

Weathering mineral facies in altered granites: The importance of local small-scale equilibria

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SUMMARY. Classical clay mineralogy determinations and electron microprobe analyses of weathering minerals developed in altered two-mica granites indicate that the chemical forces that produce new minerals are often constrained to small volumes, frequently on the scale of a mineral grain or contact between two grains in the granite.

Chemical potentials such as pH, alkali and alkaline earth and silica activity in the altering aqueous solutions provoke a destabilization of pre-existing minerals, which recrystallize locally to give a new multiminerall product. The chemical composition of the new phases is largely governed by the relative concentrations of the elements present in the former minerals.

Three mineral facies were observed in the weathered granites: initially a sericite-beidellitic type, then a beidellite-kaolinitic type, and finally a last stage kaolinite-oxide facies assemblage. The position of each facies is not restricted to a given depth in the profile but the relative proportions of each facies found in a thin-section size sample change towards the kaolinite-oxide facies.

The global rock chemistry reflects the type facies predominant in each sample. The first two facies are roughly silica conservative while the kaolinite-oxide facies loses silica as well as alkali and alkaline earths.

Geochemical and clay mineral studies of rock alteration should consider problems of mineral genesis at very localized sites.

A DETAILED study of the alteration products formed in the initial stages of granite weathering should provide some answers to the questions that must precede the general geochemical studies currently employed to study the interaction of granites with meteoric water (Dejou, 1958; Grant, 1963; Seddoh, 1973). A combination of classical methods (the petrographic microscope) and their modern extension (the electron microprobe) allows the investigation of authigenic minerals in their petrographic context. The problem of determining the

scope of chemical reaction (i.e. the scale of the chemical system concerned in terms of reacting species and their relative masses) must be posed once it has been ascertained exactly where new minerals are formed in the rock and exactly what the species are. The study reported here has been oriented towards a determination of the chemical or mineral elements that are active in producing weathering minerals.

The material studied forms two weathering profiles, from rock to upper horizon, developed upon a two-mica alkali granite (Meunier, 1977). In one profile, La Rayrie, the granite is slightly tectonized, the other one, La Pagerie, is overlain by a thin layer of calcareous sediments, which are now largely gone, but which still provide significant concentrations of calcium to the percolating waters that react with the granite. The character of the chemistry of the initial aqueous solution determines, to a certain extent, the subsequent mineralogy produced during weathering but the tectonization does not seem to be important. Both profiles show a distinct structure (fig. 1), which can be attributed to the physical fracturing of the initially isotropic rock. Fissures are formed in which the alteration process is more rapid than in the rest of the rock. These cracks and their alteration halo extend to the lowest levels of the profiles. The weathering mineralogy is similar in all of these zones.

The more massive sections of the profiles can be separated into two parts—that where the granite mineralogy can be identified and that where mineral boundaries are totally obliterated and the granite mineralogy has become totally unstable. Here quartz grains are identified but they appear to be less abundant than in the initial rock (26% vs.

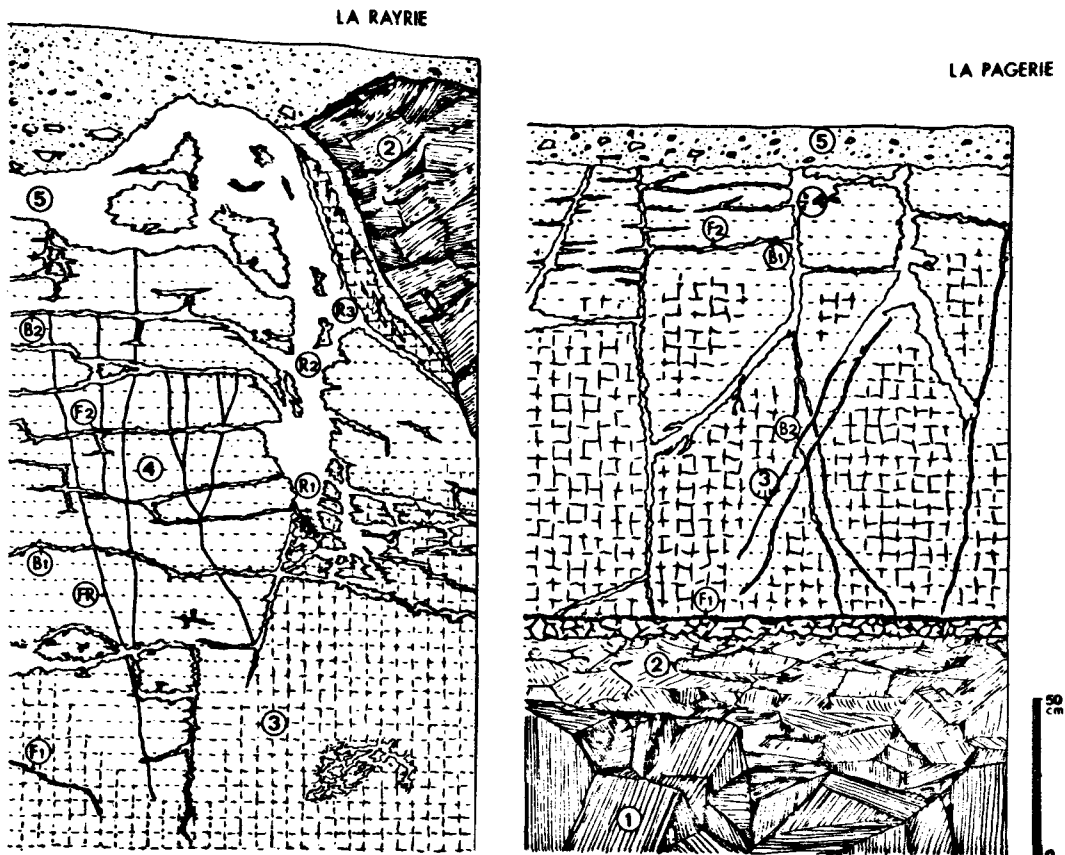


FIG. 1. Representation of the weathering profiles developed upon the granite of Parthenay (SE of Massif Armoricain, France). *La Rayrie profile* (left): (1) 'unweathered' rock; (2) compact weathering granite; (3) red coloured saprolite; (4) rose coloured saprolite; (5) white coloured saprolite. B₁-B₂: white bands; R₁-R₂-R₃: root surroundings; F₁-F₂: clay concentrations in fissures; FR: subvertical rusty band. *La Pagerie profile* (right): (1) unweathered rock; (2) compact weathered granite; (3) first saprolite level (r: red, b: white); (4) second saprolite level (r: red, b: white); (5) acid-brown soil. F₁-F₂: clay concentrations in fissures; B₁-B₂: white bands.

32% by modal analysis). One can thus distinguish three types of rock-alteration zones in these profiles: fissural, friable rock, and coherent rock.

The major initial minerals in the rock are: quartz, K-feldspar (almost always orthoclase), acid plagioclase (An₂₅₋₃₀), muscovite, and minor sericite and biotite (see Table I).

Observations on weathering phases

Fissures. In all cases the fissural reaction minerals are iron oxide and kaolinite. All pre-existing phases, whether belonging to the granite or to weathering facies, are converted to the kaolinite-oxide assemblage. One also finds a strong concentration of transported minerals in these zones, most often in the form of cutanes but also in a more

massive (on the microscopic scale) layer structure in the case of horizontal fissures (D in fig. 2). In all points investigated by microprobe analysis, this material had an Si/Al ratio of very near one—that of kaolinite. Thus the greatest part of the transported clays in the profile are kaolinites. Also, since the fissural zones are those of the greatest or at least most rapid passage for percolating water, the solutions that will be most likely to be collected from these profiles as altering fluids will have ionic activities compatible with kaolinite (Feth *et al.*, 1964; Tardy *et al.*, 1973). However, these fissural zones are not the only sectors where clay minerals are formed and they in fact represent but 3-4% of the column of the profile.

Coherent rocks. This zone (level 2 in fig. 1A and 1B) represents the initial stages of mineral trans-

TABLE I. *Chemical composition of the principal mineralogical species identified in the two weathering profiles*

(Microprobe analysis expressed in atoms per 22 oxygens. n.a.: 'not altered'; Verm. di.: dioctahedral vermiculite; Verm. tr.: trioctahedral vermiculite; M.L. (m-v): regular mixed-layered mica-vermiculite.)

La Pagerie											
	Biotite		Muscovite		Orthoclase		Plagioclase		Fissure	Sericite in	Sericite in
	n.a.	M.L. (m.v.)	n.a.	kaolin-ite	n.a.	Beidel-ite	n.a.	Kaolin-ite	cutane	plagio- -clase	ortho- -clase
Si	5.432	5.972	6.080	6.187	8.112	6.941	8.159	5.330	5.965	6.158	6.282
Al	3.618	3.914	5.142	5.987	2.840	3.160	2.962	5.462	5.721	5.500	4.712
Fe	3.060	2.577	0.503	0.189	0.007	0.072	0.023	2.787	0.673	0.262	0.648
Ti	0.212	—	0.019	—	0.004	—	—	—	0.086	—	0.016
Mg	1.316	1.022	0.562	0.110	0.112	0.281	—	0.101	0.323	0.228	0.728
Ca	0.008	0.086	0.021	—	0.022	0.079	0.245	0.041	0.101	0.012	0.030
Na	0.112	—	0.018	0.079	0.186	0.418	2.033	0.157	0.169	0.112	0.116
K	1.692	1.002	1.842	0.639	2.530	0.321	0.019	0.274	0.269	1.754	1.748

La Rayrie												
	Biotite		Muscovite		Orthoclase		Plagioclase		Fissure	Sericite	Reac- tional mica	
	n.a.	Bio- tite ₂	Verm. tr.	n.a.	Verm. di.	n.a.	Verm. di.	n.a.	Kaolin- ite	cutane	tecto- nisme	Mus- Ortho.
Si	5.502	5.548	6.747	6.141	6.640	8.223	6.265	8.098	6.673	5.965	6.266	6.464
Al	3.322	3.984	4.945	3.462	4.941	2.847	5.855	2.969	5.219	5.721	5.336	5.203
Fe	3.372	3.546	0.319	0.303	0.182	0.006	0.344	0.012	0.550	0.673	0.206	0.270
Ti	0.204	—	0.116	—	0.059	—	—	—	—	0.086	0.006	—
Mg	1.346	0.786	0.243	0.240	0.141	—	0.132	0.051	0.141	0.323	0.196	0.066
Ca	0.010	—	0.008	—	0.012	—	0.017	0.247	0.098	0.101	0.008	—
Na	0.144	—	0.030	0.163	0.163	0.162	—	2.098	0.074	0.169	0.242	—
K	1.612	1.194	0.944	1.819	1.324	2.393	0.391	0.011	—	0.269	1.838	1.862

formation by the weathering process. New phases are found in two situations: those where two mineral grains react to form a new mineral (B in fig. 2) and those at grain boundaries where the phases form two parallel zones each with a distinct mineralogy. The chemical character of the sericite produced by the reaction of muscovite and orthoclase has been previously reported (Meunier and Velde, 1976). It can be most closely compared to an illite.

The mineralogy of the parallel reaction zones at mineral contacts is in fact the same for each species as that found internally for each mineral in the friable rock zone and will be described below. One remark should be made concerning quartz. It is apparent that this mineral inhibits reaction. Contacts between quartz and all other minerals show no new phases to be present. This is also true in this zone for orthoclase-plagioclase grain contacts, which show no weathering minerals.

Friable rock. In this zone all minerals except quartz show internal destabilization, forming, in

most cases, mineral assemblages, i.e. multiphase products (C in fig. 2). This point must be emphasized since it is very important to a description of the type of chemical system active in the rock during weathering (Table II).

TABLE II. *Secondary phases produced for each initial granite mineral in the two profiles (identified by optical properties, X-ray diffraction and thermal analysis)*

	La Rayrie	La Pagerie
Biotite ₁	vermiculitic intergrade + kaolinite + oxides + biotite ₂	regular (1:1) interstratified
Muscovite	vermiculite intergrade (dioctahedral) + kaolinite	kaolinite
Orthoclase	vermiculite intergrade (dioctahedral) + kaolinite	beidellite → kaolinite
Plagioclase	kaolinite	kaolinite

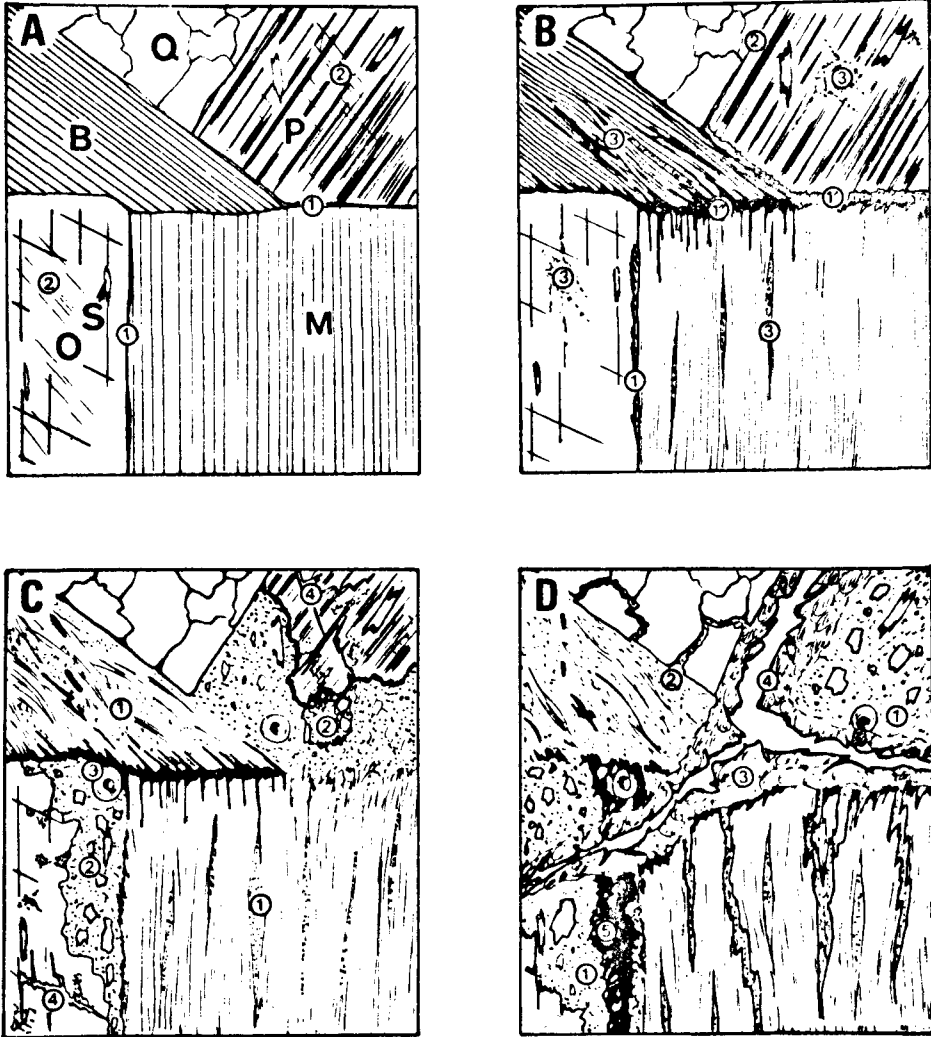


FIG. 2. Principal steps of the granite transformation during weathering. A: *Unweathered rock*: (B) biotite; Q: quartz; P: plagioclase; O: orthoclase; M: muscovite; S: sericite. (1) widening of the joints; (2) microfissuration. B: *Development of the contact microsystem*: (1) reactional micas; (1') simultaneous destabilization; (1'') diffusion (1-1'-1''); contact reactions; (2) inert contact; (3) beginning of the internal destabilization of primary minerals. C: *Development of the plasmic system*: (1) internal destabilization; (2) plasma; (3) argillane; (4) fissure. D: *Development of the fissural system*: (1) plasma; (2) fissures; (3) clay concentration; (4) recent clay deposit; (5) subvertical rusty band.

The morphological terminology for these transformations is that of a heterogeneous plasma (Bisdorn, 1967; Boulet, 1974). It is important to remember that although one cannot identify the pre-existing mineral of the granite at this stage of weathering, the pattern of weathering minerals nevertheless remains, i.e. the initial chemical system is maintained and no significant homogenization of the phases is discerned. Thus the mineral reactions

in this zone depend upon the chemistry of the initial phase and that of the percolating fluid since one can see a difference in the reaction products between the two profiles for the same reacting mineral. This is particularly striking for the biotites, where calcic solutions result in the formation of a regular mixed layered biotite-vermiculite mineral, which is found at all levels in the profile (Meunier, 1977). This influence of calcium has been demonstrated by

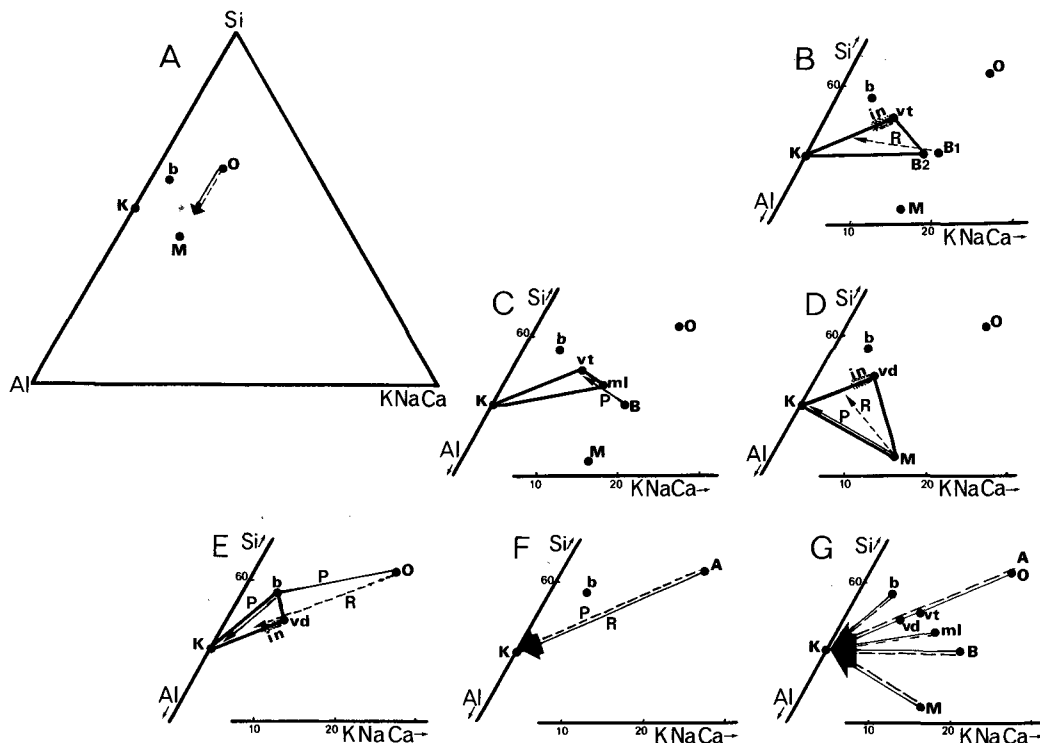


FIG. 3. Chemigraphic representation of the mineralogical transformations of primary and secondary minerals during weathering in the Si-Al-K, Na system. (O: orthoclase; A: albite; M: muscovite; Q: quartz; B: biotite; m-l: mixed-layered mica-vermiculite; Vt: trioctahedral vermiculite; Vd: dioctahedral vermiculite; in: intergrade minerals; K: kaolinite). A: muscovite-orthoclase reaction; B, C, D, E, F internal destabilization in the plasmic system of La Rayrie biotites, La Pagerie biotites, muscovites, orthoclases, and plagioclases; G: primary and secondary minerals destabilization in the fissural system.

Hoda and Hood (1972). In the case of La Rayrie, the reaction $\text{biotite}_1 \rightarrow \text{biotite}_2 + \text{vermiculite} + \text{kaolinite}$ was observed. The new biotite composition can be found in Table I.

Chemical description of the alteration process

Fig 3A to 3B represents the granite phases and the weathering phases as they plot in Si-Al-(Na,K,Ca) coordinates. The average of the granite phases that are reactive, feldspars and muscovite, lies between these phases in this plot. The final assemblage for each weathering type—fissural, friable rock, and coherent rock—is represented by the letter P for La Pagerie profile and R for La Rayrie. Microprobe analyses of the phases are taken from Meunier (1977) and are presented in Table I.

It can be seen that the new phases or phase assemblages are defined by the kaolinite-beidelite-vermiculite-intergrade-mica association. If we

consider that the aggregate reacting phases plot between feldspar and muscovite, the weathering phase assemblages represent bulk compositions of equivalent silica content but of much less alkali + alkaline earth content. There is a relatively much greater loss of sodium than potassium in the friable and coherent rock zones. The effect of calcic altering solutions at times favours silica retention (biotite and orthoclase alteration) or the reverse (muscovite alteration). Calcium seems to inhibit muscovite destabilization; in fact, the extent of recrystallization is less in La Pagerie than in La Rayrie for an equivalent position in the profile.

It is only in the more intense weathering zone of the profile (the fissures) that the silica content does decrease in a given rock column, undoubtedly due to dissolution of quartz. This occurs mainly in the presence of kaolinite. The general evolution of the whole rock composition (fig. 4) shows an initial alkali-loss trend, then a silica loss, as one goes from the coherent rock to friable rock to fissural zones.

Weathering mineral facies

We have distinguished three weathering zones on the basis of their mineral textures in thin section and in the outcrop aspect. Each zone represents a type of physico-chemical system that transforms the initial granite minerals.

Coherent rock. In this zone water evidently circulates at grain boundaries and effects mineral reaction there. The most spectacular and initial effect is the growth of illite-like mica into orthoclase at muscovite-orthoclase and some biotite-orthoclase contacts. A certain amount of reaction occurs at other grain boundaries and this increases upwards in the profile. In this zone we see the destabilization products of the minerals, which invade the grains from their edges inward. However, the mass of altered material is relatively small, globally, the new mineral facies is represented by 2:1 phyllosilicates—illite, beidellite, and vermiculites, both di- and trioctahedral (fig. 3B–F). These minerals form a bisiallitic process (Pedro, 1964). The new minerals form a general line between beidellite and muscovite in fig. 3.

Friable rock. Internal destabilization, i.e. no apparent increase from the exterior or any one point within the mineral, is the phenomenon that dominates this portion of the profile. It seems apparent that at this level in the profile the altering solutions are dilute enough to provoke the partial dissolution of the granite minerals. It also seems apparent that these solutions traverse the minerals at a great many points. They most likely do so also at lower levels in the profile but their flow rate

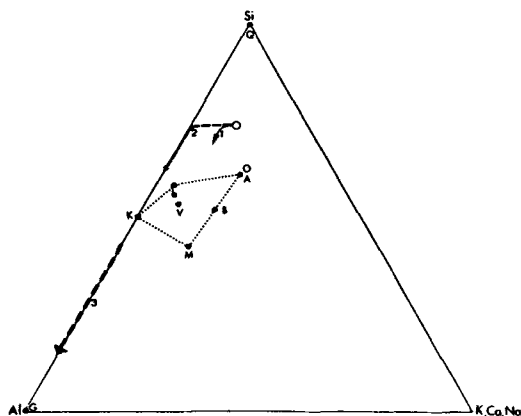


FIG. 4. Representation of the bulk evolution of weathering granites. (Q: quartz; A: albite; O: orthoclase; B: biotite; M: muscovite; K: kaolinite; b: beidellite; V: vermiculite; G: gibbsite). (1) weathering in temperate climate (Dejou, 1958; Seddoh, 1973; Meunier 1977); (2) weathering in tropical climates (Lelong, 1969; Leneuf, 1959); (3) Bauxites (Novikoff, 1974).

would be smaller than at the grain boundaries and hence they would contain more material in solution and hence would be closer to a chemical equilibrium for each mineral they slowly traverse.

The grain boundary solutions, flowing more rapidly, would not come into equilibrium with the minerals but instead they will effect an initial destabilization reaction.

The weathering mineralogy of this friable rock zone includes in most cases kaolinite-bearing assemblages for each altered original mineral; Jackson (1965) described the intergrade minerals as a transition toward kaolinite. The plagioclase is transformed into a monomineralic assemblage. The bulk composition of the altering granite moves further toward the alkali-free side of fig. 4.

It should be said here that although some minerals are present in this zone simply because they have not reacted to new chemical conditions, the regular interlayered biotite-vermiculite mineral remains present throughout the zone even in the $< 2 \mu\text{m}$ grain size fraction (Meunier, 1975). This suggests that the trioctahedral mica portion of the structure remains stable in a fairly advanced stage of weathering.

Fissures. These zones, found at all levels in the profile, represent total transformation of all pre-existing minerals, even many of those produced during weathering. Kaolinite plus oxides are the final products and even the quartz, which is largely inert in the rest of the profile, becomes subject to a destabilization process. Fig. 3G shows this evolution mineralogically. Also, these very porous zones contain much transported clay material, mineralogy of which is virtually that of the entire fissure. These zones represent the most intense weathering in the profile.

Conclusions

In summary, the initial weathering in the lowest-massive levels of the profile produces first illitic mica, then beidellite and vermiculites. The greater loss of sodium relative to potassium is notable. This might be called the mica-beidellite facies.

The second stage (friable rock) produces kaolinite-bearing assemblages in each of the decomposing minerals. One should note the predominance of multiphase assemblages in the rock. This might be called the kaolinite-beidellite facies. Then the final weathering type is that where only kaolinite is present and even the quartz begins to be dissolved in significant quantities. This is the kaolinite-oxide facies.

Now let us consider these observations in a chemical framework. First, it is evident that the initial weathering is effected through alkali loss.

Both alumina and silica remain present in approximately constant proportions. In such circumstances the chemical system appears to be one where alkalis are mobile but Al and Si are immobile at least on the scale of a thin section. Calcium does not greatly affect these trends but it can provoke the crystallization of slightly different mineral assemblages. Since the process of weathering is that of hydration, one could represent these stages of weathering as functions of mobile H^+ and K^+ , Na^+ ions in a two-inert-component system, Al_2O_3 - SiO_2 , at constant pressure and temperature.

However, from the transformation of each mineral—muscovite, orthoclase, biotite—it is evident that, even though multiphase assemblages are produced upon weathering in each mineral grain, there can be a variation in the Si/Al ratio of new phases, which is controlled by the presence of calcium and could be modified by other elements in solution. Thus, on a grain-to-grain scale, either silica or alumina is displaced. If the use of a three-component representation is pertinent to the granite minerals, the common formation of two new phases in replacing a previous mineral indicates that two elements are often acting as immobile components, to a first approximation. Supposing that silica migrates, it is necessary to assume that its activity is 'buffered' on a grain-to-grain scale as, for example, muscovite is transformed to kaolinite plus aluminous vermiculite. It is likely that one must consider the mineral pair orthoclase-muscovite to equilibrate the silica content of the weathering minerals formed from each crystal species. Since reactional gradients are rather rare, it is necessary to consider that silica activity is equivalent throughout the mineral grain—a mobile component or intensive parameter of the system in which each grain is contained. However, on the scale of several grains, or perhaps the thin section, the over-all balance of Si/Al is little changed and the assemblages taken for the several grain types (muscovite, orthoclase, etc.) represent a system more or less closed to Si-Al migration in the first two weathering mineral facies described above.

It is very important to remember that the weathering mineral assemblage formed from each mineral grain remains coherent in space—i.e. there is little tendency towards a chemical homogenization of the separate parts of the altered and altering rock. This indicates that the limits of the 'grain' system are respected even when the initial alteration process is completed. Each original mineral grain is part of a mosaic of chemical systems (Thompson, 1959), which themselves buffer at least two of the chemical components.

The fissural alteration facies represents the stage where the silicates no longer buffer silica in solution to maintain their stability. Both alkali and silica loss become apparent and the only weathering minerals that resist are kaolinite and oxides. The only other stage of weathering would be that where even quartz no longer persists and one will find only oxides stable, excluding silica. This stage is well developed in tropical countries (Delvigne, 1965; Lelong, 1969; Leneuf, 1959; Novikoff, 1974). Because the weathering profiles studied here are truncated, it is not possible to determine if an extension of the fissural system towards the surface would continue into the soil which would contain only kaolinite as a weathering silicate.

Fig. 4 shows bulk chemical analyses of the different sections in the profiles studied as they plot in Si-Al-(Na,K,Ca) coordinates. It is clear that the first stages of alteration in the profile show alkali loss at roughly constant Si/Al ratios. This shows that the first two weathering facies—muscovite-beidellite and beidellite-kaolinite—are predominant in the sample analysed. The fissural zones show decreasing Si content indicating the existence of the kaolinite-oxide facies. The bulk composition of each horizon in the weathered granite then reflects the relative importance of each alteration facies as it is developed in a specific sample. The first alteration processes are more or less silica-conservative in aggregate and the last or most intensive process shows silica loss. However, at each level in the horizon, the three facies are represented and thus the global clay mineralogy represents the products of several alteration facies. It is clear then that analysis of clay minerals and dissolved components in water circulation through an altering rock must consider the processes that are taking place in each section of the sample in a microscopic scale. Global geochemical descriptions of an alteration process necessarily average the effects produced by each micro-system that produces weathering minerals.

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