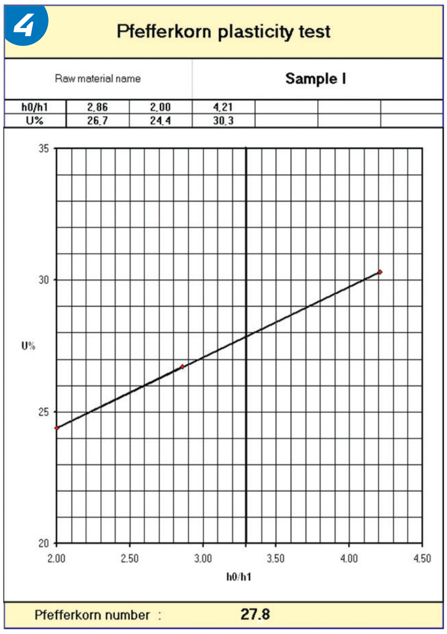

Fig. 4 • Pfefferkorn diagram resulting from plasticity test made on Sample I

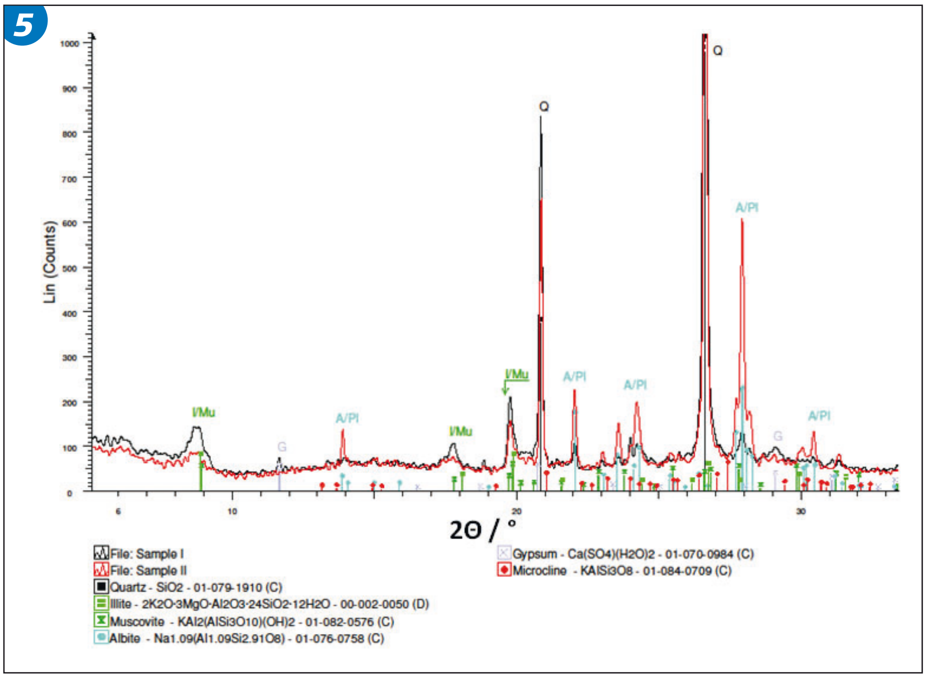

Fig. 5 • Comparison XRD diagram of Darbazi samples I and II

- mineralogical analysis (XRD)
- differential thermal analysis and thermogravimetric analysis (DTA/TG)
- firing test in a laboratory muffle kiln.

4.1 Residue >45 µm

200 g of Sample I were sieved with a 45 µm net after one night of rest in wet conditions with added sodium tripolyphosphate. The result was 81.1 mass-% >45 µm. This value indicated that fine material made up a low proportion of the altered clay sample.

4.2 Pfefferkorn Plasticity test

To verify the workability of Sample I we used a Pfefferkorn plasticity tester. The measured result (PI = 27.8 % is a medium-range Pfefferkorn index value that indicates good workability and plasticity (Fig. 4).

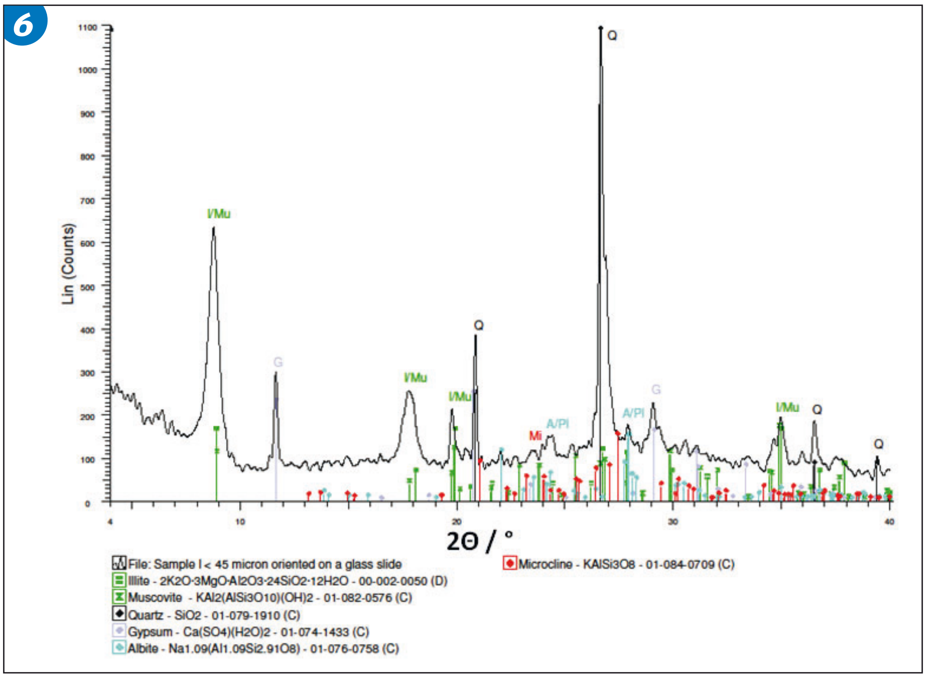

Fig. 6 • XRD diagram of Sample I <45 µm oriented on a glass slide

Table 2 • Results of the chemical analysis of Darbazi samples

Oxide	Sample I (altered part)	Sample II (hard part)
SiO_2	74.3	74
Al_2O_3	14.6	14.5
Fe_2O_3	0.6	0.64
TiO_2	0.43	0.48
CaO	0.45	0.25
MgO	0.74	0.57
Na_2O	0.61	3.09
K_2O	3.93	2.95
SO_3	0.57	0.3
LOI	3.53	2.82

4.3 Quantitative chemical analysis (XRF) with loss on ignition at 1050 °C

A chemical analysis was performed with an ED-XRF spectrometer Ametek Spectro iQ II. Tests were run on tablets made by mixing 10 g of micronized powder of each raw material and 2.5 g of Licowax C Micropowder PM used as a binder. The mixtures were shaped using specific pressure of about 2000 kg/cm². All the tested samples were analyzed under the same laboratory conditions and suitable standard procedures.

Loss on ignition was calculated after a firing test in a laboratory muffle kiln with a three hour cycle at 1050 °C and one hour at the maximum temperature. The results are listed in Table 2.

4.4 Mineralogical analysis (XRD)

The mineralogical analysis was made with a Bruker AXS D5000 X-ray Diffractometer (Siemens). A qualitative evaluation was

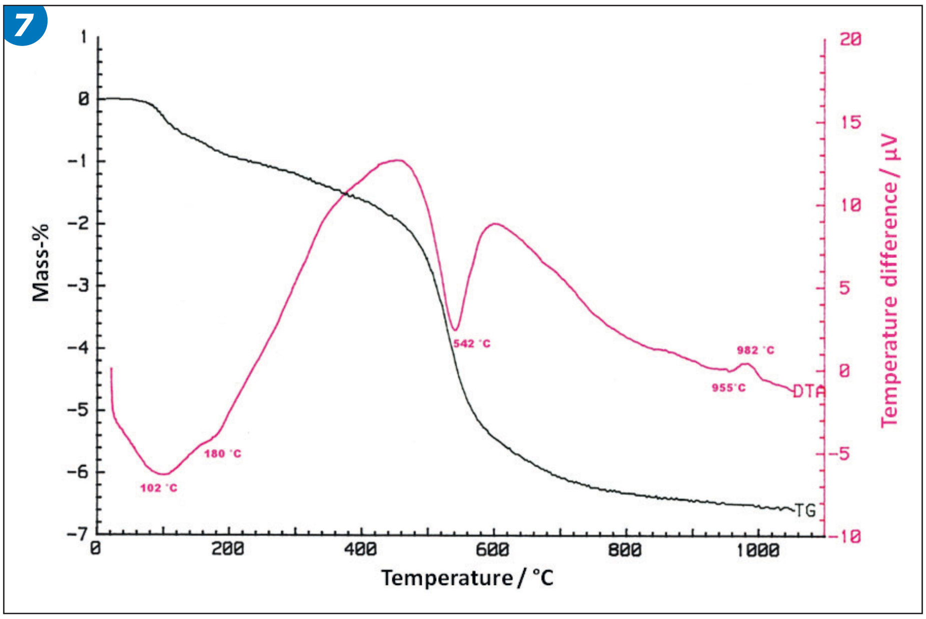

Fig. 7 • DTA/TG diagram of Sample I <45 µm

Sample I		Sample II	
Water absorption / mass-%	0.47	Water absorption / mass-%	0.15
Total shrinkage / %	4.44	Total shrinkage / %	in expansion
Colour	White / light grey	Colour	White / light grey

Fig. 8 • Aspect and physical characteristics of Darbazi Samples I and II after firing cycle in a laboratory muffle kiln. 5 h-Cycle: 1220 °C/15 min, (Buller ring 75, temperature = 1200 °C)

done using the PDF2 (Powder Diffraction File) database standards of the JCPDS-ICDD (Joint Committee on Powder Diffraction Standards-International Centre for Diffraction Data).

The XRD analysis showed that the samples mainly contained the following minerals: quartz, illite/muscovite, albite, and gypsum. Sample I differed from Sample II in showing Na plagioclase modified in the illite and a greater occurrence of gypsum. Kaolinite, usually present in these kinds of rocks is not relevant (Figs. 5–6).

4.5 Differential thermal analysis and thermogravimetric analysis (DTA/TG)

The DTA/TG analysis was performed with a Netzsch STA 409 EP simultaneous thermal analyzer. The only meaningful results were obtained from a Sample I <45 µm fraction (Fig. 7).

The DTA diagrams show a weak endothermic peak at about 100 °C that may be due to the presence of montmorillonite. The other weak endothermic peak at 180 °C and the main one at about 540 °C are typical of illite. A weak endothermic peak at 955 °C and an exothermic event at 982 °C explained by transformation of the same mineral at higher temperature.

The TG graph shows a first loss in weight at about 100 °C and the main drop occurs at 550 °C.

4.6 Laboratory muffle kiln firing test

The micronized Darbazi powder prepared from Samples I and II was also used to prepare round samples and was pressed at 400 kg/cm². The bodies were fired in a laboratory muffle kiln with a cycle of 5 h at 1220 °C/15 min.

The firing test showed good melting behaviour for the two samples after firing. Each displayed a very light colour (Fig. 8). Sample II was already in expansion after reaching its minimum water absorption value.

5 Conclusions

The preliminary analyses made on Sample I and Sample II from the Darbazi deposit surely underline their potential as a raw material interesting for ceramic applications.

Positive elements found in the laboratory tests are:

- the material is easy to crush and grind in laboratory mills
- good plasticity was measured by the Pfefferkorn method (PI = 27.8)
- the samples have low F_2O_3 and TiO_2 content
- the clay part shows presence of illite mineral and Na-feldspar
- good melting behaviour was observed at 1200 °C (<0.5 mass-% water absorption)
- the material has a light colour after firing.

Among negative elements are the following:

- the samples have low clay fraction and contain rock parts with different chemical and mineralogical compositions
- the tests demonstrated presence of SO_3 (>0.5 mass-% in Sample I)
- there were very low proportions of kaolinite in the raw material.

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