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CLAY DEPOSITS OF LAS AGUILAS FORMATION, BARKER, BUENOS AIRES PROVINCE, ARGENTINA

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Abstract—Mineralogical, chemical, and geological aspects of four types of clay deposits in Las Aguilas Formation northwest of Barker in Buenos Aires Province, Argentina, indicate that these products of lateritic alteration of Precambrian granite are predominantly kaolinitic with minor amounts of halloysite, illite, and chlorite. Minor amounts of quartz, feldspars, and ferric oxides are also present. Of the four types, one is suitable for high quality refractories, another can be used for wall tiles and china, and a third, the most abundant, can be used for red ceramics; the fourth has no special use. The abundance of the clays is calculated to exceed eleven million metric tons.

Key Words—Argentina, Kaolinite, Lateritic alteration, Refractories, Reserves.

INTRODUCTION

This paper summarizes mineralogical studies of the clay deposits of the "Las Aguilas" Formation near Barker, Province of Buenos Aires, Argentina (Zalba, 1978b), and introduces additional information on the geology and stratigraphy of the region. The area of study extends along the Cuchilla de Las Aguilas, in the Sierras Septentrionales (Figure 1), 3 km northwest of Barker, Juarez. It is 64 km² in area and is delineated by the Gauss-Kruger coordinates 5836–5840 (abscissa) and 5548–5552 (ordinate) of the Hoja Topografica no. 3760-29-3 (Barker) map, Instituto Geografico Militar. Detailed geological mapping was carried out, and the clays were systematically sampled. Mineralogical, chemical, and physical properties were measured to determine their industrial application for refractories, ceramics, and cements in the Buenos Aires region. Reserves were calculated for different clay types.

STRATIGRAPHY AND LITHOLOGY

The deposits were first mentioned by Heusser and Claraz (1863) and have been subsequently studied by several investigators (Iñiguez, 1965; Caillère and Iñiguez, 1967; Di Paola and Marchese, 1974; Iñiguez and Zalba, 1974; Zalba, 1978a,b). In this region the "Las Aguilas" Formation occurs as a south-southeast dipping succession of clastic strata, for the most part sandstones with a minor amount of shales and conglomerates. Stratigraphic sections are shown in Figure 2.

Although the whole sequence was formerly considered to be a single formation (Teruggi, 1951, 1964; Teruggi *et al.*, 1962, 1968, 1973; Teruggi and Kilmurray, 1975; Di Paola and Marchese, 1974; Holmberg, 1972), in this paper the beds in the area under study are divided in two formations: La Tinta Formation and Las Aguilas Formation, on the basis of biological, structural, stratigraphic, mineralogical, and geochronological evidence of Amos *et al.* (1972), Dalla Salda and Iñiguez (1978), Zalba (1978b), and Bonhomme and Cingolani (1978).

La Tinta Formation is represented in this area solely by orthoquartzites, whereas Las Aguilas Formation consists of basal breccia, sandstones, shales, and conglomerates. La Tinta Formation is separated from the crystalline basement rocks by an erosional unconformity. It is overlain by Las Aguilas Formation which is apparently paraconcordant with it. Modern deposits (Tertiary?–Quaternary), described as loess and siltstones by Rabassa (1973), overlie all of the beds (Table 1). Clay deposits correspond to the shales of Las Aguilas Formation and consist of white siltstones and whitish, iron-bearing claystones.

The clays are distributed in two horizons which the author has named Lower and Upper Clay Beds. The Lower Clay Bed is white (N 9, Rock Color Chart, 1963) and is well laminated. It overlies a lenticular sedimentary breccia of clastic chert in a sandy matrix. Above are grayish pink (5 R 8/2), medium-grained orthoquartzites, with cross-bedding and ripple marks of large wave length ($\lambda = 9$ m; $h = 1$ m). Its thickness is about 0.7–0.8 m. An upper shale bed, with a silicified basal section, lies above the orthoquartzites and is covered by an alternating succession of shales and sandstones. It is possible to distinguish three layers of different colors with a total thickness of 3–6 m. The basal section consists of very dark red, iron-bearing clays (5 R 2/6) that gradually change to a middle section of moderate reddish brown (10 R 4/6) clays. The upper section is a pale, greenish yellow (10 4 8/2) clay. All three clay sections are well laminated and some show deformational as well as flaser structures.

The age of La Tinta Formation is probably Precambrian based on studies of algal stromatolites by Amos *et al.* (1972), similar to those from the Bambui Group

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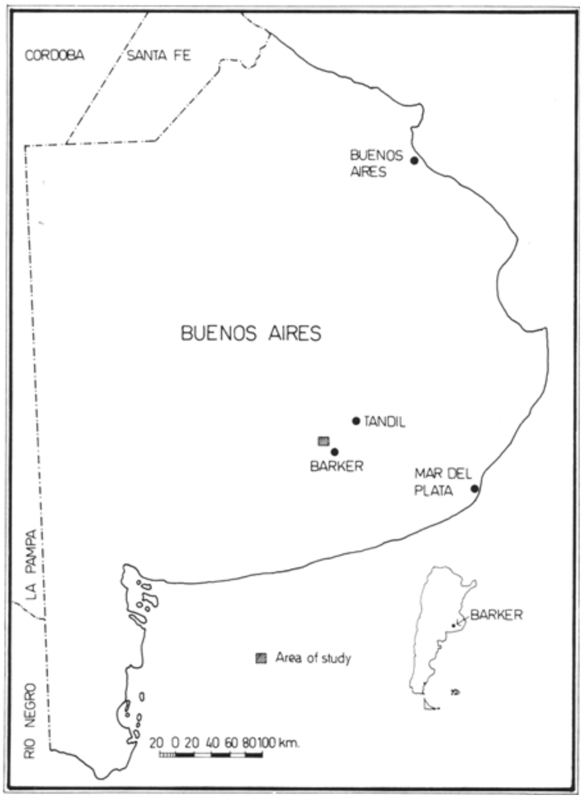

Figure 1. Map showing location of the area of study near Barker, Buenos Aires Province, Argentina.

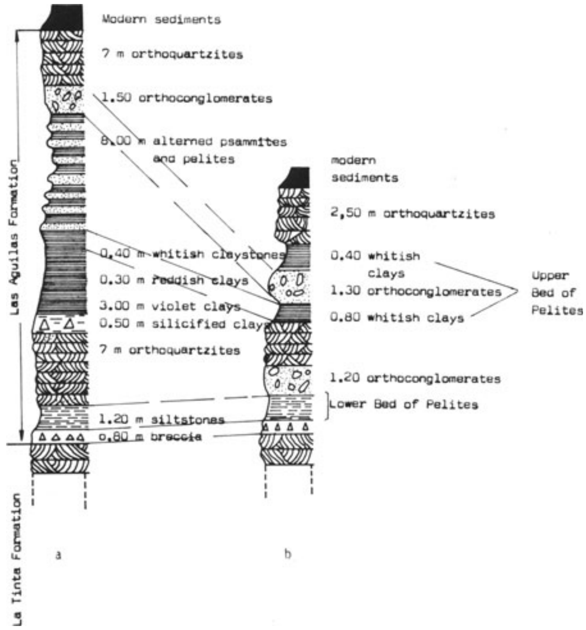

Figure 2. Type section of Las Aguilas Formation (a) near Cerro El Sombrerito and (b) in the northeasternmost exposures near Cerro El Sombrerito.

(Almeida, 1944) and which were dated by Amaral and Kawashita (1967) as between 600 and 640 million years. Recent Rb/Sr and K/Ar radiometric data obtained from clays in this formation in Sierras Bayas, Olavarria, by Bonhomme and Cingolani (1978) confirm this age. On the other hand, Las Aguilas Formation was considered Cambrian–Ordovician in age by Borello (1966) from fossil evidence.

EXPERIMENTAL

Methods

Clays in Las Aguilas Formation were characterized by mechanical, X-ray powder diffraction, chemical, differential thermal, dilatometric, thin section, and electron microscopy analyses. Most samples are strongly cemented with Fe_2O_3, and their dispersion is sometimes very difficult. Mechanical size analyses may therefore yield lower percentages of the clay fraction than is actually the case. The clays were classified on the basis of González Bonorino and Teruggi's (1952) terminology and correspond to silt clays and clay silts.

Clay characterization

Clay mineral identifications were based upon X-ray powder diffraction patterns of oriented and random aggregates of <4-μm size fractions, and their response to glycolation and heat treatment. X-ray analyses were performed with a Philips X-ray Diffractometer at 40 kV and 18 ma, with Ni-filtered CuKα radiation.

Most of the sediment samples contain several different clay minerals, the relative proportions of which vary from sample to sample. In addition, certain species appear to vary because of their hydration state and degree of crystallinity. *Kaolinite* was identified by its strong characteristic diffraction maximum which is not affected by glycolation. The peak at 10 Å is superimposed with the (001) reflection of hydrated halloysite (endellite) which hampers identification.

The presence of *halloysite* was confirmed by electron microscopy, but one of the factors that allowed its identification was the different behavior of the reflection at 10 Å (001) of illite and hydrated halloysite (endellite) which are superimposed. Both minerals are present in several samples, but when samples were calcined for 2 hr at 550°C, the 10-Å reflection became smaller indicating that the major component must be endellite. *Pyrophyllite* was identified on the basis of sharp and well defined basal reflection (001) from 9.05 to 9.40 Å. *Chlorite* is less abundant than other mineral species and was identified only in a few samples. On the basis of chemical analyses and the characteristics of the diffractograms, the chlorite appears to be a ferrous type, near thuringite (Zalba, 1978b), according to Hey's (1954) classification. The quantitative method of Johns *et al.* (1954) was used for semiquantitative estimates (Table 2). Areas under the peaks were measured with a polar

Table 1. Stratigraphy of the "Las Aguilas" Formation.

Lithostratigraphic units			Age
		Silty loess without structures Siltstones with $CaCO_3$ concretions	Tertiary? Quaternary
	Discordance		
Las Aguilas Formation		Orthoquartzites Conglomerates Claystones Orthoquartzites Conglomerates Siltstones Breccia	Cambrian–Ordovician?
	Paraconcordance		
La Tinta Formation		Upper orthoquartzites Light brown friable orthoquartzites Lower orthoquartzites	Precambrian
	Discordance		
		Crystalline basement rocks (granites and related migmatites)	Precambrian

planimeter, and the estimates were checked by means of chemical analyses (Table 3).

RESULTS

Areal and vertical distribution

According to the X-ray powder diffraction analyses of the clay fraction of the shales, it is possible to draw the following conclusions:

1. The Lower Clay Bed is composed of illite, pyrophyllite, and a small amount of kaolinite. Abundant quartz and rare feldspar occur as impurities. No lateral or vertical variation was observed, and the predominant clay is illite (30–60%). Pyrophyllite comprises 20–40% of the clay, and kaolinite, 6–10%. Up to 40% quartz is present.

2. The mineralogy of the Upper Clay Bed varies markedly from place to place. In the northeast, two whitish clay layers are present composed of kaolinite, halloysite, and pyrophyllite, with minor illite (Figure 4). Impurities present are abundant quartz and rare feldspar. Illite is the dominant clay in the upper part of the deposit. It rapidly diminishes in the lower sections where there is an increase in kaolinite, halloysite, and pyrophyllite.

In the central and northwest parts of the deposit, the two white claystone layers are replaced by three different ones with colors ranging from violet to red to white. Illite is dominant in the uppermost layer, but is less abundant or absent in places in the iron-bearing beds below. The percentage of kaolinite appears to be inversely related to the percentage of illite. Illite predominates in the iron-bearing clays and is less abundant in the upper whitish clays. Pyrophyllite is a ubiquitous constituent of the Upper Clay Bed. Its distribution is variable, but it generally predominates in reddish clays over the other clay minerals.

3. The vertical distribution of clay minerals depends principally upon segregation factors caused by preferential settling due to different average particle size.

Table 2. Quantitative estimation of clays from Las Aguilas Formation, Argentina by X-ray powder diffraction.

Sample no.	Kaolinite	Pyrophyllite	Illite	Chlorite	Quartz	Feldspars
6	42	33	25	—	scarce	scarce
12	25	15	50	—	scarce	scarce
25	60	20	20	—	scarce	scarce
35 'B'	48	20	25	7	scarce	scarce
36	45	45	10	—	scarce	scarce

Chemical analyses

Selected chemical analyses are listed in Table 3. Specimens rich in Al_2O_3 (34–40%) correspond to kaolinitic and halloysitic clays, which are in places associated with pyrophyllite and diaspore. Fe_2O_3 percent varies considerably. Higher values (up to 58%) correspond to reddish, iron-bearing clays. K_2O varies from 0.17 to 7.22% and is related to the illite content of the clays. Chemical analyses are in agreement with semiquantitative estimates by X-ray powder diffraction.

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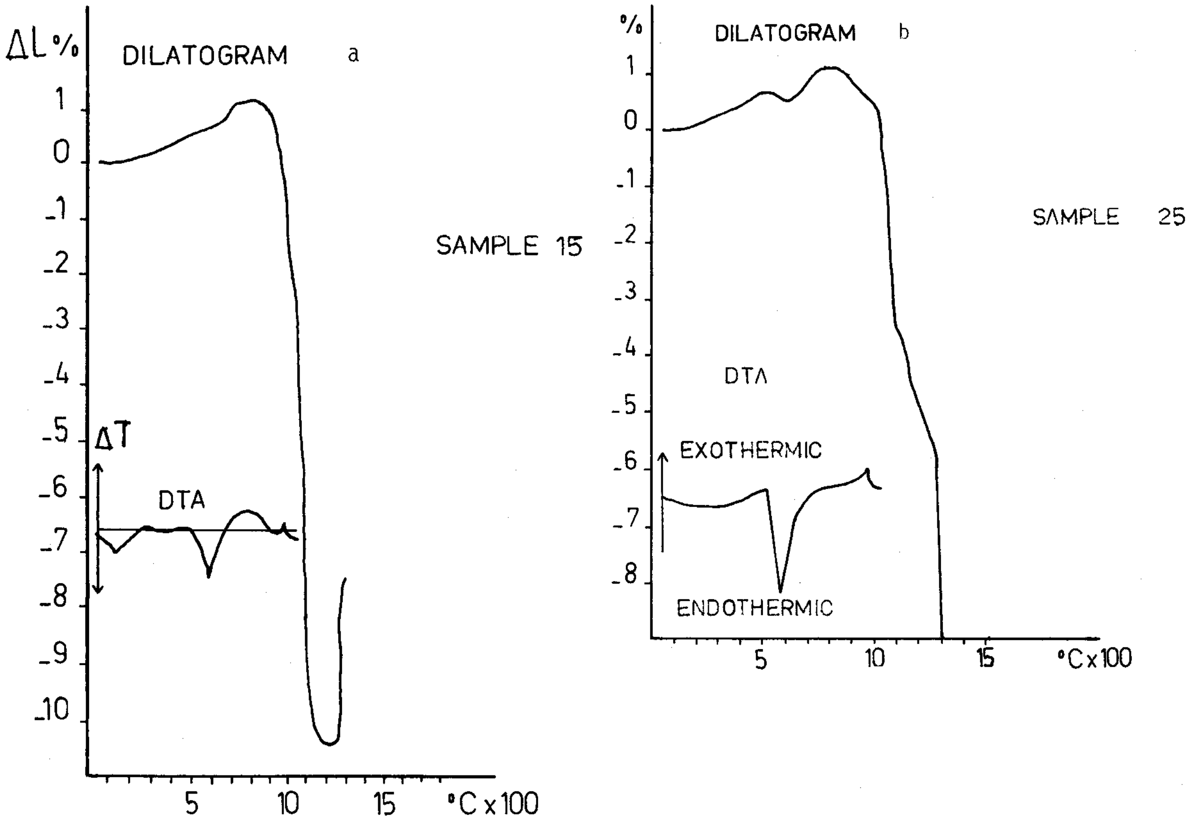

Figure 3. Differential thermal analysis diagrams and dilatograms of clays from Las Aguilas Formation (a) sample 15, (b) sample 25.

Differential thermal analyses and dilatometry

Differential thermal analyses (DTA) of more than 40 samples support the conclusions from X-ray examinations and indicate the degree of structural order in many of the clays. DTA and dilatometry patterns of two typical samples are shown in Figure 3. The first endothermic peak of sample 15 between 150°C and 200°C is characteristic of plastic clays. The second endothermic peak at 550°C is characteristic of fireclays with structural disorder. Sample 25 displays a second endothermic peak at 585–590°C, indicating that the mineral is a well crystallized kaolinite. Almost all of the kaolinite group members analyzed here are of the fireclay type and occupy an intermediate position between well-crystallized kaolinite and poorly crystallized halloysite.

Clays containing illite and pyrophyllite expand when heated between 600° and 900°C. Samples with more than 30% illite expand at higher temperatures (>1200°C). Clays with equal proportions of pyrophyllite and illite (e.g., 25% each) and with 40–60% kaolinite expand above 1200°C (sample 25). This natural mixture seems to be ideal for high quality refractories, provided that the Fe_2O_3 percentage is not greater than 1%.

Electron microscopy

Kaolinite. Electron micrographs of kaolinite clays from Las Aguilas Formation show well-formed six-sid-

Table 3. Chemical analyses of clays from Las Aguilas Formation, Argentina.

Sample no.	Color	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	MnO	Na_2O	K_2O	H_2O^-	H_2O^+
6	whitish	48.60	34.80	2.00	0.50	0.03	0.16	—	0.64	3.40	1.05	8.10
12	whitish	46	34.50	2.00	1.50	0.20	0.60	—	0.76	7.22	1.08	6.30
25	whitish	45.9	39.20	0.80	1.70	—	0.30	—	0.23	2.76	0.75	8.96
35 'B'	red	35.70	38.10	11.20	0.42	0.05	—	0.97	0.39	1.95	1.54	11.10
36	violet	18.10	12.10	58.00	0.98	0.05	0.04	0.71	0.40	0.89	1.73	8.76

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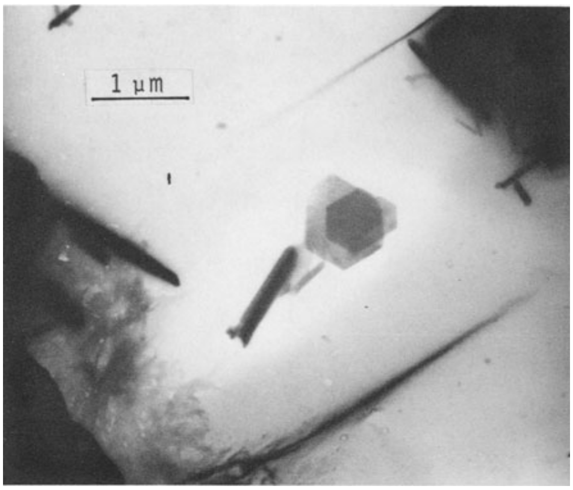

Figure 4. Electron micrograph of six-sided kaolinite crystals and elongate halloysite from Las Aguilas Formation.

ed flakes, with a prominent elongation in one direction parallel to either (010) or (110). As shown in Figure 4, the flakes are usually so transparent that the underlying crystals can be seen easily. It seems also that kaolinite is growing from a groundmass (Figure 6).

Illite. Because of its lack of distinctive features, illite from these deposits is one of the most difficult clay minerals to identify by electron microscopy. It is possible that the flakes are crystal aggregates.

Pyrophyllite. Pyrophyllite crystals have straight edges and are much larger than crystals of kaolinite (Figure 5).

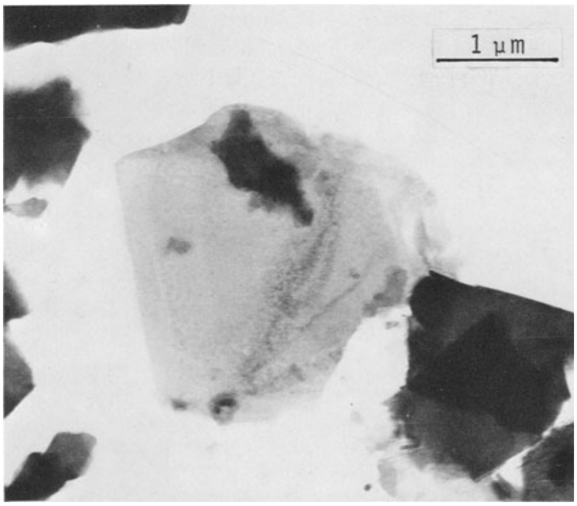

Figure 5. Electron micrograph of authigenic pyrophyllite with straight edges and shadows presumably due to diffraction effects from Las Aguilas Formation.

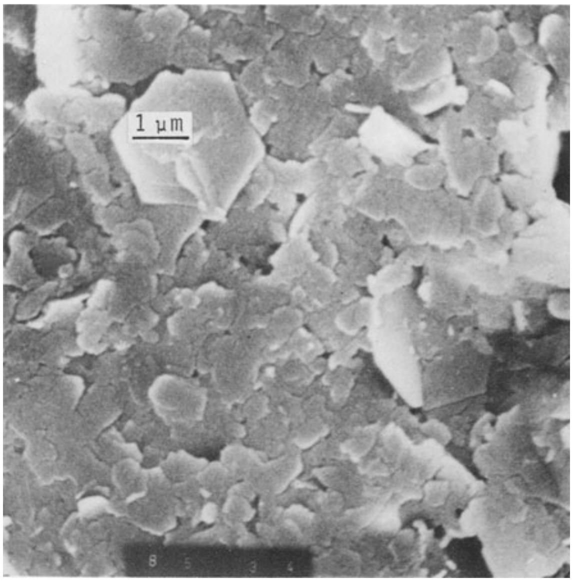

Figure 6. Electron micrograph of kaolinite growing from a groundmass from Las Aguilas Formation.

Halloysite. Halloysite occurs as elongate, tubular crystals which, in some samples, predominate over kaolinite. (Figure 7).

Summary of mineralogy

The several different analyses indicate that the clays of Las Aguilas Formation are essentially kaolinitic, with the following mineral association:

Lower Clay Bed: illite–pyrophyllite–kaolinite

Upper Clay Bed:
1. kaolinite–pyrophyllite–illite
2. kaolinite–illite
3. kaolinite–halloysite–pyrophyllite–illite

Genesis

In order to explain the origin of the clay minerals of the "Las Aguilas" Formation, Caillère and Iñiguez (1967) pointed out that these materials follow a zone of lateritic alteration formed on the granitic rocks. Iñiguez and Zalba (1974) confirmed the detrital origin of these deposits (granites and related migmatite rocks). Authigenic processes have also been involved in the formation of kaolinite, halloysite, pyrophyllite, and diaspore in shales and kaolinite in sandstones (Zalba, 1978a).

CHEMICAL-PHYSICAL BEHAVIOR CLASSIFICATION OF THE CLAYS

On the basis of engineering properties, four types of clay have been identified.

Type A: for refractories

White clays containing 43–55% SiO_2, 33–38% Al_2O_3, and 0.4–1% Fe_2O_3 may be valuable as refractory raw

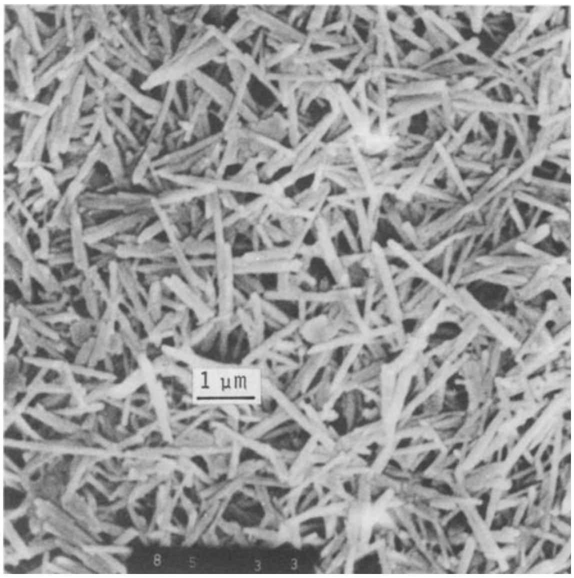

Figure 7. Electron micrograph of halloysite from Las Aguilas Formation.

materials. Their pyrometric cone equivalents (PCE) are between 31 and 32.5. The clay minerals present are kaolinite and/or halloysite, with minor proportions of pyrophyllite and illite. These clays seem to have an ideal proportion of kaolinite and illite for high quality refractories.

Type B: for china and wall tiles

Plastic white and greenish clays, containing 45–46% SiO_2, 34.5–36% Al_2O_3, and 0.9–2.0% Fe_2O_3 may be useful in the production of china and wall tile. Their PCEs are between 23 and 26. The clay minerals are kaolinite, illite, and pyrophyllite. This type of clay shows expansion between 1250° and 1255°C.

Type C: for red ceramics

Iron-bearing clays containing 47% SiO_2, 31–33% Al_2O_3, and 8–18% Fe_2O_3 may have application in the preparation of red ceramic bodies. PCEs are between 21 and 29. The minerals are kaolinite, pyrophyllite, and illite. Use in the refractory industry is limited by the Fe_2O_3 percentage; however, this is the most abundant clay type in the area.

Type D: limited use

Reddish and whitish clays showing isolated spots of Fe_2O_3 after heat treatment and characterized by 55–60% SiO_2, 23–28% Al_2O_3, variable Fe_2O_3, and PCEs between 21 and 23 are of limited technological use. The clay minerals present are pyrophyllite, kaolinite, and illite.

MINERAL RESERVES

Buenos Aires Province contributes 60% of the total Argentina mineral production (Angelelli *et al.*, 1973). This total is composed solely of construction materials such as orthoquartzites, dolomites, limestones, granites, and clays. Approximately 17,000 metric tons of clay per month are produced from the "Las Aguilas" Formation and are utilized primarily in the refractory, ceramic, and cement industries. Mineral reserves calculated in the present study are based upon outcrops, taking into account the lenticular nature of beds. All values correspond to estimated reserves separated according to different applications of ferruginous and refractory clays. There are 1,476,300 metric tons of whitish clays (types A and B) for refractory, china, and wall-tile applications, and 9,850,000 metric tons of iron-bearing clays (types C and D). The total amount of the clays available in this area for use as raw materials is therefore about 11,250,000 metric tons.

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Резюме—Минералогические, химические, и геологические аспекты четырех видов отложений глин формаций Лас Агулас к северо-западу от Баркер в провинций Буэнос Айрес, Аргентина, указывают, что эти продукты латеритового выветривания докембрийского гранита являются в основном каолинитовыми с небольшими количествами галлоизита, иллита, и хлорита. Также присутствуют небольшие количества кварца, полевого шпата, и окисей железа. Из четырех типов глин один может использоваться для изготовления высококачественных огнеупоров, другой-для изготовления кафеля и фарфора, третий, самый распостраненный, — для изготовления красной керамики, четвертый не подходит для никакого специального употребления. Вычисленные запасы превышают одиннадцать миллионов метрических тонн.

Resümee—Mineralogische, chemische, und geologische Aspekte von vier Arten von Tonablagerungen in Las Aguilas Formation nordwestlich von Barker im Buenos Aires Bezirke Argentinien, deuten an, daß diese Produkte lateritischer Veränderung von precambrischem Granit zumeist aus Kaolin mit ein wenig Halloysit, Illit, und Chlorit bestehen. Kleinere Mengen von Quartz, Feldspaten, und Eisen (III) oxyden sind auch vorhanden. Von den vier Tonarten ist eine für hochqualitäts Schamottestein verwendbar, eine andere kann für Wandkacheln und Geschirr benutzt werden, eine dritte, die am meisten vorkommende, kann für rote Keramik benutzt werden, und die vierte hat keinen besonderen Nutzen. Das Vorkommen der Tonerden ist auf über elf Millionen Tonnen berechnet worden.

Résumé—Les aspects minéralogiques, chimiques, et géologiques de quatre sortes de dépôts d'argile de la formation Las Aguilas du nord ouest de la province de Buenos Aires, Argentine, indiquent que ces produits d'altération latérique de granite Précambrien sont en majeure partie kaolinitiques avec des quantités moindres d'halloysite, d'illite, et de chlorite. De petites quantités de quartz, de feldspar, et d'oxides ferriques sont aussi présentes. Parmi ces quatre sortes, une peut être utilisée pour des réfractaires de haute qualité, une autre pour des tuiles murales et la faience, et une troisième, la plus abondante, pour des céramiques rouges; la quatrième n'a aucune utilité particulière. L'abondance des argiles est calculée être en excès de onze millions de tonnes métriques.