# INTERLAYERING OF EXPANSIBLE LAYER SILICATES IN SOILS BY CHEMICAL WEATHERING

*by*

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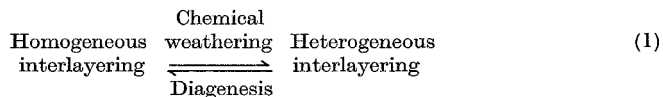
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## ABSTRACT

Interlayering of 2: 1 layer silicates varies as a function of chemical weathering from the simple, homogeneous K or Na interlayers of micas to the heterogeneous systems of mica intercalated with expanded 2: 1 layer silicates. "Frayed edge" type of weathering at dislocation planes of mica is collated with K release and preferential cation-exchange uptake of K relative to Ca by such expansible layer silicate systems; mica islands maintain alignment of the silica sheet cavities, which facilitates recapture of lattice K. Intercalation of the expanded 2: 1 layer silicates with alumina interlayers appears to be a characteristic function of chemical weathering in soils, with the formation of 2: 1–2: 2 intergrades not only of 14 Å spacing but also of swelling 18 Å types that give small 12, 14, 18 Å and higher spacing peaks (along with the 10 Å peak) at 550°C. Interlayer precipitates appear to be characteristic of soil clays, contrasting with "pure" minerals of deposits developed in less "open" environments than those of soils. The "2: 2 lattice building" phenomenon in expansible 2: 1 layer silicates relates to layer charge density and crystal size, and frequently tends to inhibit the formation of free gibbsite in soil chemical weathering so long as there are expansible layer silicates present to become intercalated with aluminum hydroxide—a weathering phenomenon that may be called an "antigibbsite effect". Accumulation of alumina (possibly with some iron, magnesium, and allophane) as interlayers in 2: 1 minerals of soils is seen as a genetic stage in the 2: 2 → 1: 1 weathering sequence through which kaolinite and halloysite develop in soils.

## INTRODUCTION

Homogeneous or monospecies interlayers in layer silicates consist of intercalated sheets such as K in micas. Other examples are the water (with more or less of exchangeable cations) in vermiculite or montmorillonite or halloysite, and the hydroxide sheets of chlorite. (The dictionary definition of *intercalation* covers any class of insertion between other layers, such as intrusives between sedimentary beds or interlayering of water in crystals.) Chemical weathering reactions induce heterogeneous intercalations of layer silicate crystals:

$$\begin{array}{c} \text{Chemical} \\ \text{Homogeneous} \quad \text{weathering} \quad \text{Heterogeneous} \\ \text{interlayering} \quad \xrightleftharpoons{\phantom{xxx}} \quad \text{interlayering} \\ \text{Diagenesis} \end{array} \qquad (1)$$

The present paper considers the types of heterogeneous intercalations that

29

arise through chemical weathering of layer silicates in surficial deposits of soils and mantle rocks.

## INTERLAYERING BY MICA CLEAVAGE

Several types of simple, homogeneous interlayers occur in layer silicates. Examples are K in muscovite or biotite or Na in paragonite. Homogeneous interlayers of water without cations were first proposed for montmorillonite (Hofmann and others, 1933) and for vermiculite (Gruner, 1935). Interlayer cation exchange capacity was then proposed for montmorillonite (Marshall, 1935) and for vermiculite (Barshad, 1948).
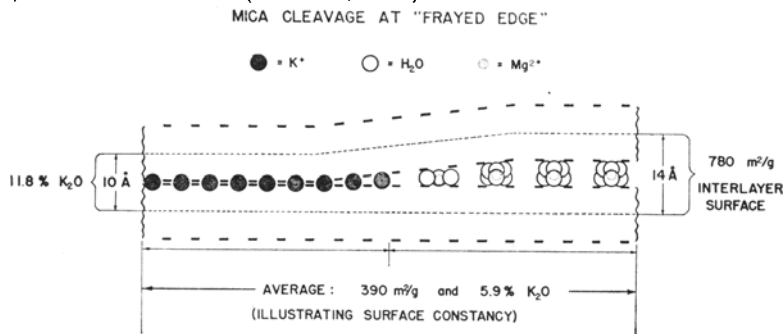


FIGURE 1.—"Frayed edge" type of interlayering caused by mica weathering, an example with 50 per cent expanded interlays and 50 per cent residual mica interlays. The analytical data illustrate the tendency for interlayer specific surface constancy.

One may imagine a perfect mica crystal becoming exposed to weathering by a rock fracture on a Precambrian mountain in central Wisconsin (or elsewhere). Even with no further physical weathering following the fracture, chemical weathering soon begins if water is present. Interlayer K is subjected to depletion through chemical equilibrium with the soil solution and gain in replacing ions and water, according to the equation of Jackson and others (1952):

$$\text{Mica} \rightleftarrows \text{illite} \rightleftarrows \text{vermiculite} \rightleftarrows \text{montmorillonite} \qquad (2)$$
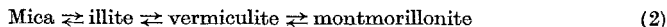
This equation has been amplified to include interlayer "2:2 lattice building" (discussed in next section). The role of low K concentration in shifting equation (2) to the right (to give expansion) has been demonstrated by K precipitation with cobaltinitrite (White, 1951) and tetraphenylboron (De-Mumbrum, 1959). Shift to the right has been accomplished by replacement with strongly sorbed alkyl ammonium ions (Weiss, Mehler, and Hofmann, 1956). It is proposed that mica cleavage and expansion (Barshad, 1948; Walker, 1949; Jackson and others, 1952; Mehra and Jackson, 1959) represented by the arrows pointed to the right in equation (2), occurs at frayed edges (Bray, 1937; MacEwan, 1949) as shown in Fig. 1 and develops into interstratification (Hendricks and Teller, 1942) as shown in Fig. 2 when there are

large cleavage rate differentials (Jackson and others, 1952). A high correlation was found between the total of the specific surface of edges in soils and the equilibrium level of exchangeable potassium in soils (Milford and Jackson, 1962). The cleavage is visualized at mica layer dislocation edges as well as the particle edges. The characteristic mottled appearance in electron micrographs of weathering mica particles (Fig. 2 in Jackson, Whittig and Pennington,
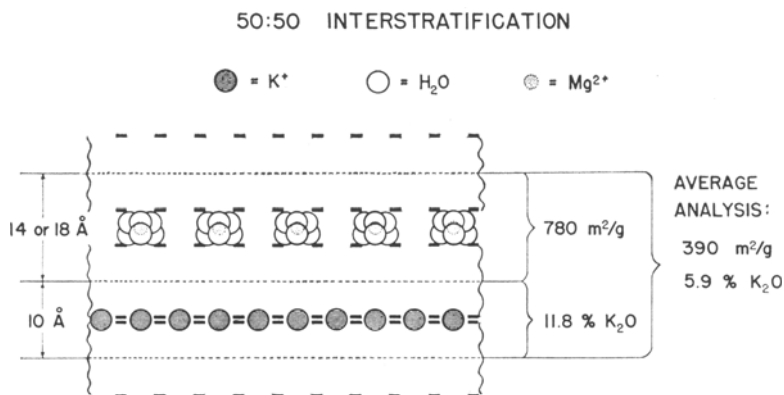


FIGURE 2.—Interstratification type of interlayering caused by mica weathering, illustrated by an example of 50 per cent mica interlayers and 50 per cent expanded interlayers. The ¦analytical ¸data illustrate the tendency for interlayers specific surface constancy.

1950) is visualized as equation (2) applied at the various crystallographic dislocations on mica crystal faces.

Such cleavages of mica produce a heterogeneous population of interlayers, composed of residual K interlayers and cation-water interlayers (Figs. 1 and 2). Further interlayer heterogeneity is implicit in the proposal of hydronium as a replacing cation for K without expansion (Brown and Norrish, 1952; Harrison and Murray, 1959)—possibly a result of diagenesis or "closed" environment. The tendency (shown in Figs. 1 and 2) of interlayer K-surface constancy of mica–vermiculite (Mehra and Jackson, 1959) suggests that under fairly intensive weathering conditions any given mica interlayer segment is completely filled with K or else completely available for interlayer swelling.

The interstratification type of heterogeneity (Z-axis disorder) was quickly recognized as a reality in soil clays (Alexander, Hendricks, and Nelson, 1939). On the basis of a survey of soil clays from diverse sources and weathering conditions, the general occurrence of interstratification was proposed (Jackson and others, 1954) and its general occurrence in sediments was demonstrated through thousands of analyses by Weaver (1956).

The model of weathering of micas to expanded layer silicates, summarized in equation (2), appears to have become widely accepted (Barshad, 1948; Walker, 1949; Murray and Leininger, 1956; Whittig and Jackson, 1956;

Brown and Jackson, 1958; Weaver, 1958; Hensel and White, 1960; Loughnan and others, 1962). The rate of weathering (equation 2) is greatly influenced by the permeability of rocks (Glass, 1958) and soils (Jackson, 1959). The slowly permeable soils from shale-derived till of Wisconsinan age have remained illitic (Dixon and Jackson, 1960), in contrast to the permeable soils derived from loess (Glenn and others, 1960), limestone and sandstone in southwestern Wisconsin in which micas have weathered to montmorillonite and mixed layer mineral assemblages high in montmorillonite.

Reversibility of equation (2), was suggested by potassium fixation in layer silicates (Truog and Jones, 1938; Page and Baver, 1940; Wear and White, 1951; van der Marel, 1954; Weaver, 1958). Preferential sorption of K in certain collapsing interlayers adjacent to more highly charged layers was presented (Jackson and others, 1952) and further indicated by specific surface measurements (Dyal and Hendricks, 1952). The full mica K content would not be resorbed by K fixation in layers of lower than mica charge; however, diagenesis in deep burial may accomplish complete reversal to mica. Drying with potassium silicate solution resulted in tighter collapse of expansible silicate than with simple K saturation (Mortland and Gieseking, 1951).

Preferential adsorption of K over Ca in cation exchange from dilute solutions containing both ions increases with montmorillonite relationship to mica origin (Schwertmann, 1962a, 1962b) as indicated by geologic source and $BO_3$ in sediments (Frederickson and Reynolds, 1960). Table 1 summarizes Schwertmann's data.

TABLE 1.—EXCHANGEABLE POTASSIUM SELECTIVITY AS A FUNCTION OF ORIGIN OF CLAYS
(AFTER SCHWERTMANN, 1962a, 1962b)

| Montmorillonite source | $K^+$ selectivity* $(k \cdot 10^2)$ | $B_2O_3$ % |
|---|---|---|
| Wyoming bentonite, $-2\mu$ | 3 | 0·03 |
| Marsh soil $-0·08\mu$ | 11 | 0·11 |
| Alluvial soil $-0·08\mu$ | 13 | 0·05 |
| Loessial soil $-0·08\mu$ | 14 | 0·08 |
| Ground muscovite $-2\mu$ | 23 | — |

* Gapon equation:

$$\frac{K^+ \text{ sorbed}}{Ca^{++} \text{ sorbed}} = k \quad . \quad \frac{K^+ \text{ solution}}{\sqrt{Ca^{++} \text{ solution}}}$$

Sorbed K and Ca are expressed as meg. per unit weight of clay, and K and Ca in solution are expressed in millimoles per liter. In the equilibration solution, meg. of K = meq. Ca and the total concentration was 0·01 N.

It is proposed that the K preference in cation exchange may occur by K "lattice building" in the wedge-zone between weathered and unweathered

portions (Fig. 1), because of $a$- and $b$-axis alignments of the cavities in layers held in proper orientation by unweathered mica cores (left side of Fig. 1) to provide juxtaposition of K coordinating cavities in the upper and lower silica sheets. Structural K "lattice building" inherent in mica layer structural relations (Radoslovich, 1960) may also contribute to K preference in cation exchange. The cation exchange preference for K would be a consequence of "frayed edge" heterogeneity in the XY (or ab) planes of weathered layer silicates. Residual nonexchangeable K is nearly always found with montmorillonite (Mehra and Jackson, 1959) and may indicate unweathered mica cores of small magnitude (left, Fig. 1), although it could also occur as a few unexpanded interstratified layers (Fig. 2) as suggested earlier (Jackson and others, 1952). The presence of K does not result in a 10 Å diffraction peak when only small amounts are present with montmorillonite (Schmehl and Jackson, 1956).

## INTERLAYERING IN 2:1–2:2 INTERGRADES

Intergradient 2:1–2:2 layer silicates have interlayers incompletely filled with respect to the brucite or gibbsite interlayers. Most natural intergrades so far described have shown 14 Å diffraction spacings and have been termed "dioctahedral vermiculite" (Brown, 1953; Rich and Obenshain, 1955), "vermiculite" (Loughnan and others, 1962), or "chlorite-like" (Klages and White, 1957). The interlayers are heterogeneous with respect to islands of brucite (Grim and Johns, 1954) or gibbsite (Dixon and Jackson, 1962, their Fig. 3) distributed in interlayers otherwise filled with water and exchangeable cations as characteristic of vermiculite or montmorillonite. Intergradient in properties between 2:1 and 2:2 layer silicates, these materials have appropriately been termed "2:1–2:2 intergrades" (Dixon and Jackson, 1959; Jackson, 1962). When stripped by extraction of the interlayer islands, they may show either vermiculite (Rich, 1960; Sawhney, 1960) or montmorillonite spacings (Tamura, 1957; Dixon and Jackson, 1959).

*Degrading chlorite weathering mechanism:* Weathering of chlorite to 2:2–2:1 intergrades has been suggested through acid removal of Fe from the (Fe, Al, Mg) interlayer (Harrison and Murray, 1959) and by replacement of OH by $OH_2$ (Droste, 1956). A lacy residuum of aluminum (and possibly iron) hydroxide interlayers (Dixon and Jackson, 1962) would remain with "holes" (internal edges) if Mg were selectively removed by weathering. The chemistry of layer edges and these "holes" has been discussed elsewhere (Jackson, 1960, 1963).

*Swelling 2:1–2:2 intergrades:* Stephen and MacEwan (1951) described "swelling chlorite" as having one surface of the brucite layer unattached to a silicate layer; the interlayer positions thus would be heterogeneous with respect to brucite and water-cation layers. The occurrence of such interlayering in montmorillonite of natural soil clays was indicated (Sawhney and Jackson, 1958) in a Queensland, Australia, soil clay weathered from basalt.

A sharp 18 Å peak with Mg saturation and glycerol solvation, and a greatly broadened 10 Å peak with K saturation and 550°C heating, characterize the expanding montmorillonite–chlorite intergrade type of clay. Extensive occurrences of swelling interlayered soil clays have now been observed (the one represented in Fig. 3 is representative of many, to be reported separately,
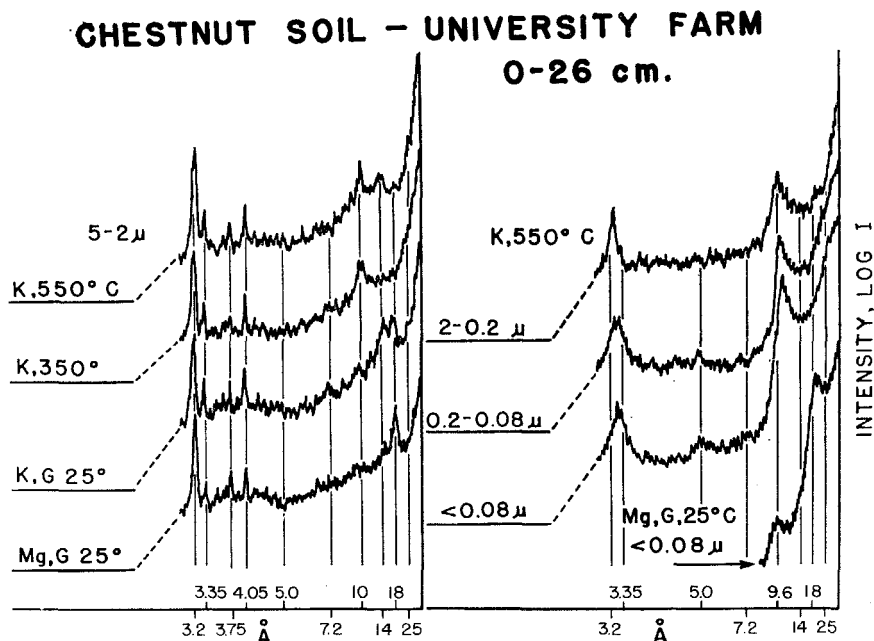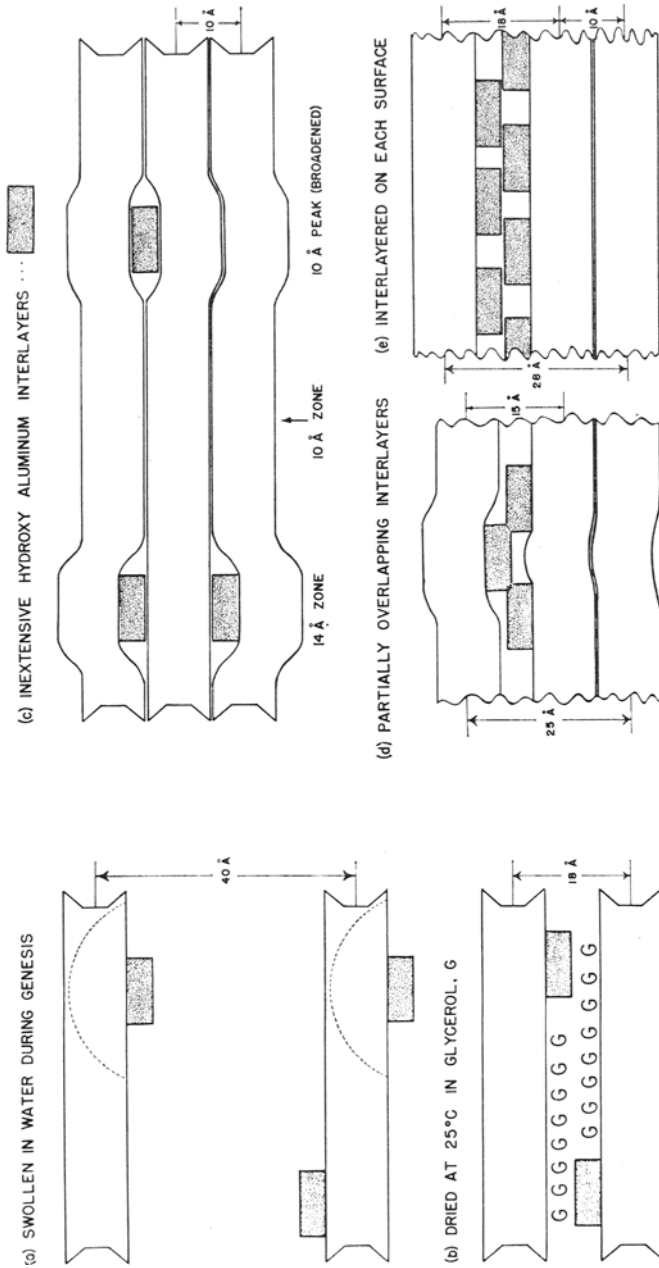


FIGURE 3.—X-ray diffractograms of soil-derived swelling 2: 1–2: 2 intergradient montmorillonite–chlorite. This alluvial soil occurs on Ataturk university Farm, Erzurum, Turkey (from Wisconsin studies with Dr. O. Baykan). Some feldspar is also present. The initial 18 Å spacings with Mg and glycerol indicate a montmorillonite-like expansion. The 12, 14, 18, and 24–28 Å enhanced spacings after heating to 550°C indicate some hydroxy aluminum interlayering (of chlorite-like character depicted schematically in Fig. 4) in the montmorillonite (10 Å after heating).

Baykan and Jackson, in manuscript). The sharpness of the 18 Å peaks (Fig. 3) suggests swelling of the interlayer (Fig. 4,b) rather than interstratification of 14 Å with 18 Å spacings. The greatly broadened 10 Å peak on heating (Sawhney and Jackson, 1958) is shown by the finer fractions in Fig. 3. Flexibility of the layers (Fig. 4,c) accommodates some interlayering without disturbing the main spacing. Even 14 Å peak reinforcement occurs in many samples, as is characteristic of a chlorite-like character in the portion of the sample more completely interlayered (fine silt and coarse clay, Fig. 3). When interlayering is still more complete, i.e. alumina on *both* surfaces of the swollen montmorillonite (Fig. 4,e), then collapse is only to 18 Å at 550°C (coarse clay and fine

FIGURE 4.—Schematic representation of swelling 2: 1–2 :2 intergradient montmorillonite-chlorite which could give the observed diffractograms (Fig. 3). The dotted lines in (a) and (b) suggest possible spheres of (—) countercharge effected by (+) charges of interlayers.

silt, Fig. 3). The genesis of the soil clay shown (Fig. 3) is in a slightly alkaline alluvial material, thought to be of basaltic origin because of the unusually high feldspar content and the near absence of quartz. In several soils examined, a little chlorite frequently occurs with the 2:1–2:2 intergrade clays as, for example, in the Chestnut soil of Iraq (Hanna and Jackson, in manuscript).

In alkaline soils of arid regions, montmorillonite is relatively stable to weathering and so is of great frequency distribution. Characteristically, it is partially interlayered. The weathering mechanism by which swelling intergradient 2:1–2:2 montmorillonite–chlorite is formed appears to be tied to the swelling character of montmorillonite (Fig. 4,a) which has spacings of 40 Å or more in water (Norrish, 1954). In a feldspar-rich mantle, alkalinity and hydrolysis yield a plentiful supply of freshly precipitated aluminum hydroxide, positive in charge at just under pH 8.3 (Jackson, 1963). Some excess silica is available for leaching as feldspars weather. The presence of much montmorillonite makes for moist conditions and as a result the clay in soil is kept swollen to 20 or 40 Å spacing. Intercalated gibbsite sheets thus can attach on only one layer so long as the layers are widely separated. Laboratory syntheses also have involved attachment of gibbsitic interlayers to one surface while the montmorillonite is in a swollen state (Slaughter and Milne, 1960). Gibbistic character of the interlayer has been indicated in synthetic preparations (Shen and Rich, 1962) in which the cation exchange capacity was considerably lowered. Restoration of the cation exchange capacity by HCl suggests that dealumination may occur where $OH_2$ substitutes for OH in gibbsitic sheets covering the (—) sites, as discussed elsewhere (Jackson, 1963).

The polymeric hexaluminohydroxyhydronium cation, $Al_6(OH)_{12}(—OH_2)_{12}^{6+}$, which has the OH/Al ratio = 2, has a one-surface coverage of approximately 100 Å$^2$, calculated from the $a = 8.62$ Å dimension of gibbsite (Bragg, 1937, p. 207) plus one oxygen diameter. This is equivalent to 16.6 Å$^2$ one-surface coverage per (+) charge. In comparison, $(a \times b) = 46.6$ Å$^2$ per (—) charge on one surface* of mica and 117 to 195 Å$^2$ per (—) charge of montmorillonite surface (Table 2). An excessive separation of (+) charge from (—) charge would be involved in one-surface attachment on montmorillonite of the 6+ hexameric ion or other acidic polymeric cations. Only 8 to 17 per cent coverage of the layer would completely neutralize the layer (Table 2). More extensive "lattice building" of gibbsitic sheets (higher OH/Al ratios, Jackson, 1960, p. 447) would be required for the (+) . . . (—) charge proximity (observed by Shen and Rich, 1962). With a limited amount of Al precipitation (Ragland and Coleman, 1960), little loss of cation exchange capacity occurs. The "lattice building" could occur with polar units of gibbsite of the nature, $Al_6(OH)_{12}(OH_2 . . . OH)_6$ in which the $(OH_2^{0.5+} . . . OH^{0.5-})$ pair represents a broken-edge pair, comparable to that proposed by Weiss (1958) for layer silicate edges, as discussed elsewhere (Jackson, 1963). A unit of this size

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* The $(a \times b)/2$ dimension for muscovite, $(5.18 \times 9.02)/2$ (Bragg, 1937, p. 20.5), gives an average of 7 Å separation of the (—) charge on each layer of mica given earlier (Jackson, 1960).

TABLE 2.—CALCULATED PERCENTAGE COVERAGE OF EXPANSIBLE LAYER SILICATES FOR CHARGE BALANCE BY THE POLYMERIC ION $Al_6(OH)_{12}(-OH_2)_{12}^{6+}$ OF AREA 99.5 Å², 16.6 Å² PER $(+)$, AND OH/Al RATIO $= 2$.

| Mineral | Charge density $(-)$ | | % area covered | |
|---|---|---|---|---|
| | Meq per 100 g | Area per Charge, Å² | One Surface* | Two Surfaces† |
| Muscovite | 251 | 46.6 | 35.6 | 71 |
| Vermiculite | 160 | 73.1 | 22.7 | 45 |
| Montmor.-Vermi. | 125 | 93.2 | 17.8 | 36 |
| Montmorillonite | 100 | 117 | 14.2 | 28 |
| Montmorillonite (low charge) | 60 | 195 | 8.5 | 17 |

* Swelling 2:1–2:2 intergrade (18 Å spacing in glycerol); if counter charge is effected through the layer, the "coverage" would be equivalent to that for two surfaces in next column.

† Nonswelling 2:1–2:2 intergrade (14 Å spacing in glycerol).

TABLE 3.—MOLARITY OF INTERLAYER SPACE IN EXPANSIBLE LAYER SILICATES

| Mineral | Interlayer Charge Density, on Two Surfaces | | Inter-charge Distance, One Surface Å | $c$ Spacing | | Volume per M+ | Molarity |
|---|---|---|---|---|---|---|---|
| | meq 100g | Å² $(-)$ | | Cell Å | Inter-layer Å | Å³ | |
| Muscovite (expanded) | 251 | 23.3 | 6.8 | 14 | 4 | 93.2 | 17.8 |
| Kaolinite-NH₄NO₃ | — | 23.3* | — | 11.6 | 4.4 | 102.5 | 16.2† |
| Kaolinite-KOAc | — | 23.3* | — | 14 | 7 | 163.1 | 10.2† |
| Vermiculite | 160 | 36.6 | 8.6 | 14.7 | 4.7 | 172 | 9.7 |
| Montmorillonite | 100 | 58.5 | 10.8 | 18 | 8 | 468 | 3.5 |

* Å² per silica sheet cavity.

† Salt concentration when intersalated; cavity contains K+ or NH₄+ and space is occupied by anion.

could sorb on the surface of montmorillonite between the $(-)$ exchange sites, which are approximately 11Å apart (Table 3). A few $(+)$ valence charges on the interlayer cation at unpaired edge sites could attach to $(-)$ exchange sites.

*Weathering processes forming* 2:1–2:2 *intergrades of* 14 Å *spacing:* Mobilization of layer cations, particularly aluminum from layer edges occurs in acid soils through the bonding of protons in silicic acid edges as outlined previously (Jackson, 1960, 1963), the silicic acid ($pk_1 = 9.5$) acting as a "proton sink".

4

This is a key to driving the release of aluminohydronium ($pk_1 = 5$) from layer edges by $H_2CO_3$ ($pk_1 = 6.4$). The decomposition of clay by $H_2CO_3$ with dissolution of silica in excess of alumina (which precipitates) has been demonstrated for montmorillonite (Correns, 1961) and for kaolinite (Polzer, 1961). The exchange of H from Si–OH groups at edges of clays occurred in alcohol at pH 10.1 (Weiss, 1958), while the silica sheets dissolved in aqueous alkali. Release of octahedral Mg from acid clays was shown by Barshad (1960).
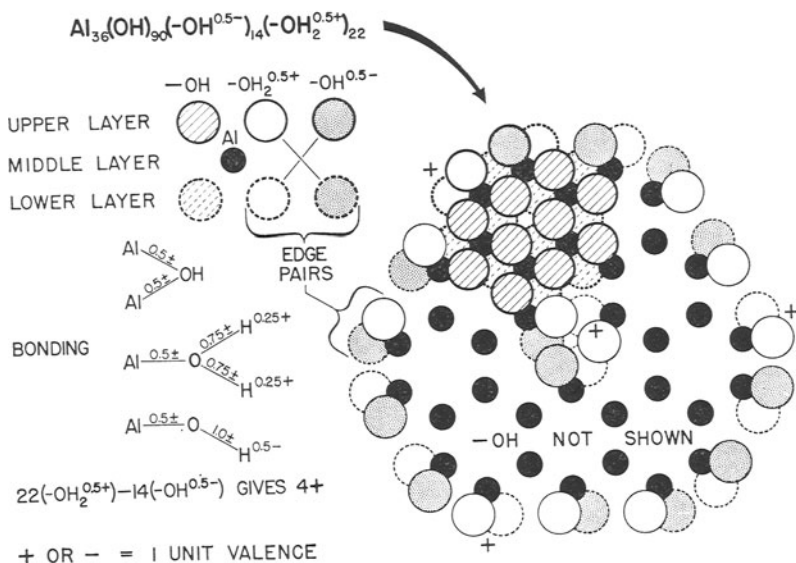


FIGURE 5.—Aluminum hydroxide, illustrating the mechanism of varying positive charge with varying number of hydroxyl and alumino-hydronium (OH ... OH₂ pairs, Jackson, 1962) at edges and in defects or "holes."

Polymerization of aluminum into hexaluminohydroxyhydronium units such as $Al_6(OH)_{12}(-OH_2)_{12}^{6+}$ (Jackson, 1963, Fig. 3) and larger, less highly charged units (Fig. 5) can occur in the vermiculite interlayer. Since vermiculite is characteristically spaced at 14 Å, the positively charged polymers can attach to both surfaces and the interlayer (+) charge proximity to (−) of layers is closer (Table 2) than for swelling intergrade. Less extensive interlayer "lattice building" in vermiculite is thus necessary than in montmorillonite for cation exchange capacity decrease. The "2:2 lattice-building" process is sterically assisted by cation exchange sorption in the vermiculite interlayer space because the charge concentration is on the order of 10 molar (Table 3).

Lattice building of interlayer $NH_4NO_3$ at a concentration on the order of 16M (Table 3) in kaolinite and dickite occurred from 6 to 9M solutions of $NH_4NO_3$ (Andrew and others, 1960).

Montmorillonite, characteristically of smaller crystal size and hence of greater specific edge surface, is attacked by weathering acids at a greater percentage rate than is vermiculite which is of larger particle size, freshly formed by mica cleavage (discussed in above section) and thus of lower specific edge surface, as pointed out (Jackson, 1960). The tendency for montmorillonite to become interlayered in acid weathering tends to be negated by relatively rapid weathering away of the small particles characteristic of this species (Fig. 6). An amorphous relic clay that exemplifies this model was

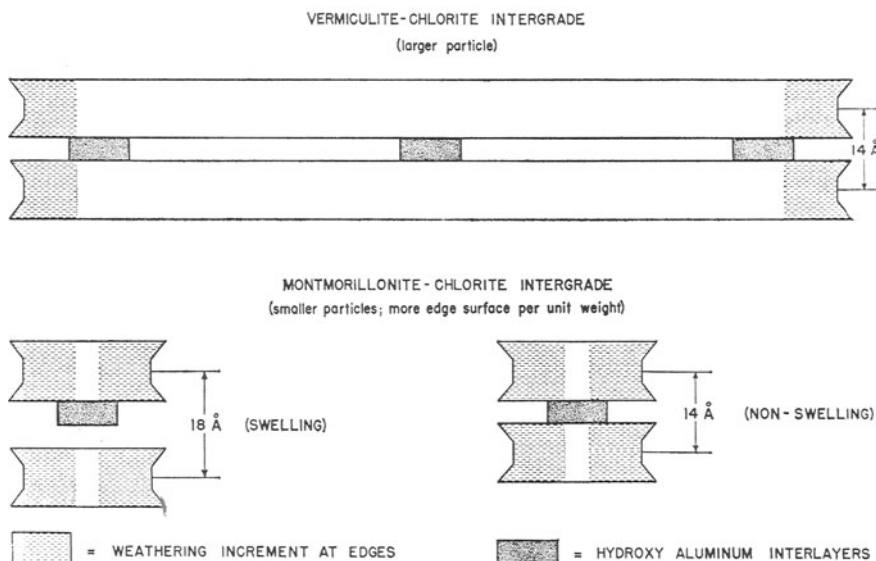## WEATHERING RATES OF 2:1-2:2 INTERGRADES



FIGURE 6.—Selective weathering away of montmorillonite in an acid environment (Jackson, 1960), owing to higher specific edge surface of montmorillonite particles, which are mainly of finer particle size. Concurrently, vermiculite formed from cleavage of larger mica particles becomes interlayered at 14 Å by hydroxy-aluminum and accumulates as weathering stage or index 9 just ahead of kaolinite of index 10 (Jackson and others, 1948).

found in the −0.08 micron fraction of the Chester soil derived from schist in Maryland (Pennington and Jackson, 1948). The survival of vermiculite–chlorite intergrades, in warm humid weathering (Fig. 3 in Aquilera and Jackson, 1953; Rich and Obenshain, 1955; Rich and others, 1959; Loughnan and others, 1962) is thus explained on the basis of claysize vermiculite formation

weathering from sand and silt sizes of mica particles. The vermiculite can weather progressively by equation (2) to montmorillonite in an acid environment, while (and after) being interlayered and thus montmorillonite sometimes appears on stripping out the interlayers (Tamura, 1957; Dixon and Jackson, 1959). A weathering index of 9 (equal to montmorillonite) has been proposed* for 2:1–2:2 intergrade clays.

*Frequency distribution:* Layer silicates with intergrade 2:2–2:1 character appear to occur rather generally in soils, varying from a small to extensive degree, and seldom negligible, as reviewed elsewhere (Dixon and Jackson, 1962). Absence of alumina interlayering of vermiculite and montmorillonite appears to be characteristic of "pure" deposits such as certain vermiculites and certain bentonites. Characteristic pedogenic occurrences of little interlayered soil clays include certain grumusols (Kunze and others, 1955; Sawhney and Jackson, 1958) and acid gley soils (Hutchinson, Lewis, and Seay, 1959). The fact that wetness and gleying "clean up" the interlayered 2:1 minerals suggests that Fe may be involved with the alumina interlayering. Concurrent occurrence of Fe with Al in pedogenic interlayers was indicated by Sawhney (1960) and Dixon and Jackson (1959). The possibility of allophanic interlayering has been indicated (Dixon and Jackson, 1962). Montmorillonite can persist in highly montmorillonitic soils long after the soils become acid (Jackson, 1959), but such soils finally give way to kaolinite (Ferguson, 1954), possibly through an interlayered vermiculite-like and chlorite stages.

Extensive weathering and soil development under good drainage apparently can rather completely develop interlayer precipitates (Fig. 5, extended) and approach the 2:2 end-member of the series, i.e. chlorite (Theisen and others, 1959; Glenn, 1960; Brydon and others, 1961). These occurrences appear to be distinct from reported occurrences of ferromagnesian chlorites (Jackson and Sherman, 1953, p. 236).
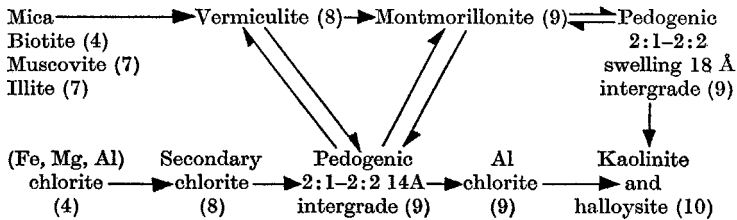
*Weathering reactions:* The 2:2–2:1 intergrade clays appear to accumulate alumina in acid soils nearly to the 2:2 level during weathering; then a subsequent weathering step, $2:2 \rightarrow 1:1$, involving silica tetrahedra inversion, could give kaolinite (Glenn and others, 1960) by translation of silicon atoms as proposed by Brindley and Gillery (1954) for daphnite (2:2–1:1 chlorite–kaolinite interstratification). Some inversion of silica tetrahedra has been proposed for montmorillonite (Edelman and Favejee, 1940) and chlorite (Bradley, 1955). Dickite pseudomorphic after mica by hydrothermal alteration has been reported by Jasmund and Riedel (1961), whose illustration is shown (by permission) in Plate 1. Structural control of mica weathering to kaolinite in residual kaolins has been noted (Sand, 1956). These two instances may represent more rapid $2:1 \rightarrow 1:1$ weathering transformation or may involve the $2:1 \rightarrow 2:2 \rightarrow 1:1$ pedogeochemical reaction proposed (Glenn and others, 1960).

Weathering of ferromagnesian chlorite (discussed above) or the incomplete

* By the author in his Kearney Foundation lectures, University of California, Dec. 1960.

reconstitution of ferromagnesian chlorite through diagenesis yields in soil clays a secondary chlorite having less thermal stability (some interlayer collapse at 550°C) than the typical ferromagnesian chlorite (14 Å reinforcement at 550°C). It appears that the ferromagnesian-related secondary chlorites, earlier (Jackson, 1959) given weathering index 8 (with vermiculite, including both trioctahedral and dioctahedral), have a lower weathering stability index than the pedogenic 2:1–2:2 intergrades of both 18 Å swelling and 14 Å type. The later have interlayer precipitates most of which are highly aluminous and are more usefully given weathering index 9 (as montmorillonite). The weathering accumulation of sufficient interlayer aluminum to prevent collapse to 10 Å at 300°C of a K saturated dioctahedral layer silicate suggests the probability of pedogenic (weathering index 9) intergrade. Trioctahedral character rather reliably indicates intergrade of index 8, but some lithologic intergrades may be dioctahedral. Although the analytical differentiation of the lithologic–diagenic relic intergrade (index 8) from pedogenically interlayered (Rich and Obenshain, 1955) intergrade (index 9) is as yet not fully worked out, the pedogenic significance of the differentiation is sufficient to warrant the attempted separation.

The various weathering reactions and indexes are summarized in equation (3):

Mica ⟶ Vermiculite (8) ⟶ Montmorillonite (9) ⇌ Pedogenic
Biotite (4)                                              2:1–2:2
Muscovite (7)                                            swelling 18 Å
Illite (7)                                               intergrade (9)

(Fe, Mg, Al)  Secondary   Pedogenic       Al        Kaolinite
chlorite  ⟶  chlorite ⟶ 2:1–2:2 14A ⟶ chlorite ⟶  and
(4)          (8)        intergrade (9)   (9)        halloysite (10)

in which the numbers in parenthesis represent weathering indexes.

*"Antigibbsite effect":* The selective accumulation of exchangeable aluminohexahydronium in the interlayer space of expansible layer silicates causes polymeric hydroxyaluminum precipitation there while the ion product in the equilibrium solution is approximately equal to or slightly less than the solubility product of free aluminum hydroxide (Ragland and Coleman, 1960) as discussed in more detail elsewhere (Jackson, 1963). The silicate layers may serve sterically (Jackson, 1960) as a template for nucleating gibbsite-like crystal units during interlayer polymerization, in addition to serving as a negative surface for sorbing the positive hydroxy ion units. The net result may be termed the antigibbsite effect, namely so long as there are expansible layer silicates in the weathering mantle, the aluminum released by weatheirng tends to be deposited in 2:1–2:2 intergrades with aluminohydroxyhydronium interlayers (Fig. 5) rather than in free gibbsite. For example, Red–Yellow Podzolic soils in southeastern United States derived from mica schist, such as the Cecil soil (Rich and others, 1959) develop alumina interlayers and then

weather on to kaolinite, and tend not to develop free gibbsite, while weathering of basic rocks more readily produces gibbsite in soils.

The occurrence of gibbsite in a mixture with expansible layer silicates could tend to indicate geomorphic erosion and depositional mixing (Erhart, 1956) of materials weathered in different sites. Very rapid weathering of mica to kaolinite (equation 3) appears to be able to produce gibbsite concurrently from feldspars. Fluctuations of the pH of the soil solution system back and forth across the isoelectric pH 4.8 of crystalline gibbsite could nucleate a separate crystalline gibbsite phase as follows: templated (+) aluminum hydroxide units sorbed on clay below pH 4.8 would become negative above pH 4.8 and thus be repelled and released, an "anti-antigibbsite effect", discussed elsewhere (Jackson, 1963). Intense pedogeochemical leaching of the sorbed (+) units would remove soluble anions and accelerate crystal growth of the released gibbsite phase.

## SUMMARY

Mica cleavage associated with crystallographic dislocation planes in the mica faces is seen as a special weathering source of both (a) frayed edge and (b) interstratification or mixed-layering in the equation:

$$\text{mica} \rightleftarrows \text{illite} \rightleftarrows \text{vermiculite} \rightleftarrows \text{montmorillonite}$$
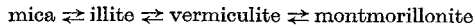
of Jackson and others (1952). The tendency in weathered layer silicates for surface constancy (K unit cell surface plus sorption surface = constant) applies to both (a) and (b) and suggests that during weathering K occupies its full mica interlayer concentration until cleavage releases the K and simultaneously yields glycol adsorption surface. Cores of unweathered mica are seen as holding $(hkl)$ orientation and juxtaposition of the K cavities in the frayed edge vermiculite and montmorillonite, thus accounting for K ion preference by the species of these two minerals weathered from micas as compared the species of these minerals derived by weathering of minerals other than layer silicates.

Intergradient 2:1–2:2 layer silicate minerals develop in soil clays by lattice building or precipitation of aluminum hydroxide attached on one interlayer surface of montmorillonite (18 Å swelling intergrade) or on two interlayer surfaces of vermiculite (14 Å intergrade). The 18 Å intergrade appears to form more frequently in alkaline soils in which montmorillonite is more stable, while the 14 Å intergrades appear most frequently in acid soils in which the reactions

$$\text{mica} \rightleftarrows \text{vermiculite} \rightleftarrows 14 \text{ Å intergrade}$$
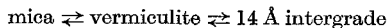
are favored by relatively rapid decomposition of any montmorillonite formed (higher specific surface of edges) with release of aluminum for interlayer "lattice building" in the more highly charged and larger vermiculite particles forming from mica. This disposition of aluminum tends to preclude free
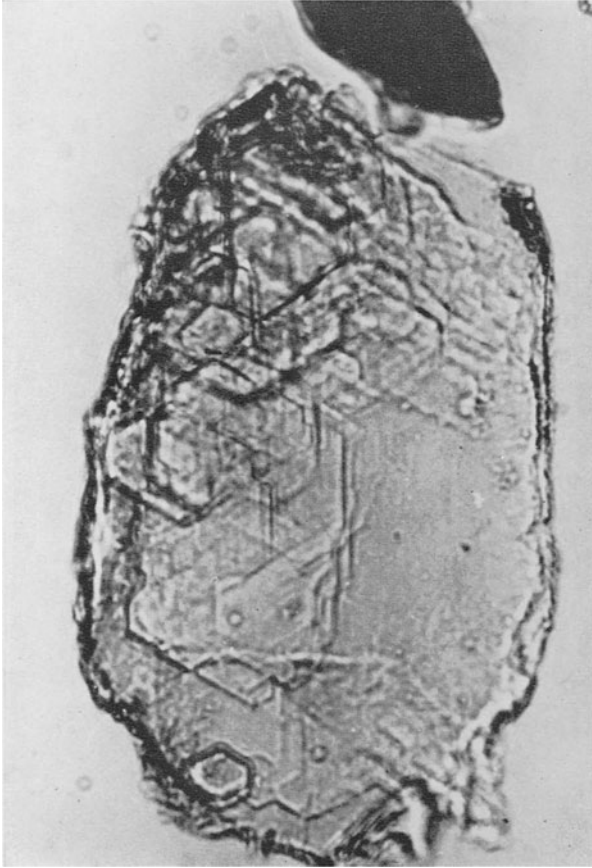
PLATE 1.—Crystals believed to be dickite formed through hydrothermal alteration of mica in sandstone (from Jasmund and Riedel, 1961, courtesy *Bull. Geol. Inst. Univ. Uppsala*).

Al $(OH)_3$ formation in soils so long as there are actively weathering 2:1 layer silicates present in soils ("antigibbsite effect"). Weathering proceeds by the reactions:

$$14 \text{ Å intergrade} \rightleftharpoons \text{Al-chlorite} \rightleftharpoons \text{kaolin}$$
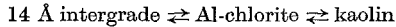
Free gibbsite can appear through weathering when leaching rates are rapid and the supply of silica is limited.

## ACKNOWLEDGMENTS

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