

## NORDSTRANDITE IN BAUXITE DERIVED FROM PHONOLITE, LAGES, SANTA CATARINA, BRAZIL

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**Abstract**—Nordstrandite from Lages represents the first occurrence of this mineral in an oxisoil that does not involve the influence of a calcareous substrate. This is also the first reported occurrence in Brazil. Nordstrandite occurs near the city of Lages only in the weathered bedrock facies, mostly where the circulation of meteoric water is restricted. There is also a close association between nordstrandite and the weathering of modified pseudoleucite structures. The interaction of meteoric water and feldspathic alkaline country rock with uncommon texture and mineralogy (rich in nepheline, sodalite, nosean, analcime and natrolite) promotes the development of solutions enriched in alkaline ions with basic pH, necessary to form nordstrandite. A detailed study by X-ray diffraction (XRD) and infrared (IR) spectroscopy in natural or chemically leached samples was performed to identify the  $\text{Al}(\text{OH})_3$  polymorphs of gibbsite and nordstrandite.

**Key Words**—Bauxite, Boehmite, Gibbsite, Nordstrandite, Paraná Basin, Phonolite, Weathering.

### INTRODUCTION

Since the pioneering work of Van Nordstrand *et al.* (1956) describing the synthesis of a new form of  $\text{Al}(\text{OH})_3$ , later named nordstrandite by Papée *et al.* (1958), only a few occurrences of this aluminum hydroxide polymorph have been reported in natural conditions (Table 1). Four polymorphs of aluminum hydroxides have been described in natural environments: gibbsite, bayerite, nordstrandite, and doyleite. Gibbsite is the most common and the others are rare. Milton *et al.* (1975) suggested that the limited occurrence of nordstrandite was a consequence of its misidentification as gibbsite, because of close similarities in their XRD patterns. Nevertheless, a quarter of a century later, the number of occurrences of nordstrandite in natural environments continues to be limited to a few citations, emphasizing not only the problem of identification but also its rarity and the difficulties in preserving nordstrandite so that it may be analyzed.

According to Chao and Baker (1982) and Chao *et al.* (1985), four groups of natural occurrences of nordstrandite (Table 1) are distinguished: (1) the most important, as a weathering product in bauxitic profiles influenced by carbonate country rocks; (2) as a vein or fissure-filling mineral in dolomitic oil shale; (3) as an alteration product of dawsonite and aluminohydrocalcite; and (4) as a late-forming mineral in pegmatitic pockets and miarolitic cavities associated with nepheline syenite and sodalitic alkaline rocks. This paper describes the first occurrence of nordstrandite in Brazil. The occurrence is related to group 1 but is slightly different because the nordstrandite is formed in a highly weathered bauxite deposit derived exclusively from feldspathic alkaline rocks and there is no associated carbonate.

Environmental conditions may influence the polymorphic nature of an initial aluminum hydroxide precipitate (Rodgers *et al.*, 1991); these include pH, the  $\text{NaOH}/\text{Al}$  ratio, temperature, kinetics, and the presence or absence of ligands and foreign ions (Schoen and Roberson, 1970; McHardy and Thomson, 1971; Elderfield and Hem, 1973; Violante and Jackson, 1981; Hemingway, 1982; Hsu, 1988; Singer and Huang, 1990). Of these, high alkali concentration and neutral to basic solutions seem to be the most important factors that promote nordstrandite formation in preference to other aluminum hydroxides.

### GEOLOGICAL SETTING

Situated in southern Brazil, with the city of Lages inside its domain, the Alkaline District of Lages is a domed structure that occupies an area of 800 km<sup>2</sup> (Figure 1). The nordstrandite reported here is not widespread but restricted to bauxites developed upon alkaline rocks of Serra da Farinha Seca, located ~25 km northeast of Lages at 27°33'S and 50°13'W. The alkaline rocks crop out as isolated bodies forming dikes and sills cutting the Gondwana sequence of sedimentary rocks that form the Paraná Basin. Locally, the emplacement of these alkaline rocks follows faults and fractures associated with sills and dikes of diabase that are part of the lavas of the Serra Geral Formation (extruded between 137–127 Ma. (Turner *et al.*, 1994). Alkaline intrusions also form elongated bodies that follow the bedding of the older sedimentary rocks of the Paraná Basin. These intrusions were dated from Upper Cretaceous to Paleocene and have a median radiometric K-Ar age of 70 ± 6 Ma (Scheibe *et al.*, 1985).

Following the nomenclature proposed in the alkaline rock classification system of Le Maitre *et al.*

Table 1. Reported geological occurrences of nordstrandite.

Geological setting, location, and type	Associated Al minerals	References
Ferruginous limestones and bauxite, Jamaica <sup>1</sup>	Gibbsite, boehmite	Davis and Hill (1973)
Interface limestone and fossil soil derived from basalts and tuffs, Sarawak, Borneo <sup>1</sup>	Gibbsite, diaspore?	Wall <i>et al.</i> (1962)
Interface limestone and weathered intermediate to basic igneous rocks, Guam <sup>1</sup>	Gibbsite, boehmite	Hathaway and Schlanger (1962)
Interface limestone and weathered volcanic rock, French Polynesia <sup>1</sup>	Gibbsite, boehmite	Jamet <i>et al.</i> (1991)
Karst limestone terra rossa, Hungary <sup>1</sup>	Gibbsite, boehmite	Náray-Szabó and Péter (1967)
Karst limestone terra rossa, Montenegro <sup>1</sup>	Gibbsite, boehmite	Tertian (1966)
Karst limestone, terra rossa, Croatia <sup>1</sup>	Gibbsite, boehmite	Maric (1968)
Solution cavities on carbonate rocks, former USSR <sup>1</sup>	No data	Kulikova <i>et al.</i> (1974)
Fissure fillings in dolomitic marlstone and oil shale, Rio Blanco, Colorado, USA <sup>2</sup>	Dawsonite	Milton <i>et al.</i> (1975)
Authigenic in marine and in fluvio-deltaic strata, Sydney Basin New South Wales, Australia <sup>3</sup>	Dawsonite, alumino-hydrocalcite	Goldberry and Loughnan (1970)
Cavities in nepheline syenite, Narssarsuk, Greenland <sup>4</sup>	No data	Petersen <i>et al.</i> (1976)
Pegmatites, miarolitic cavities and xenoliths in nepheline syenite, Mont Saint Hilaire, Canada <sup>4</sup>	Doyleite, dawsonite	Chao and Baker (1982)
Bauxite, Lages, Brazil <sup>5</sup>	Gibbsite, boehmite	This study

<sup>1</sup> Weathering product in bauxitic soils associated with limestones.

<sup>2</sup> Vein or fissure-filling in dolomitic shale.

<sup>3</sup> Alteration product of dawsonite and alumohydrocalcite.

<sup>4</sup> Late mineral in alkalic igneous rock.

<sup>5</sup> Weathering product in bauxite derived from alkalic igneous rock.

(1989), foidites, feldspathic alkaline rocks (mainly phonolites) and carbonatites have been identified in the Lages Alkaline District. Phonolites, more undersaturated in silica and enriched in alkaline elements, crop out in the northern part of the district and form a sub-type of the feldspathic alkaline rocks. These phonolites are enriched with feldspathoids and zeolites due to more intense alteration related to a post-magmatic process. Especially at Serra da Farinha Seca, these phonolites are deeply weathered and form the bauxite containing nordstrandite. The bauxites are believed to be associated with a continental passive border, which have experienced extensive exposure to the Lages alkaline rocks since the Upper Tertiary, as estimated from fission track data obtained from apatites, corroborated by geomorphological and global paleoclimate reconstruction.

A complete bauxite profile at Lages has five zones on the basis of macroscopic features visible in the field: unweathered and weathered bedrock, a gibbsitic facies, reworked soil, and recent soil (Figure 2).

### *The bauxite profile*

A typical bauxite profile developed by weathering of alkaline rocks in Brazil has a sharp contact between parent rock and bauxite and lacks a saprolite horizon (Bárdossy, 1983; Delvigne *et al.*, 1987). Near Lages, the profile is different, however, and unweathered bedrock, weathered bedrock, gibbsitic facies, reworked soil and recent soil are clearly apparent (Figure 2).

The mineralogy of the unweathered phonolite is dominated by a felsic mineral assemblage (~90% of the total volume of the rock) with a sodic/potassic composition: K-rich feldspar, nepheline, sodalite (sodalite/nosean), albite and zeolites (analcime and natrolite). Mafic minerals are minor constituents and consist mostly of aegirine-augite. The fabric of phonolite associated with bauxite is porphyritic, in a degree unseen in the other varieties of felsic alkaline rocks of the district. Among the phenocrystals, octagonal sections are indicative of original leucite. Nevertheless, microscope and microprobe studies show that the leucite is

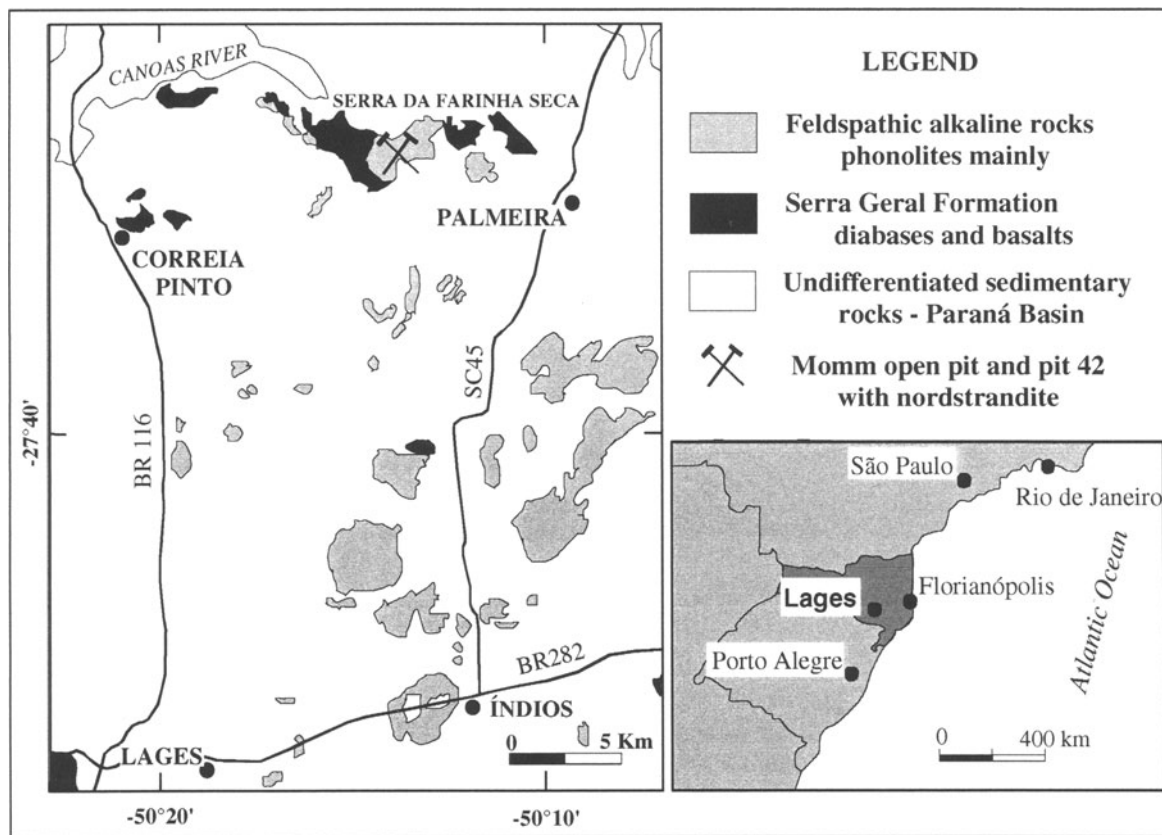


Figure 1. Simplified geological map of the Lages Alkaline District. Nordstrandite was recognized in Serra da Farinha Seca (Momm Open Pit and Pit 42).

rarely preserved and that phenocrysts are in fact transformed to an aggregate of microcrystalline K-rich feldspar and nepheline, producing pseudoleucite structures similar to those described by Edgar (1987). The phonolite associated with the bauxite also exhibits a more pronounced post-magmatic alteration that affected the whole rock and was not restricted only to rock fractures. In these fractures there was substitution of primary minerals, especially nephelines and pseudoleucite structures by sodalite, nosean, analcime and natrolite which are now dominant.

The weathered bedrock facies is a transition zone between the fresh rock and the friable upper facies (Figure 2). At the bottom of the profile, weathering develops along rock fissures and joints. With increasing weathering, there are blocks with multiple concentric zones which form the typical spheroidally weathered phonolite. Here, the partially weathered residual blocks have two main zones or rinds. Each block consists of internal fresh rock with increasing weathering towards its border. The inner weathered zone remains indurated but has a pale gray color in contrast to the dark gray color of the fresh core. The outer weathered zone, in contrast, is friable, has a white to orange color and has widespread dissolution features that contribute

to the increase in porosity and permeability. Between the blocks and, locally, between the spheroidal or ex-foliation planes, there are infiltrated halloysitic and gibbsitic veins (cutans).

The gibbsitic facies consists mainly of relict textures and structures. This facies has a very high porosity and permeability, and is the economic target of mining companies. This facies consists of hard gibbsite blocks (bauxite), embedded in a soft halloysitic saprolite with a predominant orange-red color. The proportion of bauxite blocks to halloysitic saprolite is variable laterally and depends on drainage conditions. Well-drained profiles, as in the interfluves, promote the mobilization of silica and favor gibbsite concentration, and thus the formation of a very high-quality bauxite.

At the top of the profile and below the recent soil, there is an irregular zone of reddish color formed by indurated debris within a predominant halloysitic clay matrix. Where this zone is well developed, the debris ranges in size from 5 mm to 10 cm, is poorly sorted, and has a sharp contact with the recent soil, but a gradational contact with the underlying zones. Among the debris, products of extreme weathering occur, such as bauxite blocks (pebbles) originating from gibbsitic facies and pedogenetic relicts derived from ancient

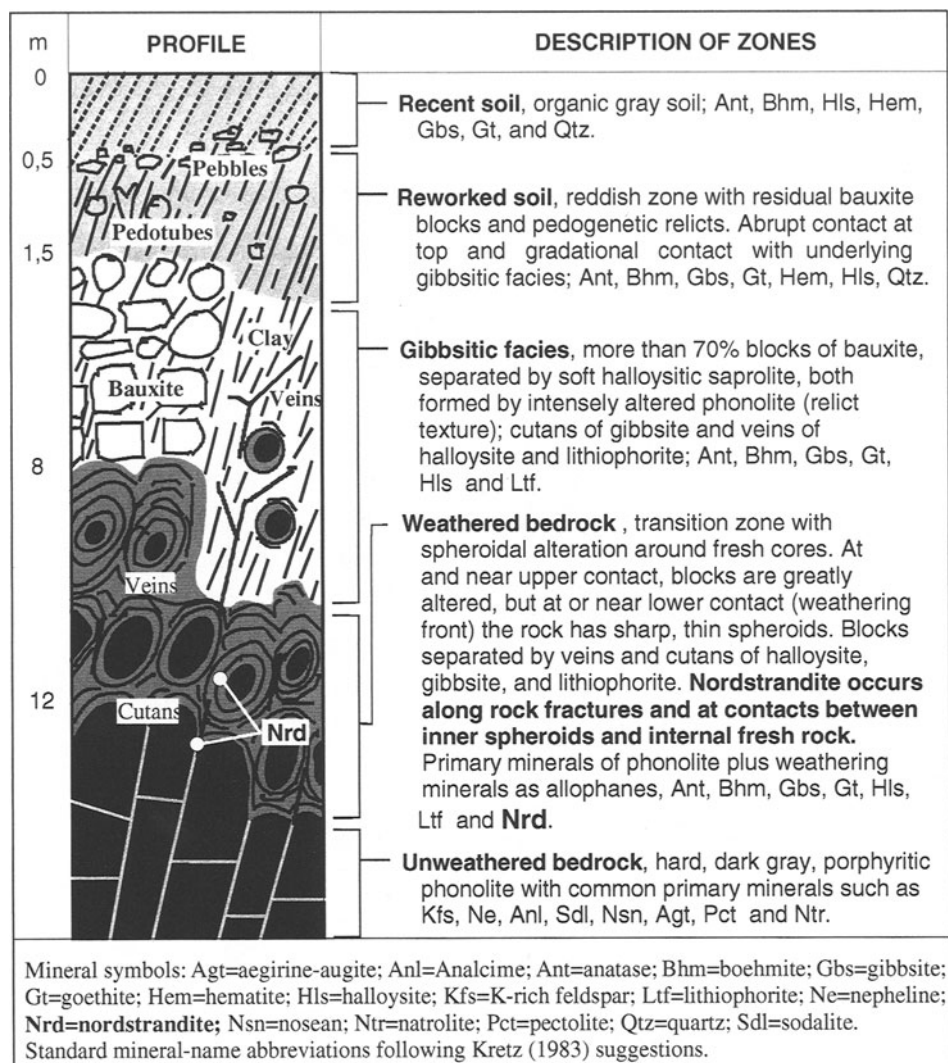


Figure 2. Typical weathering profile at Serra da Farinha Seca (Lages) and occurrence of nordstrandite.

roots of plants (pedotubes). The abundance of pebbles increases to the top of this zone. The zone was formed by the mechanical removal of the clay matrix (illuviation).

The recent soil is a thin, soft clay layer <0.5 m thick. It has a gray color owing to the influence of organic matter.

#### METHODOLOGY

The relevant rocks from the Lages area and the related weathering profiles were examined by XRD to determine the bulk mineralogy. Some samples from Serra da Farinha Seca contained nordstrandite. Additional samples were thus examined from exploratory pit 42 and the Momm Open Pit. These open pits contain profiles that are representative of the Farinha Seca Bauxite with good exposures that allow the observation of the complete succession of facies. Thirty eight

samples were obtained from the profile and sub-divided based on color, texture, homogeneity and friability. Petrographic examination identified primary minerals from which alteration products were extracted with a micro-sampler. Where possible, minerals were separated from each weathering microsystem (inner fresh rock, inner weathered zone, and outer weathered zone) to determine the effect of increasing weathering. Thin-sections and isolated fragments were studied using a conventional petrographic microscope and a scanning electronic microscope (SEM), a Jeol 6400 (equipped with a Kevex energy dispersive system).

Alteration products were studied by XRD and IR spectroscopy. The XRD was performed using a Philips PW1730 diffractometer (CoK $\alpha$  radiation at 40 kV and 40 mA). The IR spectra were obtained from KBr disks (2 mg/100 g KBr) using a Nicolet Fourier-transform spectrometer in the interval of 400–4000 cm<sup>-1</sup>. Com-

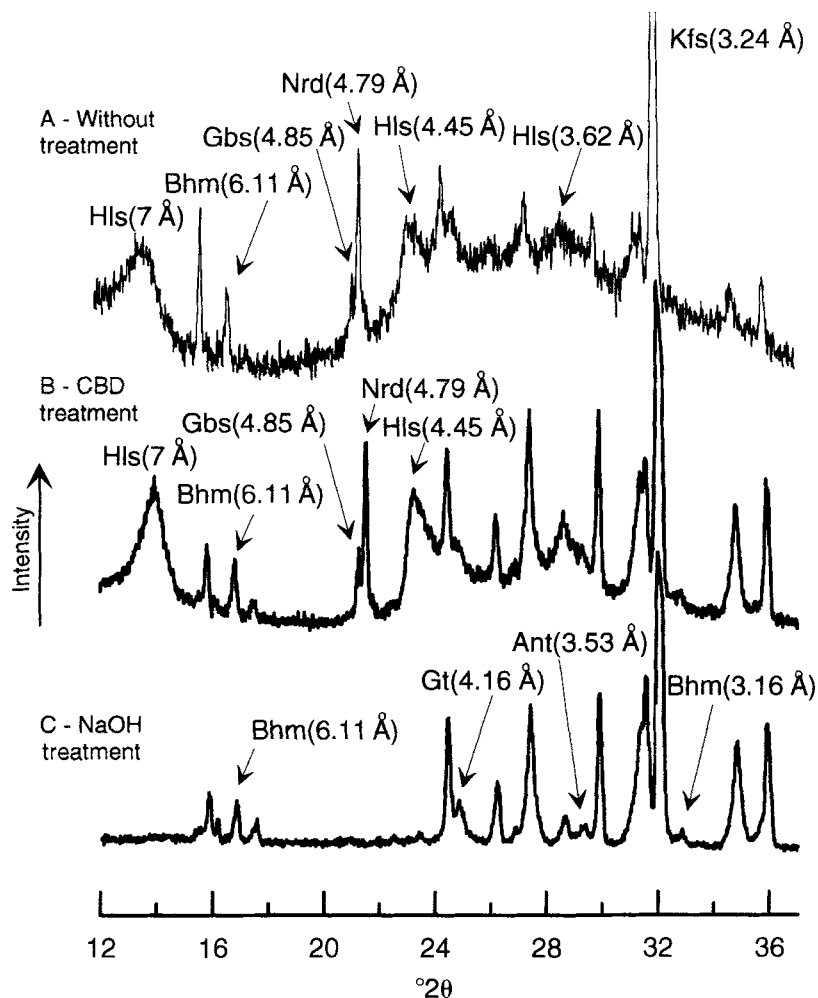


Figure 3. Powder X-ray diffraction pattern using a random powder mount of material derived from weathered bedrock facies. Peak labels follow the mineral symbols used in Figure 2.

plementary to the study of microsamples, the  $<2 \mu\text{m}$  size-fraction was extracted along the profile. The presence of nordstrandite was determined by XRD.

Additional XRD and IR spectra were obtained from chemically leached samples using 5 M NaOH (1 g of sample in 100 mL of NaOH solution allowed to boil for 60 min) and CBD (citrate-bicarbonate-dithionite) treatments (1 g of sample in 300 mL of solution of extraction CBD after 15 min at  $75^\circ\text{C}$ ), as described by Norrish and Taylor (1961), and Mehra and Jackson (1960). Each chemical treatment selectively dissolved secondary Al- or Fe-bearing minerals, thereby permitting improved identification by XRD (Kämpf and Schwertmann, 1982) and IR procedures.

Bulk density was measured in sample fragments covered by paraffin, weighed in both air and water using an analytical balance with a precision of 0.01 g. Porosity was determined by the Melcher Method, as described in Krumbein and Pettijohn (1938). Density

and porosity were used as parameters to indicate weathering stage and degree of groundwater circulation, respectively.

## RESULTS

### Identification

The combination of XRD and IR analyses together with selective dissolution proved to be effective in the identification of each aluminum phase. The XRD patterns (Figure 3A) indicate the presence of halloysite (7 and 4.45 Å), gibbsite (4.85 Å), nordstrandite (4.79 Å), and boehmite (6.11 Å) in the weathered bedrock facies. According to Berry (1974), the peaks at 4.85 and 4.79 Å are characteristic of gibbsite and nordstrandite, respectively. The CBD treatment produced a decrease in background, improved the quality of XRD patterns and enhanced the peaks of the Al-bearing secondary minerals (Figure 3B). The NaOH

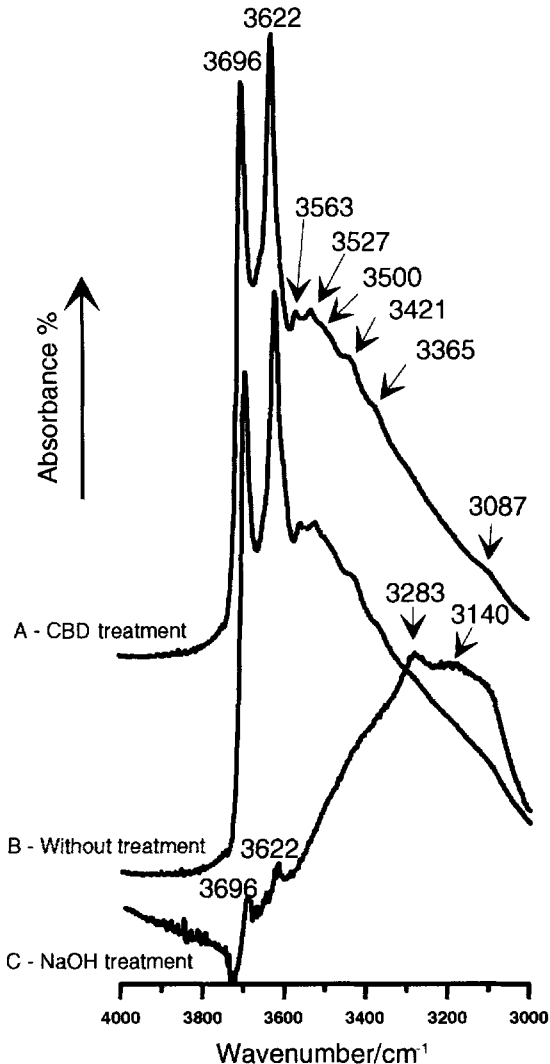


Figure 4. Detail of IR spectra of the 4000–3000-cm<sup>-1</sup> region of weathered bedrock facies with untreated and with CBD and 5 M NaOH sample treatment.

treatment (Figure 3C) dissolves halloysite, nordstrandite and gibbsite and consequently enhances the diffraction peak of other secondary phases, such as goethite (4.16 Å), anatase (3.53 Å) and boehmite (6.11 Å). Boehmite resists dissolution and this makes identification easier.

The IR spectra from the weathered bedrock sample (Figure 4A, 4B) show the vibration bands of OH groups at 3563, 3527, 3500, 3421 and 3365 cm<sup>-1</sup>, typical of nordstrandite (Palmieri *et al.*, 1985). The IR spectrum of the sample treated with NaOH (Figure 4C), where the aluminum hydroxides were dissolved, is different. However, small bands at 3696 and 3622 cm<sup>-1</sup> show the high sensitivity of this method for the identification of halloysite still remaining in this sample. In the same spectrum, the large band at 3140 cm<sup>-1</sup> is

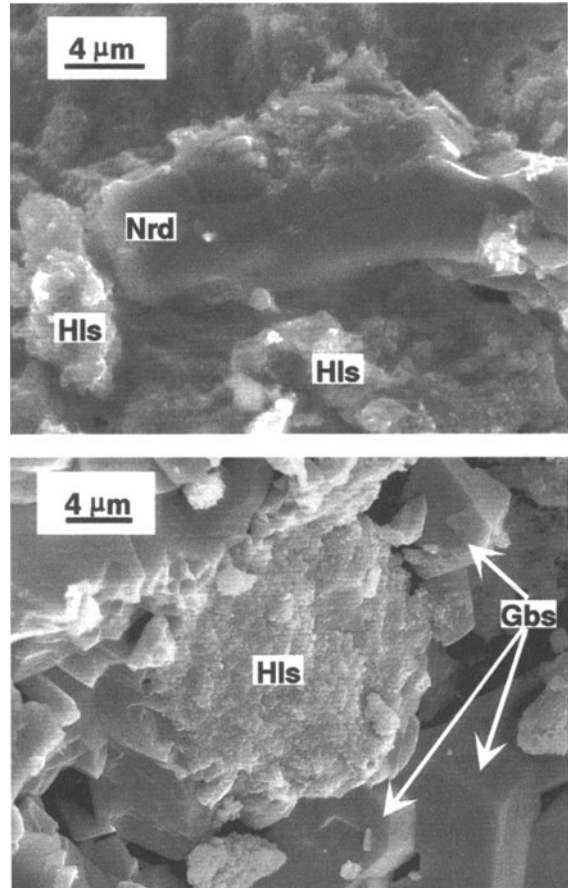


Figure 5. Scanning electron photomicrograph of samples derived from weathered bedrock facies from Lages. Upper, morphology of nordstrandite (Nrd). Lower, morphology of gibbsite (Gbs). Hls = spherical halloysites.

typical of goethite (Farmer, 1974) which was concentrated in the sample by chemical treatment. The 3283 cm<sup>-1</sup> and the 3087 cm<sup>-1</sup> bands, from the NaOH and the CBD-treated samples, respectively, were described as boehmite absorption bands (Ryskin, 1974; Violante and Huang, 1985; Lee and Condrate, 1995).

At Lages, the intimate association of nordstrandite and gibbsite makes the morphological study with the SEM difficult, but, according to Violante *et al.* (1982), nordstrandite crystals frequently form rectangular parallelepipeds. The SEM images confirm that where XRD and IR show nordstrandite to be dominant, rectangular sections are common (Figure 5, upper) and, where XRD and IR show gibbsite to be abundant, hexagonal sections prevail (Figure 5 lower).

#### Occurrence of nordstrandite in the profile

The results show that gibbsite is the common polymorph of aluminum hydroxide in Lages. Nordstrandite is rare and restricted to sites close to fresh rock in the weathering front (Figure 2). At Lages, nordstrandite

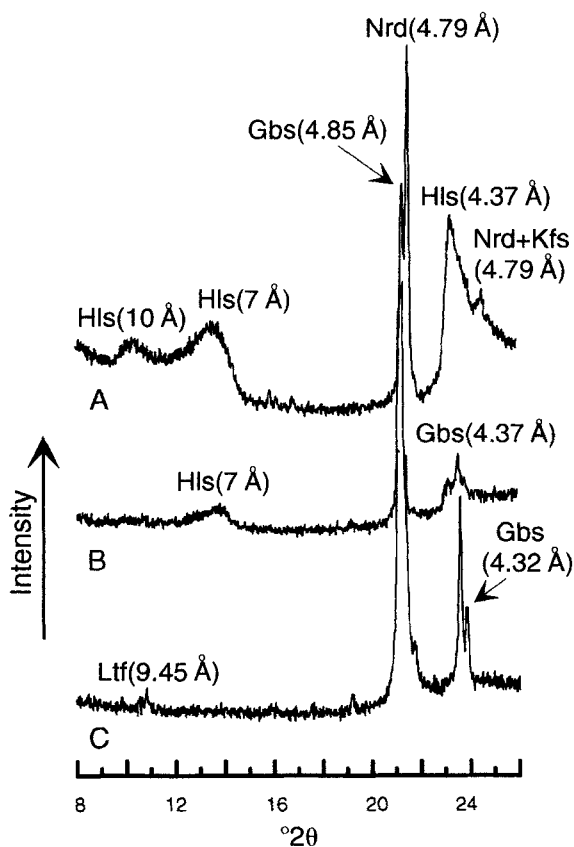


Figure 6. XRD pattern using a random powder mount of minerals separated from thin layers along fractures inside the weathered bedrock facies. Peak labels as in Figure 2.

was detected only where the process of weathering is incipient, notably inside the inner spheroidal zone and in the limited space of fractures in weathered bedrock. In rock joints in the interface between unweathered and weathered phonolite, there are mm-sized thin layers of friable material where nordstrandite occurs (Figure 6A). Detailed study shows that halloysite, gibbsite and a manganese mineral (lithiophorite) commonly occur in these sites (Figure 6B and 6C), followed by allophane and boehmite. In addition to occurrences along joints or fractures, nordstrandite is only clearly identified inside the inner portion of spheroidally weathered phonolite, close to fresh rock. The XRD patterns of the inner weathered rock halo (Figure 7, trace B) show that the proportion of nordstrandite to gibbsite is nearly equal, but the proportion of gibbsite increases greatly a few centimeters beyond the inner halo and nordstrandite nearly disappears (Figure 7, trace A). Conversely, the fresh phonolite (Figure 7, traces C, D and E) exhibits only reflections of the phonolite primary minerals.

#### DISCUSSION

The geological occurrences of nordstrandite (Table 1) show that the origin of this aluminum phase is related

to endogenic or exogenic domains. Endogenic origins are linked with late-stage minerals in a magmatic environment (Narssarsuk, Greenland and Mont Saint Hilaire, Canada) or with diagenetic phases (Rio Blanco, Colorado, USA and Sydney Basin, Australia). For both origins, nordstrandite is associated with uncommon minerals such as aluminum carbonate and dawsonite,  $\text{NaAlCO}_3(\text{OH})_2$ . In all other known occurrences, nordstrandite is a weathering mineral directly associated with gibbsite and boehmite, forming in oxisoils derived from limestones.

At Farinha Seca, nordstrandite is only of exogenic origin and evolved directly from weathered alkaline rocks without the presence of carbonate substrate. This interpretation is consistent with the absence of nordstrandite outside the zones of groundwater circulation. At Lages, nordstrandite occurs only at the weathering front in phonolite joints and in spheroidally weathered phonolite, where it is associated with weathered aggregates of postmagmatic Na-bearing minerals (feldspatoids and zeolites). Nordstrandite develops early, close to the weathering front, in a process involving dissolution that promotes the crystallization of nordstrandite and boehmite. An example of the transformation of Na-bearing minerals to nordstrandite is easily observed in the pseudoleucites. In Serra da Farinha Seca phonolite, pseudoleucites are transformed to an aggregate of sodium aluminous silicates, mainly sodalite, nosean, analcime, and natrolite due to a postmagmatic process. Material extracted from altered pseudoleucites at the weathering front clearly shows the crystallization of nordstrandite, gibbsite, and boehmite (Figure 8).

Additionally, measurements of density and porosity indicate that low porosity occurs at sites where nordstrandite is found (inner spheroidal zone in Figure 9). Consequently, nordstrandite clearly occurs only in environments where water circulation is restricted.

As at other nordstrandite occurrences (Table 1), boehmite is an associated mineral at Lages. The genetic similarities between these two aluminum minerals in the Serra da Farinha Seca suggest that crystallization of boehmite occurs in an early weathering stage. The direct crystallization of boehmite does not agree with the suggestion of a late origin of boehmite by gibbsite dehydration (Tardy, 1994).

Figure 10 summarizes the behavior of nordstrandite at Lages. Nordstrandite is not present in the highly weathered zones (gibbsitic facies) where the secondary  $\text{Al}(\text{OH})_3$  polymorph typically found is gibbsite. Boehmite persists from the incipient weathered facies to highly weathered gibbsitic facies as a residual phase. The dissolution and destabilization of nordstrandite is related to increasing rock porosity. Indeed, the free circulation of meteoric water changes the chemical conditions, removing alkalis and decreasing the pH to produce mildly acid conditions, under which

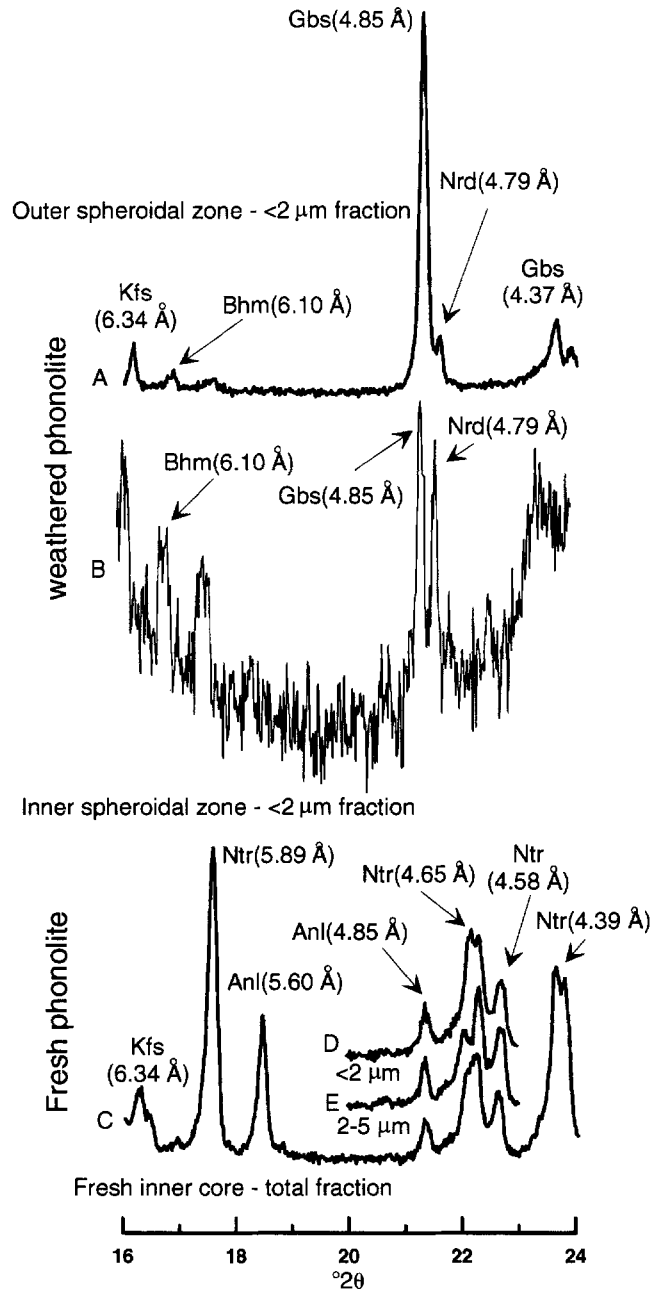


Figure 7. XRD patterns involving a spheroidally weathered phonolite from weathered bedrock facies: (A) outer spheroidal zone, (B) inner spheroidal zone, and (C), (D), and (E) internal fresh core. Mineral labels as in Figure 2. Vertical axis is intensity.

nordstrandite is no longer stable and gibbsite crystallizes.

The mechanism involving meteoric alteration was shown to be controlled by local chemical equilibria (Righi and Meunier, 1995), which is influenced by the composition of primary minerals, secondary minerals, and fluids. In the present case, nordstrandite formation appears to be strictly related to local

chemical conditions established by the dissolution of feldspaths and zeolites and associated with the pseudoleucite structures. At the weathering front (low permeability), these highly unstable minerals react with meteoric water to produce alkali-rich concentrated solutions with basic pH. Here, the microsystem conditions favor the precipitation of a polyminerale secondary assemblage where the  $\text{Al}(\text{OH})_3$



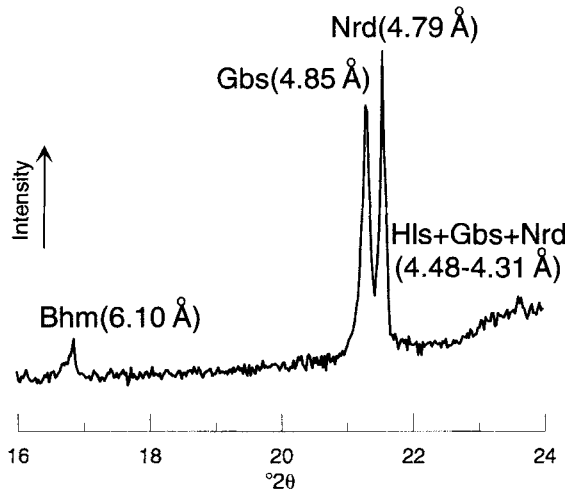


Figure 8. XRD pattern (random powder) of material extracted from weathered pseudoleucite (see Figure 2 for mineral labels).

polymorphs are nordstrandite and gibbsite, followed by the oxy-hydroxide boehmite.

The dissolution of feldspathoid and zeolite-rich aggregates increases the porosity and the permeability of the host rock. Then, the solutions can be renewed, thereby leading to the dilution of dissolved compo-

nents. In open microsystems, the alkali concentration of solutions becomes too low for nordstrandite to remain stable. This early-formed phase is dissolved in turn and replaced by a more stable phase: gibbsite. In highly permeable zones, the circulation of meteoric water controls the dilution of the solution. Feldspathoids and zeolites are replaced directly by gibbsite which forms cutans, frequently observed in the bottom of the profile or as a relict, in the superposing gibbsitic facies.

For all known occurrences of nordstrandite in the weathering environment, nordstrandite is associated with oxisols and bauxites developed on calcareous substrates (Wall *et al.*, 1962; Hathaway and Schlanger, 1962; Davis and Hill, 1973; Jamet *et al.*, 1991). This is confirmed by experiment showing that neutral to basic pH and high alkali concentrations are needed to crystallize nordstrandite (Violante and Violante, 1980; Hsu, 1967). In contrast, gibbsite precipitates from acid, alkali-poor concentrated solutions (Barnhisel and Rich, 1965). In the Lages bauxite profile, nordstrandite forms directly from phonolite weathering, without a calcareous substrate. Nordstrandite is restricted to the weathering front and strongly associated with parent rocks containing Na-bearing feldspathoids and zeolites. In these sites of incipient weathering, nordstrandite is associated with allophane, gibbsite, boehmite and halloysite.

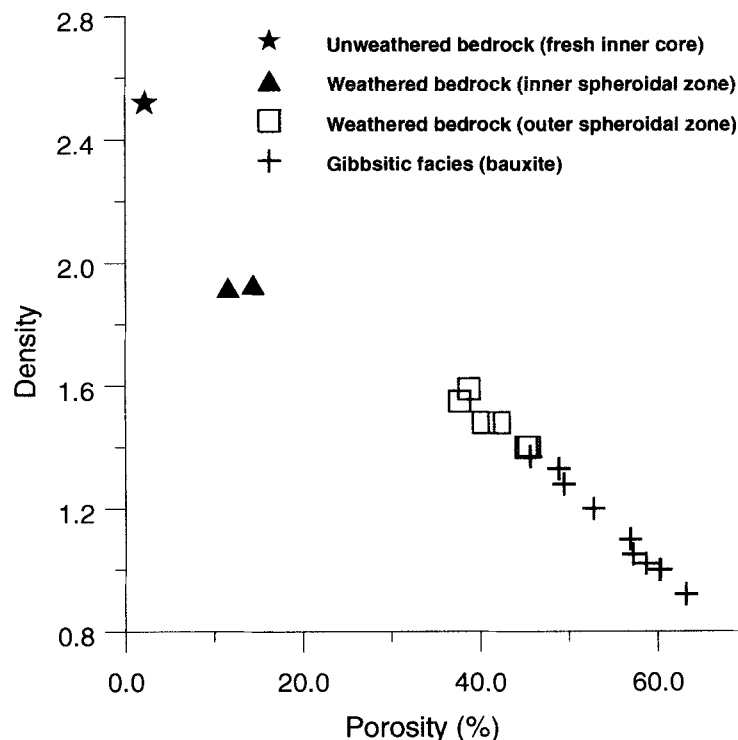


Figure 9. Plot of density vs. porosity measured in samples that represent coherent facies with an increase in weathering (unweathered phonolite to gibbsitic facies).

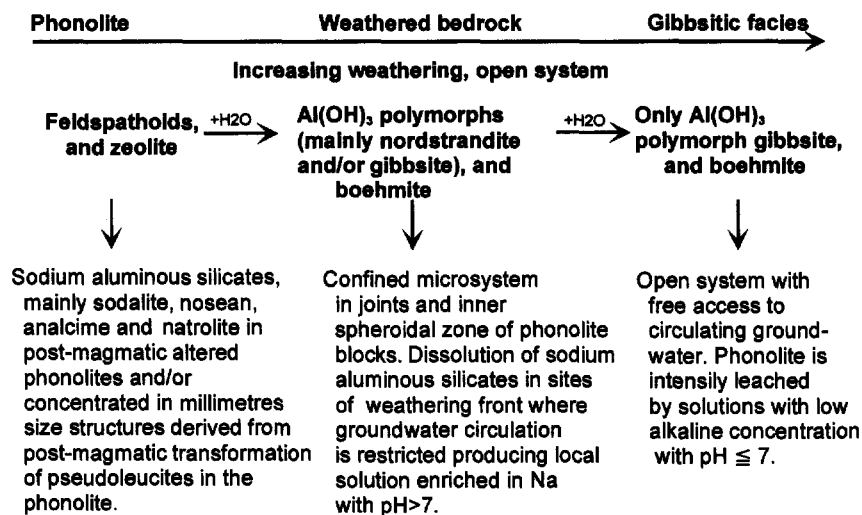


Figure 10. Proposed sequence of Al precipitation products as influenced by different solutions and pH characteristics.

## CONCLUSIONS

(1) Nordstrandite crystallizes as a weathering product and occurs in bauxite that evolved directly from phonolite weathering without the influence of a limestone substrate as is common elsewhere. (2) Nordstrandite is restricted to facies with an incipient degree of weathering. (3) The crystallization of nordstrandite is controlled by local chemical equilibria (microsystems). Rock composition, associated with a low level of water flow found in microsites of the weathering profile, provide the conditions for nordstrandite crystallization. (4) Two essential factors influence the formation of nordstrandite: (a) presence of feldspathoids (nepheline, sodalite) and zeolites (analcime, natrolite); and (b) low porosity and permeability (confined microsystem). (5) Nordstrandite is unstable in highly porous and permeable facies where the level of water flow is high.

## ACKNOWLEDGMENTS

The authors acknowledge I. Valetton for her efforts in the study of bauxite genesis in Brazil and the editorial assistance of P.E. Potter. This research was supported by Project Capes/Cofecub N° 185/96, Capes (Brazil), Project Pronex 7697100600 and Fapergs (RS/Brazil).

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(Received 15 March 2000; revised 13 December 2000; Ms. 434; A.E. Warren D. Huff)