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FIRECLAY AND KAOLIN DEPOSITS IN ROMANIA

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ABSTRACT

The fireclays in Romania are in general high-graded as regards their refractoriness, and are the most frequently connected with the Lower Liassic formation and more rarely with Pliocene deposits.

The main kaolin deposits are originated through hydrothermal alteration of some Tertiary rocks and by transport and sedimentation under continental-lacustrine conditions of kaolinitic sands and/or clays, resulting from reworking of some old weathering crusts.

The exogenic residual deposits are less important, being represented only by relics of fossil weathering crusts.

The Romanian kaolins are very seldom utilizable as raw material for industrial purposes; generally they are utilizable after washing and/or chemical treatments.

INTRODUCTION

Fireclay and kaolin deposits of Romania are characterized by a large genetic and facial variability, are covering a long age interval and are widespread in about all the country.

Hydrothermal kaolins are especially connected with Tertiary effusive rocks of East Carpathians and Apuseni Mountains, relics of old kaolinitic weathering crusts are recognized in Baia Mare area or in South Carpathians, continental sedimentary deposits of weakly coherent sands and clays are located in Dobrogea, Transylvania and Banat, tightly coherent kaolinitic clays (refractory clays) are well developed in Apuseni Mountains and South Carpathians; the last ones are transformed into pyrophyllitic schists.

The shape of the kaolin-bearing deposits are quite various: beds, lenticular beds, lenses and pockets for sedimentary kaolin deposits, and irregular bodies or zones, veins, dykes and sills for hydrothermal kaolins.

The kaolins from the weathering zone, which are considered as the most important for exploitation, are frequently high-grade materials.

The main kaolin-bearing deposits mined in Romania are the hydrothermally kaolinized rocks from Harghita and Parva, the sedimentary kaolinitic sands and clays from Aghiereş, and Medgidia, and the fireclays from Suncuiuş and Schela-Viezuroiu. The Schela-Viezuroiu fireclays are wrongly called refractory clays; in fact these are pyrophyllitic schists with refractory qualities. An improper name is used for Harghita kaolin to (as commercial product), constituted mostly of hydromica.

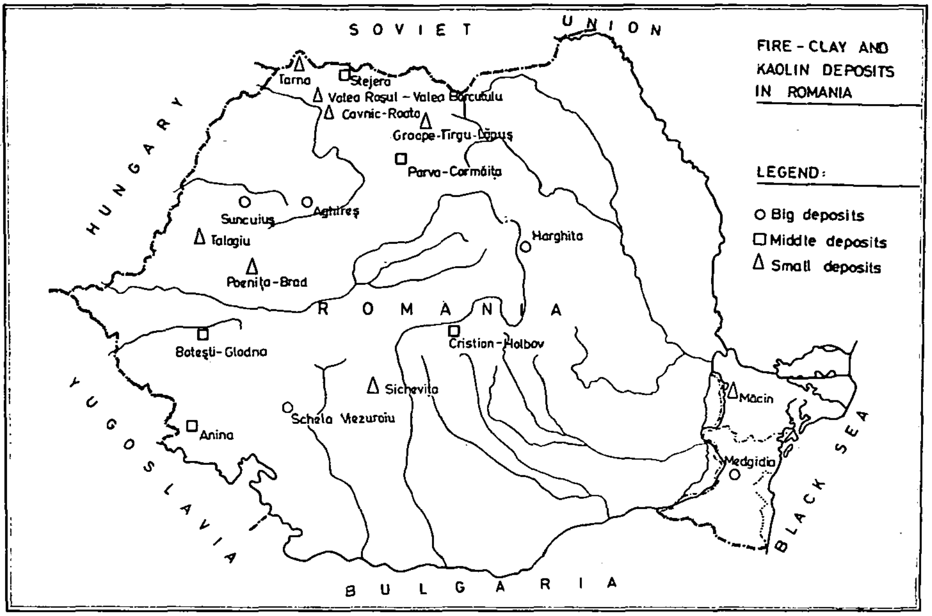

Fig. 1.

FIRECLAYS

Suncuiuş. The most important refractory clay deposits of Romania are located within Apuseni Mountains, Pădurea Craiului Massif.

The thickness of the Lower Liassic Gresten Formation with fireclay deposits varies between 40—100 m and it is represented by a complex of coarse sandstones and conglomerates with clayey, frequently refractory intercalations (bed-lenses). The productive horizon is formed by red clays encompassing intercalations and lenses of calcareous conglomerates and breccia in the basis, and sandy limestones, in the top of the horizon. Areas of repartition of refractory clays covers 120 km^2. A number of 15 fireclay levels are to be considered; their shapes are lenses and lenticular beds, ranging from 0.2 to 15 m in thickness, and extending laterally over hundred metres. The Suncuiuş fire-clay deposits are mined for a minimum thickness of 0.2 m and a refractoriness exceeding 169 PI.

Mineralogical composition of clay fraction is dominated by kaolinite T with subsidiary illite and quartz. Illite contents increase in nonrefractory clays, reaching to 20—40%, and kaolinite pM occurs usually within carbonaceous clays. Hydrargillite, montmorillonite, dickite, nacrite (along fissures in association with pyrites, sericite, chlorite and melanterite generated by pyrite oxidation), goethite, hematite and siderite have been incidentally recognized. Among heavy minerals there are recognized tourmaline, magnetite, ilmenite, zircon, rutile, staurolite and garnets.

The beneficiation tests carried out by washing of nonrefractory clays led to the result that 54% of the washed clay has a refractoriness of 171—173 PI; concomitantly the plasticity and compression strength increase to 22% and 50%, respectively.

Anina. The refractory clay deposits of Anina are associated with important pit coal deposits. These deposits (coal and fireclays) are mined underground up to 900 m depth.

The fireclay and pit coal bearing formation represents the Lower Liassic Gresten facies of the Semenic Unit; it is conformably overlain by a bituminous shale horizon and is overlying unconformably the red gritty deposits of the Lower Permian. The productive complex, so-called "coaly horizon", located in the southern limb of the Anina anticline is 250 m thick and consists of micaceous clayey sandstones, coaly shales, coals and fire-clays (2 beds). The tectonic framework of the area is characterized by important faults with clips up to 600 m which are dividing these clay and coal deposits into numerous blocks.

The mineralogical composition of the clay fraction is dominated by kaolinite; there also occur illite, quartz and small amounts of feldspars, iron oxides and hydroxides, carbonaceous matter. The kaolinite shows a crystallinity Hinckley index of 1.3—1.5 (T variety) for 70% of samples and 0.8—1.2 (pM and M varieties) for the remaining 30% of samples. Concomitantly with the increase of the tectonic pressure, pM and M varieties turn into kaolinite T.

Cristian and Holbav. In the vicinity of Cristian locality, two parallel synclines include Lower Liassic deposits containing refractory clays. The productive formation consists of sandstones, clayey breccia, clayey-coaly shales, coal and refractory clays; they are unconformable overlying the Anisian limestones and support the Carinian. In the Joedere Valley are 5 fireclay layers and in the Poiana Poieniţa 9, layers of 0.8—2 m thickness are encountered.

In the Holbav locality area, the fireclays are included in a coal-bearing clayey-gritty formation. There are 5 fireclay beds (up to 2 m thick) intercalated within the productive complex.

Mineralogical composition of Cristian and Holbav fireclays consist in kaolinite, illite, quartz, feldspars and sporadically pyrite, siderite, hematite.

Like at the Anina fireclays, the exploitation is carried out underground and presents difficulties due to tectonic causes.

Boteşti-Gladna. The refractory clay deposits of Boteşti-Gladna area consist in a few layers of 2—4 m thickness included in the Pannonian sandy-clayey sequence of the Caransebeş Basin.

Mineralogical composition of the Boteşti-Gladna fireclays consists in kaolinite (50—60%), illite (20—40%), montmorillonite (0—20%), quartz (5—30%), feldspars (0—5%), iron oxides and hydroxides. Refractoriness ranges from 155 to 171 PI.

Concerning their economic value these deposits present a reduced importance.

WEAKLY METAMORPHOSED FIRECLAYS

Schela-Viezuroiu. The geological framework of Schela-Viezuroiu area is represented by the Danubian Autochtonous, built up in this region by Suşiţa granite, Schela formation (sedimentary rocks with fireclay, weakly metamorphosed) and sedimentary rocks of Sarmatian age.

The fireclay-bearing complex is represented by Schela formation, which includes pyrophyllitic schists with intercalations of quartzitic sandstones and microconglomerates. The productive horizon, 140 m thick, comprises many intercalations of pyrophyllitic schists associated with bed lenses of anthracite. The thickness of pyrophyllitic schist intercalations ranges from 0.2 to 10 m.

The Lower Liassic deposits of kaolin type (fireclays) pertaining to the Gresten facies, grade into pyrophyllitic schists; concomitantly, the weak metamorphosis of coals of the thick intercalations leads to formation of anthracite and transforms the finaly disseminated coaly matter into graphite. The main part of the central (productive) complex consists of chloritoid-quartzose pyrophillitic schists, deriving from the weak metamorphism of some sandy kaolins. The transformation of kaolinite into pyrophyllite, the grading of illite into sericite is noticed. Along fissures or in geodes there was encountered an almost pure pyrophyllite (only 4% nacrite), sometimes associated with quartz, pyrite or chlorite.

The pyrophyllitic schists are composed mainly of pyrophyllite (80—90%), followed by chloritoid (1—19%), sericite (1—10%) and kaolinite (nacrite (1—7%). There also occur quartz (1—4%), graphite (1—4%), iron oxides and hydroxides, sporadically diaspore, chlorite, carbonates, rutile, tourmaline, zircon, pyrite. In the Baia de Arieş area the pyrophyllitic schists include larger amounts of chlorite-kaolinite (about 40%) and more sericite than Schela-Viezuroui schists.

The pyrophyllitic schosts present a refractoriness of 158—169 PI and are utilized in refractories.

The Schela formation is intensively folded and dislocated by faults showing numerous boundinage and lensing phenomena.

SEDIMENTARY KAOLINS

Aghireş. In the north-western part of Transylvania there are important kaolin sandy deposits of Oligocene age developed, exploited in the Aghireş area (Corneşti quarry).

The productive bed is underlain and overlain also by kaolin-bearing levels which, however, do not present any economical value. The kaolin-bearing sand bed considered to be mined show a variable thickness, ranging from 2 to 8 m (in average 4 m).

The coarse fraction consists of quartz (80—85%), micas (up to 10%) and sporadically chlorite. Heavy minerals (tourmaline, hyperstene, diopside, ilmenite, magnetite, garnets, rutile, staurolite, hematite, apatite, pyrite) may be noticed. Goethite is a common mineral present as impregnation in sands.

The fraction below 63 microns represents 14—16% of the raw material. Mineralogically it is constituted of a well-crystallized kaolinite T (75—85%), hydromica (10—15%), quartz (5—10%) and iron oxides and hydroxides, carbonaceous matter and calcite.

The Aghireş washed kaolin is utilized in fine ceramics and refractories industry. The commercial product presents a refractoriness of 169—171 PI-.

Medgidia. The Aptian continental deposits of South Dobrogea include essential accumulations of weakly coherent kaolinitic clays well developed especially within Medgidia area.

The clay deposits occur as lenticular beds with variable thickness (0.2—20 m), and occasionally with great lateral extent (hundreds of meters). In the kaolin-bearing complex there are intercalated cross-bedded sands and gravels which may become dominant in some areas. The clays, sands and gravels are white, yellowish, grey, red, red-violaceous due to the Fe and Mn oxides and hydroxides. Blackish and grey clays may also occur within some horizons rich in carbonaceous matter. The Aptian deposits are overlying the Jurassic and Barremian limestones or even directly the greenschist basement, and are transgressively overlain by the Albian, Cenomanian, Turonian or Sarmatian deposits.

The mineralogical composition of the clay fraction is characterized by kaolinite, subsidiary illite and montmorillonite, with incidentally chlorite. Among non-clay minerals quartz, feldspars, calcite, siderite and goethite can be recognized.

The kaolinitic clay from Megdidia can be used in paper industry, in foundry and ceramic industry. The marketable product for refractories must have a minimal refractoriness of 165 PI, and for paper industry a minimal whiteness of 65%

RESIDUAL KAOLINS

Stejera. Kaolin-bearing rocks are represented by kaolinized gneisses and kaolinitic arkosic sandstones forming an irregular layer 0.8—6 m thickners. These deposits show a residual character representing an pre-Paleogene weathering crust of lateritic type, preserved under the Eocene transgressive deposits.

Kaolinitic sandstones consist of quartz and angular fragments of micaceous quartzite encompassed in a kaolin groundmass.

The fraction below 63 microns represents about 10% of the raw kaolin-bearing rocks and is composed of kaolinite, illite, haloysite, incidentally montmorillonite and chlorite; beside clay minerals quartz, feldspars, micas, goethite, dolomite, calcite, rutile and pyrite are encountered.

Sichevița. Another relic of fossil weathering crust is known at Sichevița. These residual deposits occur as an irregular layer of 0.2—9 m thickness represented by a quartzitic sandstone; at the contact level of sandstone and granite there are encountered lenses of white kaolinitic sands.

HYDROTHERMAL KAOLINS

Harghita. The rocks of the area consist of vocanogenic sedimentary formations, pyroclastics and lava flows which are overlying the crystalline basement and the Mesozoic and Tertiary sedimentary deposits. The effusive rocks encountered in this region are pyroxene andesites, pyroxene amphibole andesites, amphibole andesites, biotite amphibole andesites and basalt andesites.

Argillization, silification and pyritization are the main types of hydrothermal alteration and are often connected with volcanic craters.

Hydrothermally altered zones are nonhomogeneous, rendered impure by limonitization of pyrite and are also including partially argillized blocks.

In the advanced phase of argillization the following stages can be distinguished: a first stage of formation kaolinite, a subsequent one of formation of hydromica and a final stage of formation of montmorillonite, chlorite, vermiculite and mixed-layer minerals.

The commercial product called „colloidal kaolin" from Harghita is composed of 10.5 Å hydromica (over 90%), kaolinite (7%), swelling clay minerals and quartz (under 3%).

10.5 Å hydromica shows an advanced degree of dispersion, a very good orientation and a high crystallinity.

"Harghita kaolin" has low refractoriness, high plasticity and presents thixotropy. Its absorption capacity is 55—60% of that of washed kaolin. „Harghita kaolin' is used in the paper and fine ceramic industry.

Parva-Cormăița. In the Rodna Mountains the mesozonal crystalline basement is overlain by Eocene and Oligocene sedimentary deposits. These rocks are pierced by 3 rhyolite dykes at Parva and a subvolcanic biotite dacite body at Cormăița. The eruptive rocks are more or less kaolinized.

In the Parva zone the rhyolites are hydrothermally altered by kaolinization, weak sericitization, pyritization and silification. The rocks are more strongly argillized where their fissuration is more intense.

Down to a depth of 40—50 m, weathering processes are superimposed on the hydrothermal alteration, leading to a more intense kaolinization, accompanied by montmorillonitization, calcitization and limonitization.

The mineralogical composition of the clay fraction is as follows: 55—76% kaolinite, 5—15% illite, 5—15% montmorillonite, 5—10% quartz, 0—5% feldspar and sporadically cristobalite and calcite.

The raw kaolin can be utilized in fine ceramics, due to its satisfactory contents of feldspar and quartz. The washed kaolinized rhyolite may be used in fine ceramic industry (pottery, electroporcelain), in paper industry, rubber, white cement, etc.

Concerning the kaolinized body of the Cormăiţa area, the following mineralogical composition of the clay fraction is to be noted: 35—50% kaolinite, 10—25% illite, 10—25% montmorillonite, 0—10% chlorite, 5—10% fedlspar, 5—7% cristobalite, 1% quartz.

The Cormăiţa kaolin can be utilized in ceramic industry.

OTHER HYDROTHERMALLY KAOLINIZED ZONES

At *Poieniţa-Brad* there are encountered kaolinized, silicified and pyritized andesites and tuffs with a reduced plasticity.

At *Cavnic-Roata* there is hydrothermal argillization of rhyolites and andesites. Argillization zones with kaolinite, illite, montmorillonite and weins formed of kaolinite, dickite and pyrophyllite occur.

At *Tarna* there are encountered zones of hydrothermal argillization of andesites and veins filled up with kaolinite and illite.

At *Groape-Tîrgu Lăpus* the crystalline formations are crossed by aplite and pegmatite veins which are intensely kaolinized in the neighbourhood of limestones under the action of hydrothermal solutions. Occasionally there occur kaolin pockets within limestones. The kaolinized material includes mostly well-crystallized kaolinite T (60—90%), montmorillonite (10—20%), illite (5—15%) and quartz (0—2%). The first argillization phase is the kaolinization, followed by montmorillonitization and chlorite-vermiculitization.

At *Măcin* the quartz porphyry and porphyrite veins are kaolinized owing to the circulation of hydrothermal solutions along the fault lines. The mineralogical composition of the fraction below 10 microns is as follows: 60—80% kaolinite, 10—15% illite, quartz and felspar, 10—30% montmorillonite.

At *Valea Roşie* and *Valea Borcutului* there are outcrops of intensely kaolinized, pyritized and silicified andesites, dacites and rhyodacites. The washed kaolin consists of 50—70% kaolinite, 15—50% illite, 5—15% quartz and feldspar. The degree of whiteness ranges from 40 to 70% and plasticity ranges between 47—51.

At *Talagiu* the andesites underwent some hydrothermal alteration processes such as argillization, garnetization, propylitization, sericitization, chloritization, carbonation, opalization, alunitization, and pyritization. Argillization, the most widely spread process, is represented by montmorillonitization (40%), pyrophyllization (30%), kaolinization (20%) and illitization (10%). Refractoriness of pyrophyllite-bearing rocks is 163 PI and the degree of whiteness ranges from 71 to 79.

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