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FELDSPAR WEATHERING IN LATERITIC SAPROLITE

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Abstract—Feldspars in granitic saprolite in southwestern Australia have altered to halloysite, kaolinite, and gibbsite with no evidence of noncrystalline material. The secondary minerals are commonly present as intimate mixtures within altered feldspar grains, but discrete zones of gibbsite or halloysite-kaolinite also are present. Variations in the chemical microenvironment within micrometer-size zones in grains apparently controlled the type and distribution of secondary minerals. The alteration of both plagioclase and alkali feldspars involved congruent dissolution by soil solution and crystallization of halloysite, kaolinite, and gibbsite from this solution. Highly altered feldspar grains consist of etched feldspar fragments embedded within a highly porous framework of subhedral to euhedral platy crystals of kaolinite and gibbsite, or of spherical and felted aggregates of halloysite.

Key Words—Feldspar, Gibbsite, Halloysite, Kaolinite, Noncrystalline material, Weathering.

INTRODUCTION

Feldspars alter during weathering and soil formation to various secondary minerals including a noncrystalline phase (Eswaran and Bin, 1978; Eggleton and Buseck, 1980), halloysite and kaolinite (Hughes and Brown, 1977; Wilke *et al.*, 1978), gibbsite (Parham, 1969; Lodding, 1972), montmorillonite (Wilson *et al.*, 1971), and muscovite mica or illite (Exley, 1976; Eggleton and Buseck, 1980). The differences in chemical macroenvironments between soils or the difference in chemical microenvironments within the same soil account for the formation of different alteration products.

The role of microenvironment in determining the nature and distribution of secondary minerals in soils is poorly understood. Eswaran and Bin (1978) concluded from a study of feldspar alteration that the formation of each secondary mineral requires a specific microenvironment. Secondary minerals forming within small pores in weathering primary mineral grains may crystallize from more concentrated solutions than those phases that form in more easily leached intergranular joints and in larger pores (Tardy *et al.*, 1973). The submicroscopic mechanisms of feldspar weathering and secondary mineral formation have not been clearly established. In particular, considerable uncertainty remains as to whether secondary minerals crystallize directly from solution or develop from noncrystalline phases. Eggleton and Buseck (1980) concluded from their high-resolution transmission electron microscope (HRTEM) study of the weathering of microcline and plagioclase that these minerals alter to illite or dehydrated smectite via noncrystalline, 250-Å diameter, ring-shaped particles. In contrast, Berner and Holdren (1979) using scanning electron microscopy (SEM) and X-ray photo-electron spectroscopy to study the surfaces of weathered feldspar grains, found no surface layer of intermediate composition, although these techniques have a resolution of a few tens of Ångstrom units. Clearly, there is need for additional data on the mechanisms of feldspar alteration as revealed by high-resolution electron microscopy and related measurements. The present paper reports an investigation of the alteration of individual feldspar grains in a granitic saprolite.

MATERIALS AND METHODS

Feldspar grains exhibiting various amounts of alteration were separated from weathered Archaean granitic rocks (adamellites) in the saprolite zones of three bauxite laterite profiles exposed in a railway cutting at Jarrahdale, 45 km southeast of Perth, Western Australia. Detailed profile descriptions and general weathering patterns for these and similar deeply weathered materials were given by Sadleir and Gilkes (1976) and Gilkes *et al.* (1973), respectively. Quartz veins and dolerite dykes which occur as intrusions in the granitic rocks are preserved within the weathered profiles, demonstrating that the profiles were formed by *in situ* chemical weathering processes. The perfectly preserved granitic fabric within the saprolite and pallid zones is evidence of the isovolumetric nature of the weathering process. Two profiles (A, B) exhibit a sequence of zones typical of the laterites in the region; parent material (>12 m thick), saprolite zone (12–9 m), pallid zone (9–5 m), mottled zone (5–1 m), and sandy pisolitic duricrust (1–0 m). A third profile (C) is only 6 m in depth, and the saprolite zone here grades

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directly into sandy pisolitic duricrust; no mottled or pallid zones are present.

Altered feldspar grains were separated by first disaggregating the bulk samples of saprolite by gentle shaking in water adjusted to pH 8 with NaOH, and then handpicking the grains under a stereo-microscope. Moderately altered grains consisting of ~50% secondary minerals were abundant in the untreated samples; however, they lacked cohesion and so were too fragile to resist dispersion. Only those grains with a connected matrix of feldspar or clay were able to resist dispersion; partly altered grains consisting of separated feldspar fragments in a poorly connected clay matrix were readily disaggregated by this treatment. Thus, studies on separated grains were restricted to the slightly and highly altered types. Chemical analyses of fresh and altered feldspar grains were made on polished thin sections using an ARL SEMQ electron microprobe (specimen current 5 μa at 15 kV). Single-grain, X-ray diffraction patterns of ~1-mm diameter, slightly altered and highly altered grains were made using a Gandolfi single crystal camera (Gandolfi, 1967). The bulk samples and altered feldspar grains were impregnated with resin and thin-sectioned for petrographic examination. Semi-quantitative determinations of the abundance of minerals in single grains were made by thermogravimetric analysis. Thermogravimetric (TG) and differential thermogravimetric (DTG) curves were obtained in flowing air at a heating rate of 10°C/min using a Perkin Elmer TG 52 instrument. DTG maxima within the temperature ranges 60–120°C, 250–285°C, and 450–520°C correspond to water losses from halloysite(10Å), gibbsite, and halloysite(7Å and 10Å) + kaolinite, respectively, were used to calculate mineral contents. The accuracy of the technique was checked using known mixtures of feldspars, kaolinite, and gibbsite.

Altered feldspar grains were investigated by scanning electron microscopy (SEM) using a Philips PSEM 500 instrument. Specimens were mounted on aluminum stubs and coated with a 50-Å layer of platinum in a vacuum evaporator. The grains were broken to expose fresh surfaces prior to coating. Pt-shadowed carbon replicas of altered feldspars were prepared for transmission electron micrographs (TEM) of the fracture surfaces of altered grains using a Hitachi HU11B instrument. Freshly exposed feldspar surfaces were also gently scraped with a scalpel blade to remove surface material for dispersion in water and deposition on carbon-coated grids for TEM examination and selected area electron diffraction (SAD). Altered feldspar grains were crushed and dispersed for TEM examination; the clay fractions of soil samples were also examined by XRD and TEM. Zones of interest within altered feldspar grains were removed from thin sections by a microcoring device, the resin was dissolved in Eposolve 70, and the minerals were examined by TEM and XRD in a Philips powder camera.

Table 1. Abundance (%) of secondary minerals in granitic saprolite.

Profile	Horizon	Depth (m)	Halloysite (10Å)	Halloysite(7Å)-kaolinite	Gibbsite
A	Lower saprolite	12–11	1	9	1.5
	Middle saprolite	11–10	2	15	<1
	Upper saprolite	10–9	2	25	<1
B	Lower saprolite	12–11	3	12	2
	Middle saprolite	11–10	5	14	1
	Upper saprolite	10–9	6	18	2
C	Lower saprolite	6.5–6	—	10	15
	Middle saprolite	6–5.5	—	12	17
	Upper saprolite	5.5–5	—	14	23

RESULTS AND DISCUSSION

Composition of saprolite

From the weight losses shown in the thermal analysis curves at 60–70°, 250–280°, and 470–480°C, the amounts of halloysite(10Å), gibbsite, and halloysite(7Å + 10Å) + kaolinite, respectively, were estimated. This technique is particularly sensitive, and our experiments have shown that it can detect less than 1% of these minerals. Residual feldspar, quartz, and mica were estimated semi-quantitatively by XRD. As shown in Table 1, the amounts of halloysite and kaolinite increase towards the surface in all three profiles; the amount of gibbsite increases towards the surface only in profile C; and halloysite(10Å) is absent in profile C. Because halloysite, gibbsite, and kaolinite are all present in the same horizons, the chemical environment must have varied significantly within each soil horizon.

Various degrees and styles of alteration of feldspar were noted in the saprolite materials. Plagioclase grains were generally more altered than associated alkali feldspars. The earliest stage of alteration of both feldspars consisted of the development of patches of cloudy material (Figure 1A). Rich (1972) showed that microcline exhibits both exsolution lamellae and cross-hatched patterns with the development of secondary minerals following these patterns. Highly altered grains consist of a fine-grained material of low birefringence, which could not be identified from its optical properties (Figure 1B). XRD, TEM, and SEM examination of this material showed it to be mixtures of feldspar, halloysite, and gibbsite.

Altered feldspar grains identified in thin sections were grouped according to the degree of alteration which was estimated by the percentage of each feldspar grain that had been replaced by clay (Figure 2). Although alteration of feldspars to clay is clearly more advanced in the upper saprolite, large differences in the degree of alteration of individual feldspar grains exist within each horizon of the three profiles. This variation sug-

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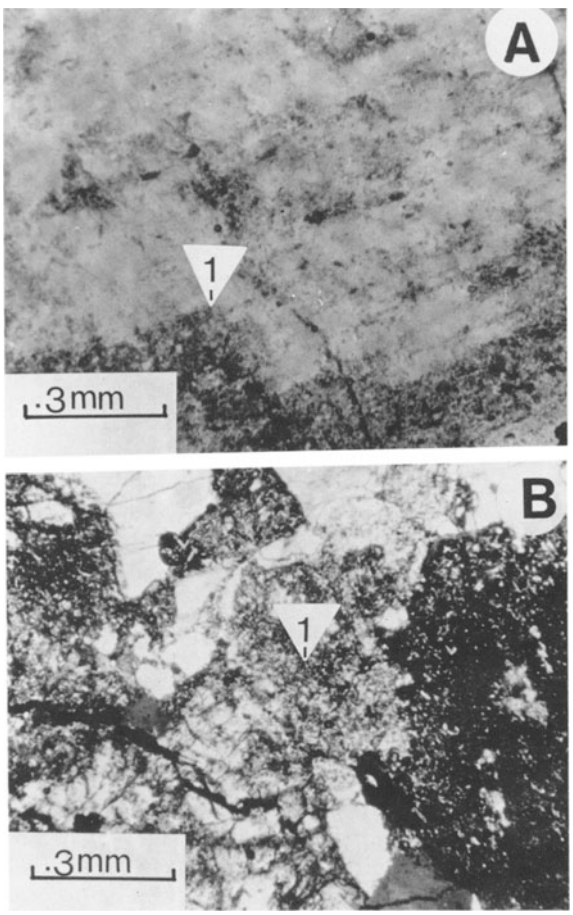


Figure 1. Micrographs of altered feldspar grains: (A) Optical micrograph of a thin section showing early stages of alteration of feldspar to a cloudy material (1); (B) Optical micrograph showing a later stage of alteration (1) to a mixture of halloysite, kaolinite, and gibbsite.

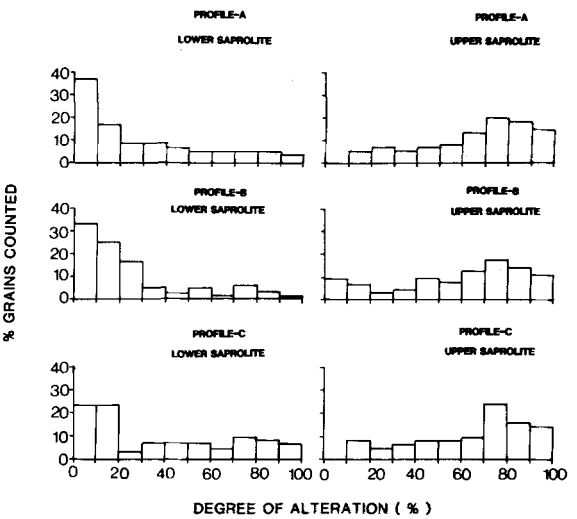


Figure 2. Histograms showing the extent of alteration of individual feldspar grains expressed as percentage of grain altered to clay vs. the percentage of altered grains estimated from thin sections of lower and upper saprolite zones of profiles A, B, and C.

gests that a range of chemical microenvironments may have existed within a single horizon.

The chemical compositions of individual altered feldspar grains and the fine-grained alteration products derived from the same grains were determined in polished thin sections of the lower saprolite from profile A by electron microprobe analysis (Table 2). Similar results were obtained for the other two profiles. The changes in chemical composition due to weathering are fully consistent with the mineralogical data described above. A marked increase in the concentration of Al, a decrease in the concentration of Si, and a loss of alkalis took place during the alteration of feldspars to halloysite, kaolinite, and gibbsite. The small amounts of Na, K, and Ca remaining in the alteration products are due to the presence of about 4% sub-micrometer-size feldspar fragments. The average Al/(Al + Si) ratios for alteration products from alkali and plagioclase feldspars within any one horizon are very similar. The values for profile A data shown in Table 2 are 0.63 and 0.61, for altered alkali and plagioclase feldspars, respectively, and are consistent with the fine-grained alteration products being a mixture of halloysite-kaolinite (Al/(Al + Si) = 0.49) and gibbsite (Al/(Al + Si) = 1.0), as shown by the XRD analyses. The Al/(Al + Si) ratio, however, varied from point to point in altered feldspar grains, and some points gave values corresponding to either pure halloysite-kaolinite or pure gibbsite. The variations in this ratio are not dependent on the feldspar species inasmuch as the alteration products derived from both plagioclase and alkali feldspar exhibited a similar range of Al/(Al + Si) ratios (Figure 3). Furthermore, the alteration products for both feldspar species mostly yielded ratios near those corresponding to halloysite-kaolinite or gibbsite. In the up-

Table 2. Average chemical composition (%) of feldspar fragments and adjacent fine-grained alteration products within partly altered feldspar grains from the lower saprolite of profile A.

	SiO_2	Al_2O_3	Na_2O	K_2O	CaO
Alkali feldspar	65.51	18.56	0.47	16.12	—
Adjacent alteration products	34.12	45.38	—	0.31	—
Plagioclase feldspar	62.40	24.16	8.59	0.12	4.74
Adjacent alteration products	33.20	46.10	0.25	—	0.15

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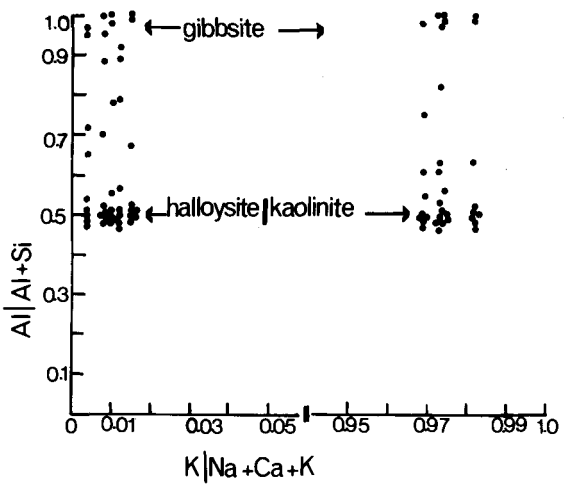


Figure 3. Plot of K/(Na + Ca + K) vs. Al/(Si + Al) for regions within single, partly altered feldspar grains from the lower saprolite of profile A demonstrating a tendency to either a gibbsite or halloysite-kaolinite composition and that there is no difference in alteration products for alkali and plagioclase feldspars.

per saprolite of profile A, the Al/(Al + Si) values for the alteration products derived from both plagioclase and alkali feldspar were ~0.50, indicating the presence of only halloysite-kaolinite, which was confirmed by XRD analysis.

The porosity of the alteration products was calculated using the values from Table 2 and assuming isovolumetric weathering (Millot, 1970), as well as no loss of Al. On average, about 0.60 ml of kaolinite-halloysite and gibbsite occupy the 1.0 ml previously occupied by a parent feldspar grain; thus, the porosity is about 40%. This highly porous structure is clearly seen in SEM images of altered grains (Figures 4B, 4D, 4F).

Mineralogy of single grains

From optical microscopic examination, feldspar grains were grouped into grains having the same degree of alteration. As explained above, feldspar grains that were approximately "half altered" lacked cohesion and could not be isolated for analysis. The mineralogical composition of 215 single grains was determined by XRD and TGA (Figure 5). Slightly altered feldspars were most common in the lower saprolite of profiles A and B (Figure 2), and the alteration products were entirely halloysite-kaolinite in profiles A and B, but contained gibbsite as well in profile C. A few highly altered grains were present in the lower saprolite of the profiles A and B and contained approximately 1:1.5 mixtures of gibbsite and halloysite-kaolinite in profile A, and a 1:6 mixture in profile B. Highly altered grains in the upper saprolite of profiles A and B had altered solely to halloysite-kaolinite. In profile C, both slightly and highly altered grains in the lower and upper saprolite had altered to 3:1 mixtures of gibbsite and halloysite-kaolinite. Large variations were noted in the ratio of kaolinite-halloysite to gibbsite for grains from a single horizon.

Morphology and mineralogy of alteration products

The surface of the residual feldspar in partly altered grains was invariably highly etched as illustrated in Figure 6A for a fragment of feldspar from which adhering clay had been removed by washing. Identical etchpits have been reported from laboratory dissolution studies of feldspars (Lunderstrom, 1970; Berner and Holdren, 1977, 1979; Keller, 1978) and for weathered feldspar grains from soils (Wilson, 1975; Tazaki, 1976; Berner and Holdren, 1977, 1979; Keller, 1978). Although some etchpits of this type follow former exsolution lamellae and twin planes (Wilson and McHardy, 1980), no evidence has been found in the present study or in published works to suggest that the crystal structure of the feldspar controls the nature and orientation of its weathering products. Halloysite, kaolinite, and gibbsite simply form in a random arrangement in etchpits and on feldspar surfaces.

Feldspar fragments are commonly coated with clay which fills etchpits (Figure 6B) and which consists of an intimate mixture of the secondary minerals (Figure 6C). Material scraped from the surface of partly altered feldspar grains consists of various mixtures of kaolinite, halloysite, gibbsite, and angular feldspar fragments (Figure 6D). In the lower saprolite, kaolinite and gibbsite occur as platy, rarely hexagonal, euhedral crystals (Figure 4A). Halloysite mostly occurs as poorly formed tubular crystals which show some evidence of unrolling in the upper saprolite samples. Where halloysite is the dominant alteration product it occurs in felted masses (Figure 4B) or as radiating spherical aggregates resembling those described by Eswaran and Bin (1978), Keller (1978), Diamond and Bloor (1970), and Gilkes and Suddhiprakarn (1980).

Although most of the particles seen in electron micrographs were easily recognized as crystalline kaolinite, halloysite, or gibbsite, some very small (~200 Å) disc- or sphere-shaped particles were noted, which on the basis of their morphology, could be a noncrystalline material (Figure 4C).

Single-crystal electron diffraction patterns of halloysite tubes (Figures 7A, 7B) showed most of them (96%) to be elongated along their b-axes with the remainder (4%) being elongated along their a-axes. This result is very similar to that obtained by Honjo *et al.* (1954). To distinguish between kaolinite and gibbsite which have similar platy morphologies and which give similar SAD patterns, an internal gold standard was used to calibrate SAD patterns and also to provide a shadow for thickness measurements. Using this procedure, the small difference in spacing between the 060 reflection

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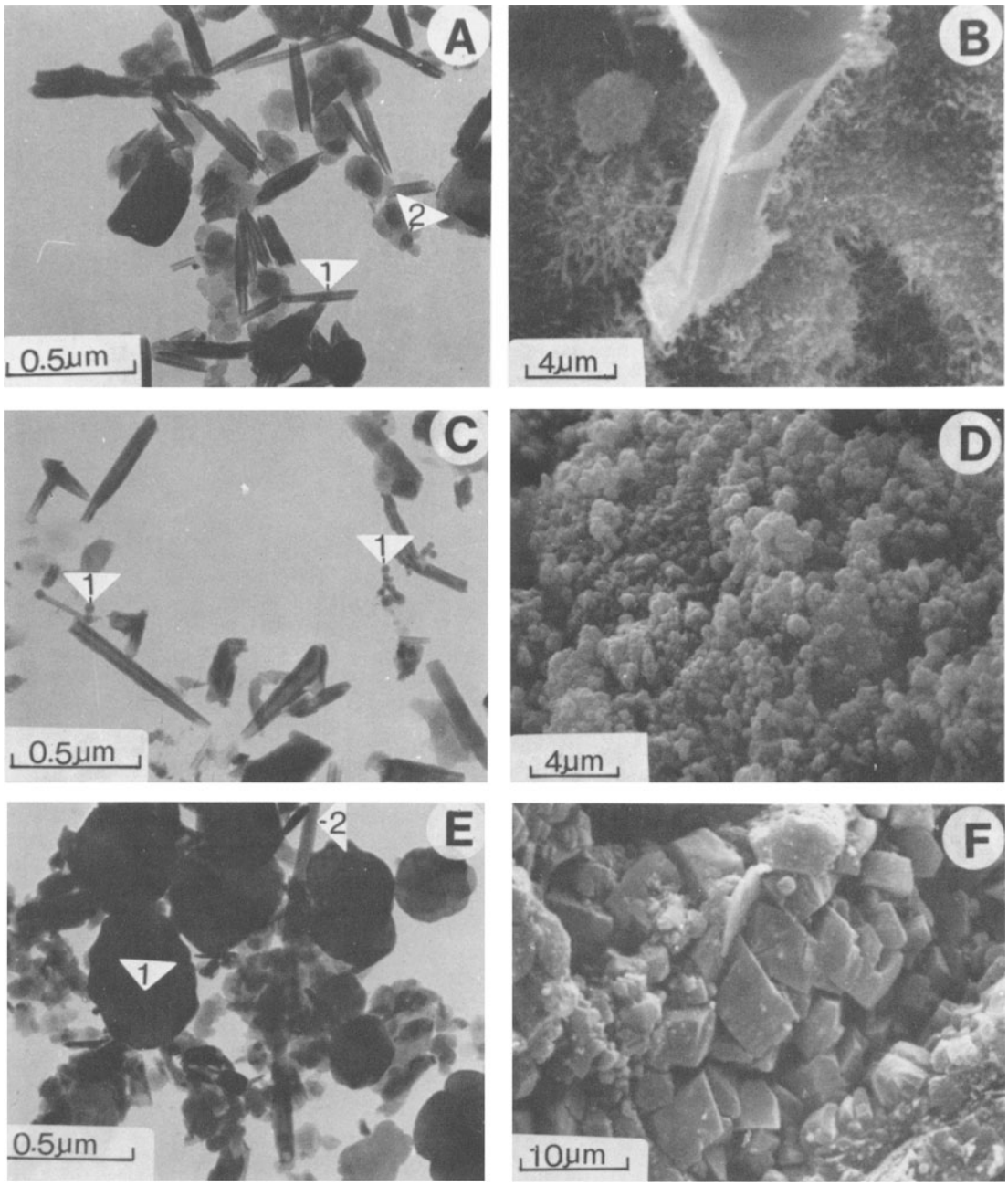


Figure 4. Transmission (TEM) and scanning (SEM) electron micrographs of materials from slightly altered feldspar grains: (A) TEM of the clay fraction of grains from lower saprolite of profile A showing irregular tubular halloysite (1) and platy hexagonal crystals of kaolinite (2). (B) SEM of a grain from the lower saprolite of profile B showing halloysite as felted masses of tubes within etchpits. (C) TEM of the clay fraction of a grain from the lower saprolite of profile A showing halloysite, kaolinite, and some small, rounded, noncrystalline(?) particles (1). (D) SEM of a grain from the upper saprolite of profile C showing aggregates of crystals of gibbsite and kaolinite arranged in a highly porous framework. (E) TEM of the clay fraction of grains from the upper saprolite of profile C showing rounded platy crystals of gibbsite and kaolinite (1) and a few tubular crystals of halloysite (2). (F) SEM of a grain from the lower saprolite of profile C showing ~10-μm euhedral crystals of gibbsite.

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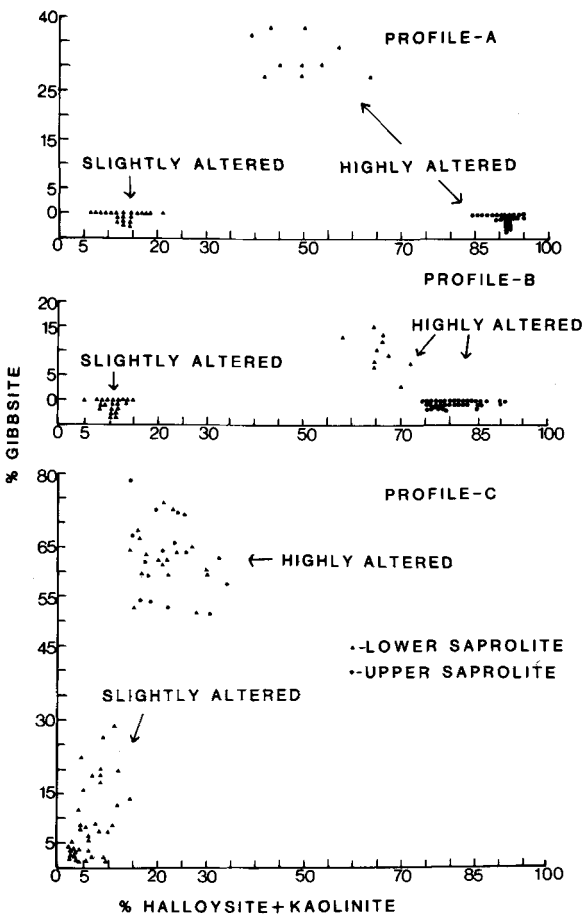


Figure 5. The distributions of (halloysite + kaolinite) vs. gibbsite in individual altered feldspar grains from profiles A, B, and C.

of kaolinite at 1.49 Å and the 330 reflection of gibbsite at 1.46 Å was determined (Figures 7C, 7D, 7E, 7F). The thickness of the ~0.3-μm diameter kaolinite and gibbsite crystals shown in Figure 7 are 0.04 μm and 0.05 μm, respectively. Most anhedral particles were found by SAD to be halloysite, kaolinite, or gibbsite.

The highly altered grains from the upper saprolite consisted mainly of porous aggregates of kaolinite and gibbsite (Figure 4D). Gibbsite commonly occurred as ~0.5-μm size, rounded platy crystals (Figure 4E) although large, ~10-μm size, euhedral gibbsite crystals were present in cracks (Figure 4F) as has been observed in lateritic soils by Eswaran *et al.* (1977).

Evidence for noncrystalline weathering products

No materials resembling the ring-shaped, noncrystalline aluminosilicates described by Eggleton and Buseck (1980) were observed. The very small spherical or disc-shaped particles shown in Figure 4C were too small to be identified by selected area electron diffraction (SAD) and may equally well be very small kaolinite or gibbsite crystals. From SAD data almost all of the dispersed alteration products were feldspar, halloysite, kaolinite, or gibbsite.

No evidence was found for the existence of noncrystalline weathering products. Particles smaller than 0.1 μm commonly gave very weak SAD patterns, and for some of these, the distinction between a noncrystalline and a crystalline structure was therefore impossible. Such particles comprised only a minor proportion of the sample; hence, if noncrystalline material is present it cannot be a major alteration product.

Although discrete noncrystalline grains were not detected, such material *could* be present as grain coatings on the various secondary minerals and feldspar fragments. Such noncrystalline surface coatings observed in bright- and dark-field electron micrographs are, however, commonly due to surface contamination in the electron microscope (Agar, 1967). Many dark field/bright field pairs for secondary minerals and feldspar fragments were taken (Figures 8C and 8E), and the edges of the grains of the bright and dark field micrographs coincided at the limits of resolution of the technique (~20 Å). This coincidence indicates that diffraction was occurring throughout the feldspar grain at these points; a noncrystalline coating thicker than ~20 Å could not be present. It is not possible to obtain coincident bright and dark field micrographs for entire grains, thus thin noncrystalline coatings or inclusions could be present in some regions of grains but would have escaped detection.

Noncrystalline material is clearly not an appreciable constituent of slightly altered feldspar grains either as discrete phases or as coatings. It is therefore unlikely to be a common intermediate phase in the alterations of feldspars to secondary minerals in these soils. This interpretation is consistent with the findings of Wilson *et al.* (1971) who observed that alteration of feldspar to montmorillonite is *via* solution without any intermediate crystalline or noncrystalline phase. In contrast, Eswaran and Bin (1978) and Eggleton and Buseck (1980) suggested that feldspars may alter *via* a noncrystalline gel phase.

These apparently contradictory results may simply be due to the fact that the various workers studied quite different material. Differences in chemical environment and alteration mechanisms may produce quite distinct alteration products. For example, the formation of noncrystalline compounds may be a consequence of the slow removal of dissolution products from primary minerals in poorly drained profiles or in microenvironments within individual grains. The lateritic soils sampled for this study are generally well drained and characterized by the formation of stable secondary minerals. Intermediate crystalline minerals such as smectite and illite are not common in these soils.

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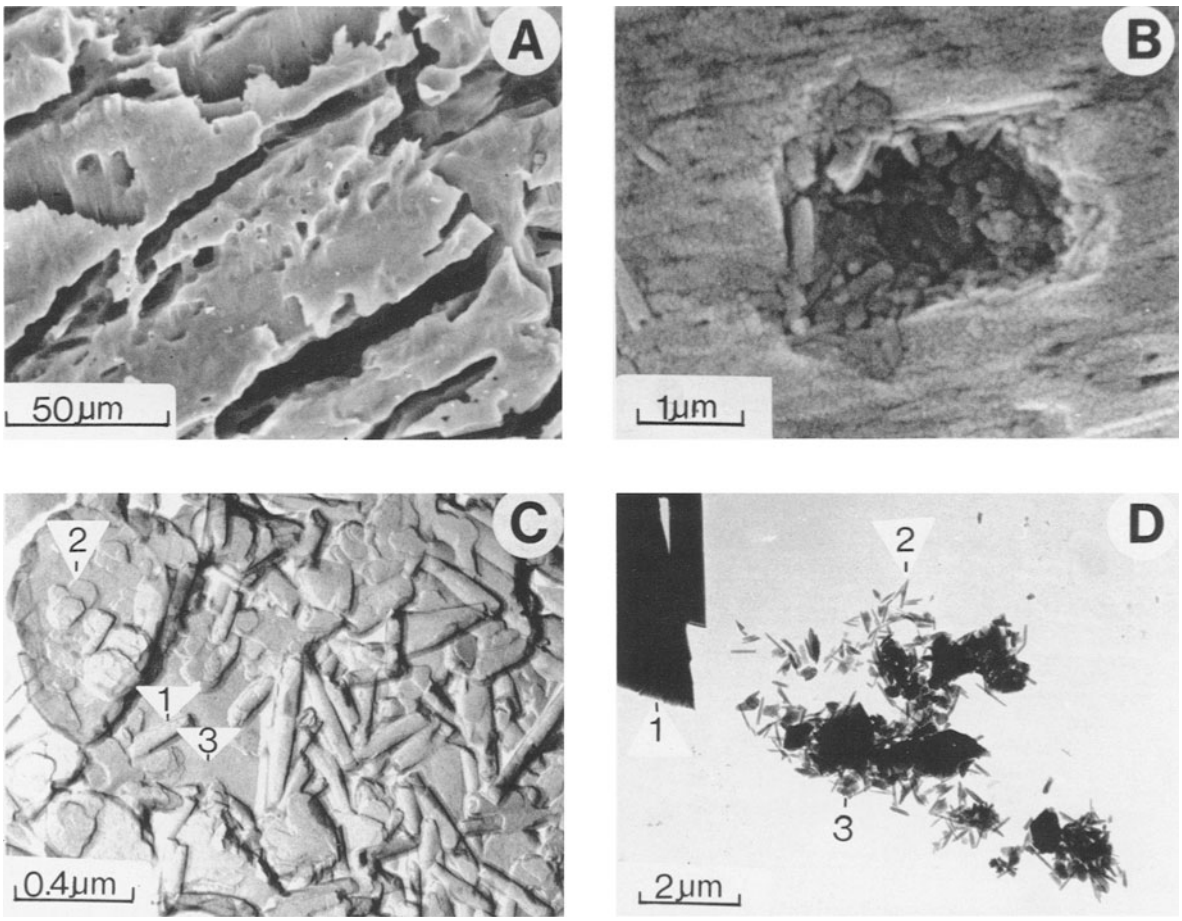


Figure 6. (A) Scanning electron micrograph of the washed surface of a slightly altered feldspar grain showing a highly etched surface. (B) Scanning electron micrograph of the surface of a slightly altered grain showing tubular halloysite crystals on the surface and within an etchpit. (C) Transmission electron micrograph of a replica of the surface of a slightly altered feldspar grain showing an intimate mixture of tubular halloysite crystals (1) and subhedral to euhedral hexagonal kaolinite crystals (2) on a smooth feldspar surface (3). (D) Transmission electron micrograph of material scraped from interior of a slightly altered feldspar grain showing angular fragments of feldspar (1) tubular crystals of halloysite (2) and platy crystals of kaolinite (3).

Small scale variations in microenvironment

Although some zones within the altered feldspar grains may contain a single secondary mineral, many zones exist in which halloysite, kaolinite, and gibbsite are intimately associated. The spatial extent of these zones can be determined by electron microprobe analysis. Typical distributions of Si, Al, Ca, and Na in a partly altered plagioclase grain are shown in Figure 9. Most of the parent plagioclase grain has dissolved, leaving feldspar fragments separated by cracks which are partially filled with secondary minerals. The distribution of Na and Ca shows the location of the residual plagioclase feldspar fragments which vary in size from 10 to 100 μm. Al and Si are present both in feldspar and in the fine-grained alteration products; these elements were determined quantitatively at many points across the altered grains. Values of the ratio Al/(Al + Si) ($\times 10$) are shown in Figure 9F. The theoretical values of this ratio for halloysite-kaolinite and gibbsite are 5 and 10, respectively. Intermediate values correspond to mixtures of these minerals. The distribution of Al and Si within the grain shows that discrete halloysite-kaolinite and gibbsite, as well as various mixtures of these, were present within the volume of about 10 μm^3 which contributes to X-ray emission from each point.

No systematic distribution of gibbsite and halloysite-kaolinite was observed within altered feldspar grains although discrete zones of gibbsite or halloysite-kaolinite were noted within the grains. Figure 10 shows discrete zones (>50 μm) of gibbsite and kaolinite-hal-

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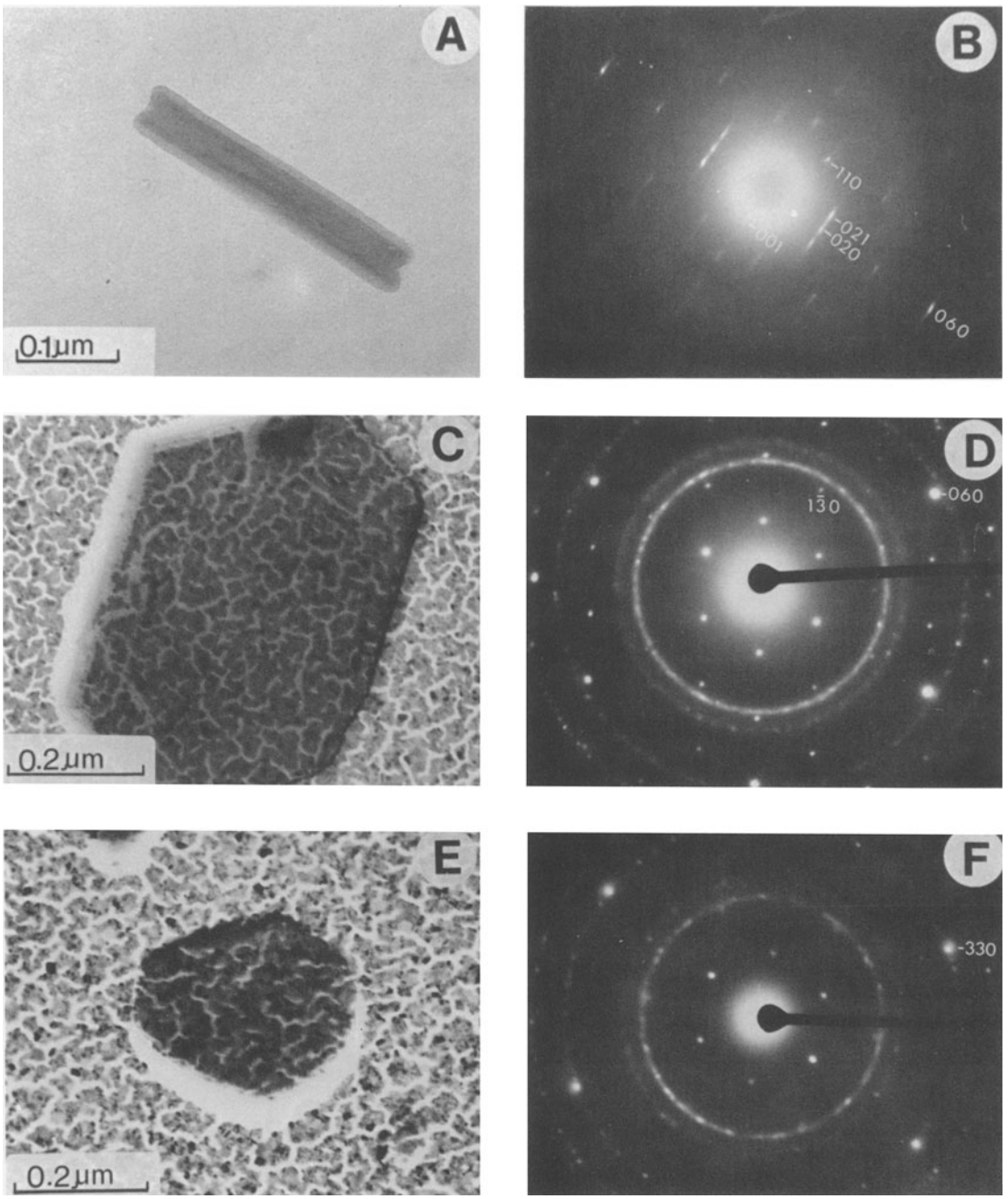


Figure 7. Transmission electron micrographs of single crystals of halloysite, kaolinite, and gibbsite (A, C, E) and their indexed selected area electron diffraction patterns (B, D, F) which are correctly oriented relative to the corresponding micrograph (i.e., no rotation error). Rings in D and F are for a gold internal standard that also provides a shadow ($\tan^{-1}$l) for thickness measurements in C and E. The halloysite tube is elongated along its b axis.

loysite in an altered alkali feldspar grain. The fine-grained alteration product in the right-hand side of the field gave a Al/(Al + Si) (×10) ratio of 5 and is halloysite-kaolinite, whereas on the left-hand side of the field the ratio is 10 and the material is gibbsite. The boundary between the two zones gave a ratio of 7, corresponding to a mixture of the two minerals. Thus, in this example microchemical environment must have

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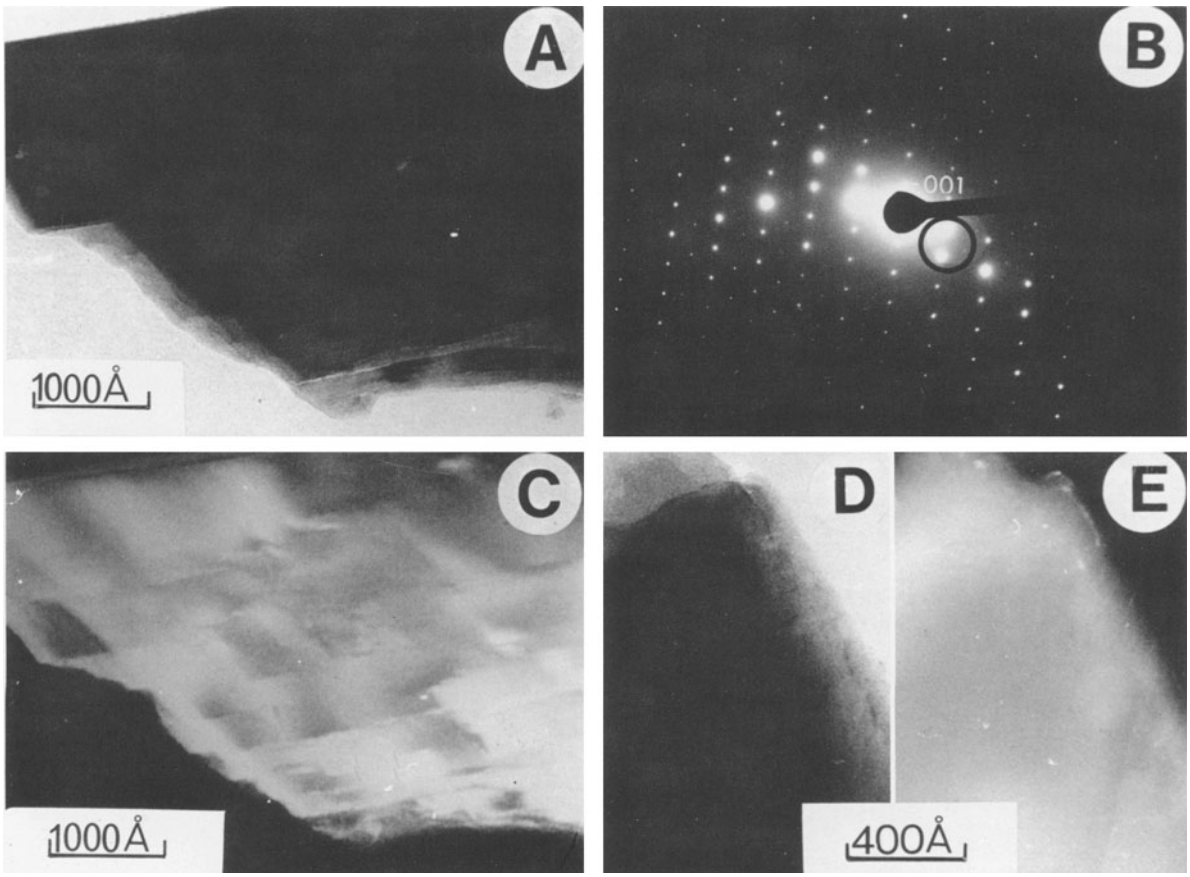


Figure 8. Bright-(A) and dark-(C) field transmission electron micrographs of a feldspar fragment and its selected area diffraction pattern (B). The dark-field micrograph was taken with the aperture selecting the ringed reflections shown in (B). (D, E) High-magnification images of part of the crystal shown in (A) and (C). In many places the edges of bright- and dark-field images of the fragment coincide, demonstrating that no noncrystalline surface coating is present.

differed within a 200 × 200 μm region in this weathering feldspar grain.

Generally the alteration products within partly altered feldspar grains are organized into zones which are either dominantly gibbsite or halloysite-kaolinite. This trend is illustrated by a histogram for data from all profiles (Figure 11). Mixtures of gibbsite and halloysite-kaolinite usually exist at interfaces between discrete mineral zones. Thus, micrometer-size zones of complete desilication or partial desilication of feldspar exist within individual weathering grains. The origins of these zones are not understood, but they may be simply a consequence of differences in permeability to water in various parts of altered grains. Zones of clay accumulation would restrict water movement and protect feldspar fragments from leach solutions and reduce the diffusion of ions from the feldspar surfaces to the soil solution in the larger cracks and pores. The complex three-dimensional organization of pores and the extent to which cracks are connected to the main drainage cracks and fissures in saprolite cannot be determined for the very small areas examined by optical and electron optical techniques. Thus, the two-dimensional variation in chemical microenvironment identified here cannot be directly related to drainage in grains which requires a knowledge of the distribution of cracks and pores in all three dimensions.

GENERAL DISCUSSION

Geochemical conditions in the lower saprolite of profiles A and B favored crystallization of halloysite, kaolinite, and gibbsite within altered feldspar grains, whereas in the upper saprolite only halloysite and kaolinite formed. In profile C halloysite, kaolinite, and gibbsite formed in both the lower and upper saprolite. Differences in the geochemical microenvironments within single grains and in the geochemical macroenvironments between both horizons and profiles affected the nature of alteration products. Both micro- and macroenvironments influenced drainage conditions

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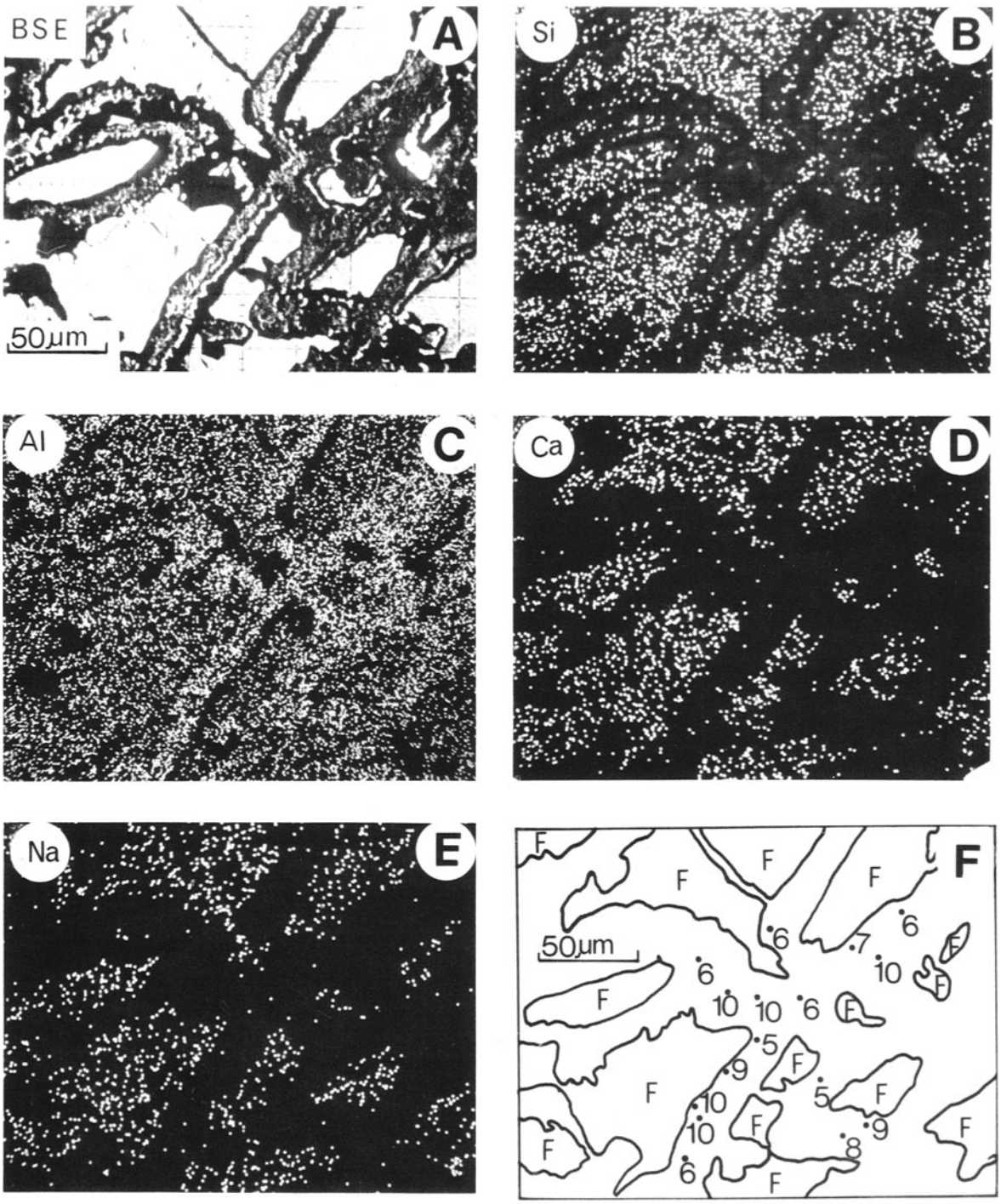


Figure 9. Electron microprobe back-scattered electron (BSE) and X-ray images showing the distribution of Si, Al, Ca, and Na in an altered plagioclase feldspar grain (B, C, D, E). Fragments of feldspar are revealed by Ca and Na concentrations and are separated by cracks partly filled with secondary minerals. (F) Drawing of the altered grain showing values of Al/(Al + Si) (×10) for secondary minerals at the points indicated. A value of 10 indicates 100% gibbsite, whereas a value of 5 indicates 100% halloysite-kaolinite; intermediate values indicate mixtures of these minerals. (F = feldspar.)

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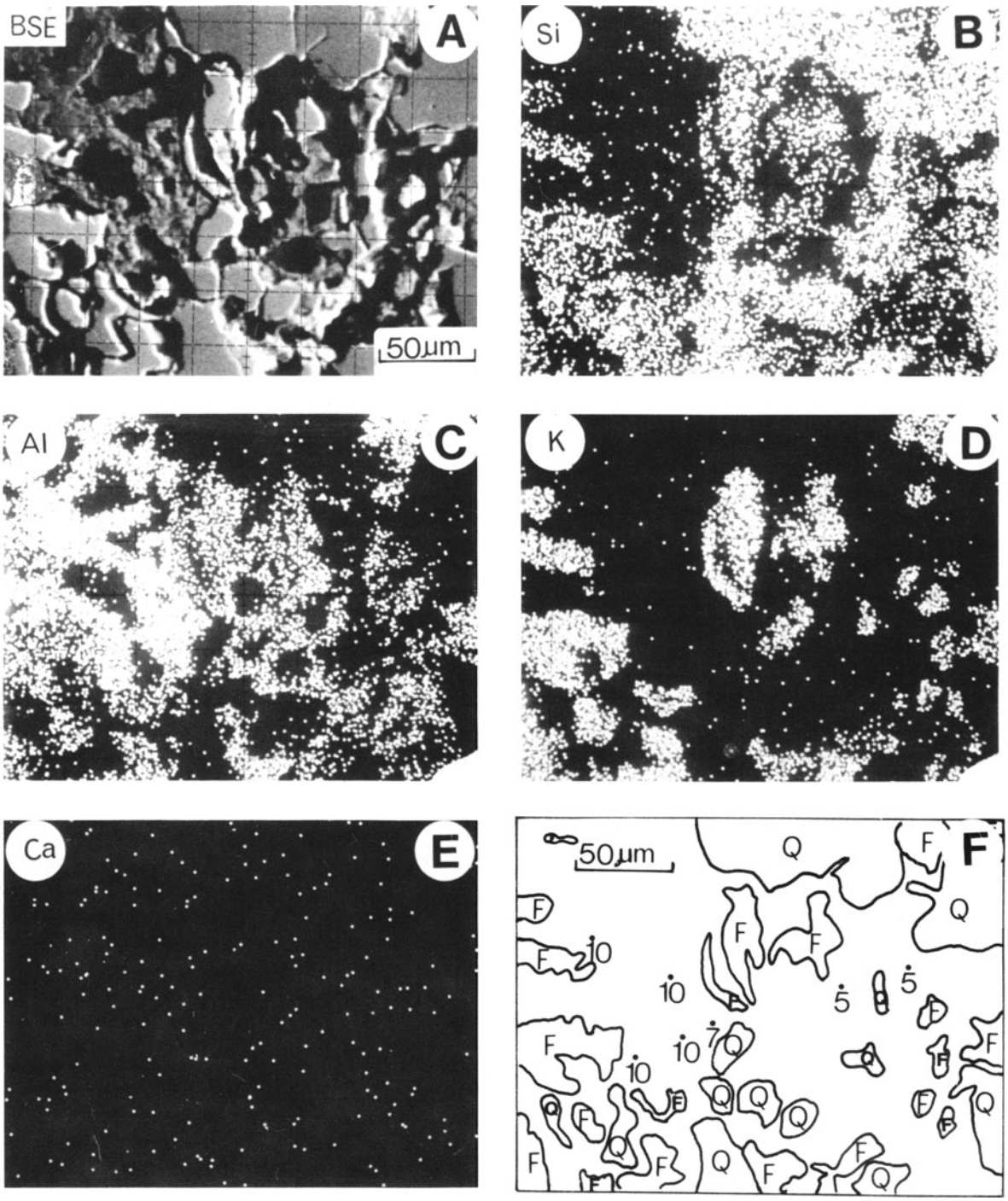


Figure 10. Electron microprobe back-scattered electron (BSE) and X-ray images showing the distribution of Si, Al, K, and Ca in an altered alkali feldspar grain (B, C, D, E). Fragments of feldspar are revealed by K concentrations and are separated by cracks filled by secondary minerals. (F) Drawing of the altered grain showing values of Al/(Al + Si) (×10) for secondary minerals at the points indicated (Q = quartz inclusions; F = feldspar).

and, as a consequence, the concentrations of Si, Al, and other ions in soil solution. In the lower saprolite, the coarse fabric dominated by sand-sized grains of feldspar and quartz allowed water to drain more freely. Thus, Si levels in the soil solution may have remained sufficiently low for gibbsite to form in well-drained microenvironments, but not so low as to prevent the formation of halloysite-kaolinite in partially enclosed

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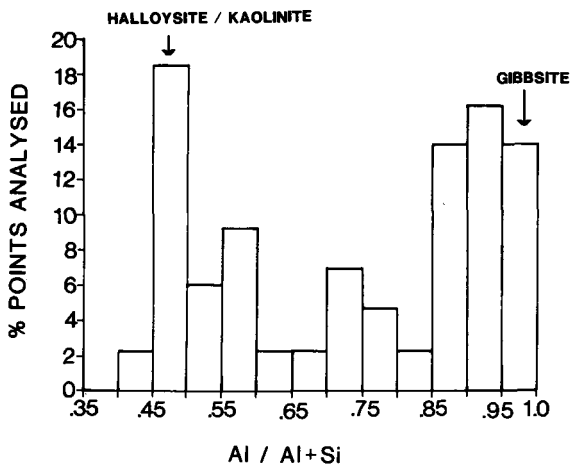


Figure 11. A histogram showing the distribution of the ratio Al/(Al + Si) amongst points analyzed in alteration products within cracks in altered feldspar grains from the lower saprolite zones of profiles A, B, and C. The ratios for gibbsite and halloysite-kaolinite are shown.

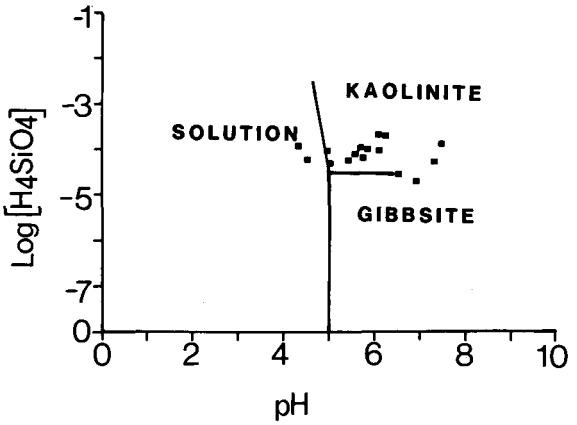


Figure 12. Plot of the values of pH and log (H_4SiO_4) for ground water from weathering granites in the Darling Range (Peck *et al.*, 1982) in stability fields of gibbsite and kaolinite taken from Garrels and Christ (1965).

and protected microenvironments. In these latter environments Si released from weathering feldspars was apparently retained. Support for this hypothesis has been provided by Tardy *et al.* (1973) who found that plagioclase altered to montmorillonite and vermiculite in narrow fissures, but kaolinite formed in well-drained pores.

Some parts of altering feldspar grains were apparently better drained than others due to an almost random pattern of cracking and to the development of clay coatings which restricted water movement and ion diffusion. Gibbsite may have formed in better-drained regions within grains where the circulation of water was most rapid. Garrels and Christ (1965) considered that both kaolinite and gibbsite form in bauxitic soils where the equilibrium concentration of Si in soil water is between 1.5 and 3 ppm which is near the value of ~0.5 ppm predicted for equilibrium between gibbsite and kaolinite. If this equilibrium was attained in regions of a partly altered feldspar grain, halloysite, kaolinite, and gibbsite could have crystallized. Concentrations of Si in pore-water in micrometer-size zones were not determined in the present study. Ground waters from many bore holes near the study area showed Si concentrations of 0.3 to 6.3 ppm, and pH varied from 4.5 to 7.3 (Peck *et al.*, 1982). Concentrations of Al in ground water were generally not determined, but the few available analyses indicate very low concentrations (<0.1 ppm). The pH and Si concentration of local ground water are plotted in Figure 12 together with the equilibrium stability data of Garrels and Christ (1965). The ground-water values are near the join of the stability fields of gibbsite and halloysite/kaolinite. Thus, the present geochemical microenvironment in grains within the saprolite may fluctuate between the stability fields for gibbsite and halloysite-kaolinite due to the poor connection between bulk ground water in large cracks and pores and the water in micropores within clay in contact with dissolving feldspar surfaces.

One final consideration is that these lateritic soil profiles may be dated from the Tertiary era and would have experienced changes in climate, hydrology, etc. that could have altered the composition of ground waters and resulted in changes in the nature of feldspar alteration products with time. Thus, the coexistence of gibbsite and kaolinite-halloysite in single grains may simply be a consequence of the successive imposition of different weathering regimes.

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Резюме—Фельдшпаты в граинтовом сапролите в юго-западной Австралии изменялись в галлуазит, каолинит и гиббсит, без доказательства присутствия некристаллических материалов. Вторичные минералы находятся обычно как внутренные смеси в зернах фельдшпата, но дискретные зоны гиббсита или галлуазита-каолинита также присутствуют. Перемены в химическом микроокружении внутри микроскопических зон в зернах, кажется, контролируют тип и расположение вторичных минералов. Видоизменение обоих плагиоклаза и щелочных фельдшпатов включало соответственно растворение в почвенном растворе и кристаллизацию галлуазита, каолинита, и гиббсита из этого раствора. Значительно видоизмененные зерна фельдшпата состоят из изъеденных фрагментов фельдшпата, окруженных сильно пороватой сетью субгедральных до евгедральных плоских кристаллов каолинита и гиббсита, либо сферических и войлочных аггрегатов галлуазита. [E.G.]

Resümee—Die Feldspäte in einem granitischen Rückstandsgestein von SW-Australien haben sich in Halloysit, Kaolinit, und Gibbsit umgewandelt, wobei es keinen Hinweis für nichtkristallisierte Substanzen gibt. Die sekundären Minerale treten meist eng vermischt in umgewandelten Feldspatkörnern auf. Es gibt jedoch auch begrenzte Zonen, in denen Gibbsit oder Halloysit-Kaolinit getrennt auftreten. Variationen im chemischen Mikrobereich innerhalb von Mikrometer-großen Zonen in den Körnern kontrollieren offensichtlich die Art und Verteilung der Sekundärminerale. Die Umwandlung von Plagioklas und Alkalifeldspat umfaßt konkruente Auflösung durch Bodenlösung und die Kristallisation von Halloysit, Kaolinit und Gibbsit aus dieser Lösung. Stark umgewandelte Feldspatkörner bestehen aus angelösten Feldspatfragmenten, die in einem stark porösen Gerüst aus hypidiomorphen bis idiomorphen tafeligen Kristallen von Kaolinit und Gibbsit liegen oder in einem Gerüst aus kugeligen und dichten Aggregaten von Halloysit eingebettet sind. [U.W.]

Résumé—Des feldspars dans de la saprolite granitique en Australie du sud-ouest se sont altérés en halloysite, kaolinite et gibbsite, sans évidence de matériel non-cristallin. Les minéraux secondaires sont communément présents en tant que mélanges intimes au sein de grains de feldspar altéres, mais des zones discrètes de gibbsite ou d'halloysite-kaolinite sont également présentes. Des variations dans le microenvironement chimique au sein de zones de taille micrométrique dans les grains ont apparemment contrôlé le type et la distribution de minéraux secondaires. L'altération à la fois de plagioclase et de feldspars alkalins a impliqué la dissolution congruente de solution de sol et la cristallisation d'halloysite, de kaolinite et de gibbsite à partir de cette solution. Les grains de feldspar fort altérés consistent en des fragments gravés de feldspar enfoncés au sein d'une matrice très poreuse de cristaux sousédraux à euédraux de kaolinite et de gibbsite, ou d'aggrégats sphériques et feutrés d'halloysite. [D.J.]

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