TRANSFORMATIONS OF MICAS IN THE PROCESS OF KAOLINITIZATION OF GRANITES AND GNEISSES

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Abstract—Micas are described from the kaolin type Tertiary weathering crusts formed on granites and gneisses of Lower Silesia, Poland. In the weathering crust, large scale processes of transformation of micas (muscovite, biotite) take place. Weathering of micas is a gradual transformation of their structures by removal of the mobile cations Mg^{2+}, Fe^{2+}, Fe^{3+} and finally K^+. Muscovite transforms into kaolinite through the intergrowths of two phases, mica and kaolinite. A degradation series, muscovite → mica/montmorillonite → montmorillonite → kaolinite also is observed. Biotite passes directly into kaolinite, but at higher concentrations of K and Al one observes the following sequence of biotite degradation: biotite → dioctahedral mica rich in Fe → dioctahedral mica poor in Fe → kaolinite. This process involves the exchange of Mg^{2+} and then Fe^{2+} of octahedral layers by Al^{3+} less mobile under weathering conditions. In the next stage of degradation, K^+ removal and kaolinite formation takes place. The micas representing different stages of this transformation, dark green, light-green, and silver-white, have been separated and investigated.

INTRODUCTION

Degradation and aggradation of micas and their formation in endogenic conditions constitute a group of important processes taking place in the lithosphere. The direction and mechanism of these processes have been studied recently by numerous workers. Many papers have been published describing experiments on degradation and aggradation of micas and also on the behavior of micas during weathering and epigenesis of sediments.

Many basic problems relating to the behavior of micas in the lithosphere have not been satisfactorily explained. No detailed data have been given on the nature of micas occurring in weathering crusts. Weathering of acidic crystalline rocks containing biotite and muscovite provides favorable conditions for observing the structural changes which occur in micas. The present work presents the results of studies of micas in Tertiary weathering crusts formed on granites and gneisses of Lower Silesia. The weathering products are of kaolinite type and have been preserved locally in cavities and hollows between the heights built of crystalline rocks (Budkiewicz, 1964). Their thickness is several tens of meters and they are usually covered with layers of Miocene clays with inserts of brown coal and Quaternary sediments. They show the zone-like distribution of minerals which is typical of weathering crusts of crystalline rocks formed *in situ* (Ginzburg, 1963; Petrov, 1967). In the lower part of these weathering crusts one may distinguish a zone of slightly changed primary rock (saprolite) above which there is a narrow kaolinite—mica zone, and higher still a kaolinite zone comprises the largest part of the weathered section.

In the lowermost of these zones (slightly changed primary rock) the main components are minerals of the parent rock such as potassium feldspar and plagioclases, quartz, biotite and muscovite. On the surface of feldspars (in particular potassium feldspars), secondary mica and kaolinite are formed. Kaolinite is formed mainly from plagioclases. Also one may observe locally the commencement of kaolinitization of micas. In this zone biotite changes its color to green. On the borders of plates and along cleavage planes there appear precipitates of oxides and hydroxides of iron. In addition to primary quartz, local precipitates of secondary low-temperature quartz are observed.

Basic minerals of the kaolinite–mica zone are quartz, kaolinite and mica. Feldspars are observed in small amounts in the form of small grains surrounded by a thick layer of their weathering products. Potassium feldspar is mainly preserved. In this zone, primary as well as secondary micas occur. One may distinguish dark-green trioctahedral micas which are transformed biotite, light-green dioctahedral micas and silver-white dioctahedral ones which are changed muscovite of the primary rock. The kaolinite–mica zone is best developed from biotite gneisses. The kaolinite zone is mainly kaolinite and quartz. Micas occur in minor quantities. They are dioctahedral micas of muscovite type of silver-white color.

Physicochemical conditions existing at the time of formation of weathering layers, governed mainly by climate and the topography of the terrain, favored formation of secondary micas in the weathering crust and relics of primary micas at different levels of preservation.

MICAS FROM WEATHERING CRUSTS OF GRANITES
AND GNEISSES FROM LOWER SILESIA

In order to study the character of micas occurring in weathering crusts, micas of different colors were separated. These were relatively coarse-grained, dark-green, light-green and silver-white micas. Elutriation, electromagnetic, and heavy liquids separation was used. In addition to these micas, illite types also were observed.

Dark-green mica

Dark-green mica is biotite changed by weathering. It differs from fresh biotite in color, dullness and weaker pleochroism. Optical properties change within one plate. In certain parts, usually at the borders, the color of the plates is lighter and often they are completely colorless. The colorless fragments show a birefrigence of 0.009 and the optical features of kaolinite. A zone-like character of the optical properties can be seen also in the direction perpendicular to 001. Occasionally transition of dark-green mica into its light-green modification with the optical properties of muscovite is observed. At the borders of mica plates and along the cleavage planes, precipitates of iron oxides occur (Fig. 1).

X-ray studies of separated plates of dark-green mica, $>60\ \mu m$ in size, demonstrate their multiphase composition. The X-ray diffractograms show the lines of trioctahedral mica and of kaolinite (Fig. 2). The ratio of the intensities of the 00l lines indicates a high content of iron in the octahedral sheets. The 060 line (1.522–1.528) also shows a bend at 1.515 which suggests the presence of a small amount of ferruginous dioctahedral mica (Fig. 3). Removal of potassium from the interlayer space during weathering of micas
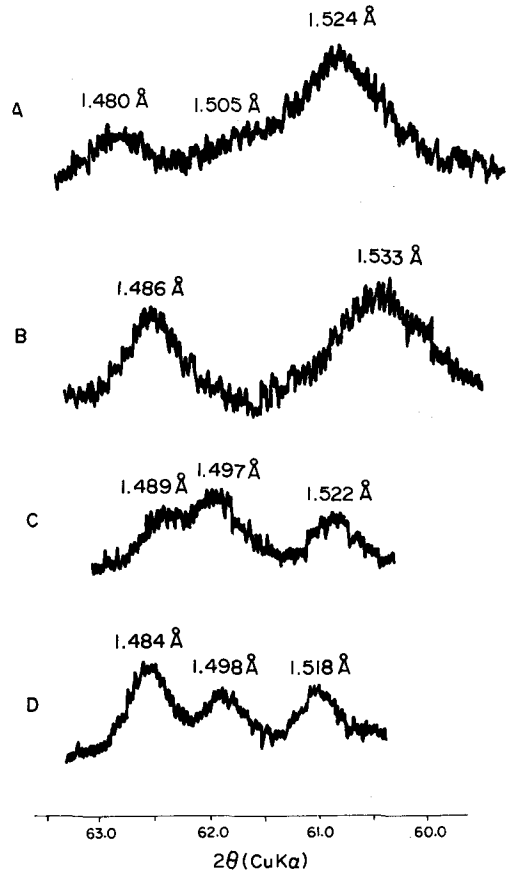

Fig. 3. 060 lines of dark-green micas (A, B) and light-green micas (C, D) from Wyszonowice and Żarów respectively.

causes a change in the position of the 060 line, but in the direction of an increase in the value of d_{060} of trioctahedral mica (Leonard and Weed, 1970)
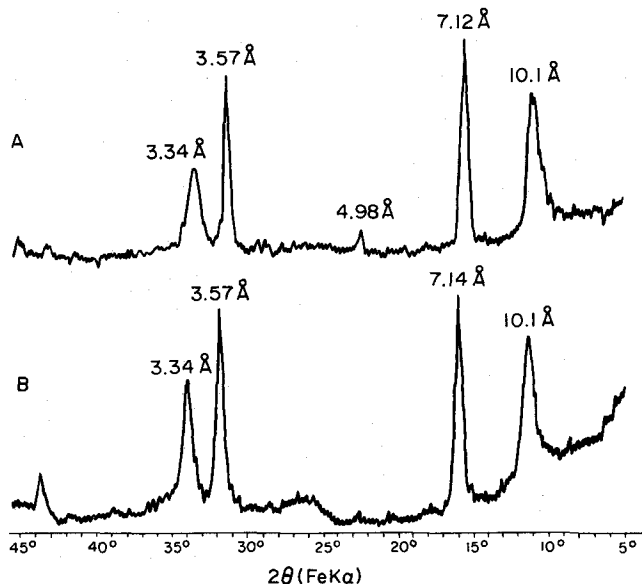

Fig. 2. X-ray powder diffraction patterns of dark-green mica, separated from: (A) weathered granite from Żarów, (B) weathered gneisse from Wyszonowice.
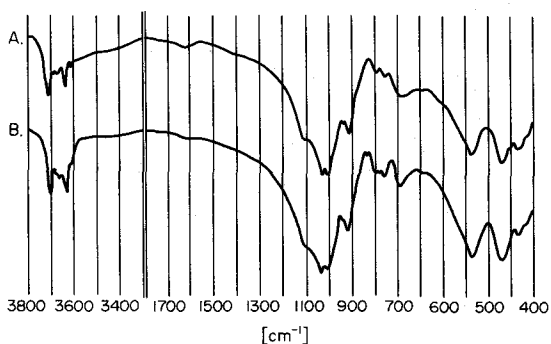
Fig. 4. I.r. absorption spectra of the dark-green mica (A) and light-green mica (B) from Wyszonowice daolin.

which cannot be the cause of the appearance of the bend mentioned above. The mica line at 10 Å shows broadening on the small angles side, which disappears after saturation with K^+ and after heating at 550°C, which indicates formation of intergrowths of mixed-layer mica–vermiculite. The i.r. absorption spectrum of the mica shows the occurrence of bands of kaolinite (Fig. 4).

On the DTA curve, an endothermic peak at 100°C is related to removal of interlayer water and 2% loss in weight, and exothermic peak at 250–400°C corresponds to oxidation of iron, an endothermic peak at 560°C resulting from dehydroxylation is accompanied by a 9% weight loss, and finally an exothermic peak of kaolinite at 980°C (Fig. 5).
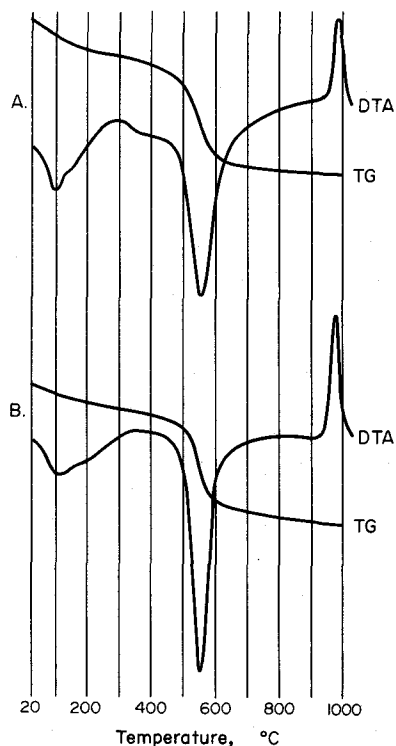
These results indicate that macroscopically homogenous plates of dark-green mica are multiphase. They are composed of trioctahedral mica, kaolinite, mixed-layer mica–vermiculite, and also small amounts of dioctahedral mica. These phases do not form a mechanical mixture of separate plates but are intergrown mutually within one mineral entity—i.e. the plate of weathered biotite.

Co-existence of different phases within plates of mica is visible in electron micrographs. Fragments of different absorption of electrons caused by local differences in chemical composition, especially the iron content, can be distinguished. Sometimes hexagonal plates of kaolinite are observed within plates of micas (Figs. 6 and 7).

X-ray probe analysis perpendicular and along 001 plates (Figs. 8 and 9) shows regions of trace content of iron, potassium and magnesium (kaolinite) and of high content of these elements (trioctahedral mica).

The chemical formula of the mica phase calculated from chemical analysis (Table 1) after taking into account the kaolinite content is as follows:
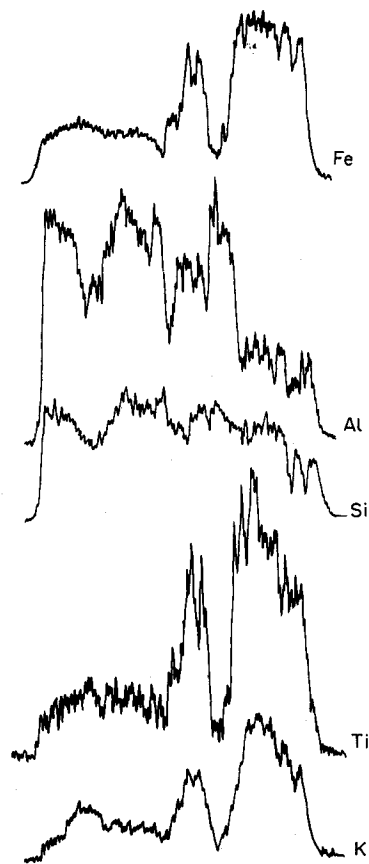
$$K_{0.86}Na_{0.05}(Mg_{0.79}Fe^{2+}_{0.79}Fe^{3+}_{1.02})$$
$$\times [Si_{2.87}Al_{1.11}O_{10}] (OH)_2.$$

Fig. 5. DTA and TG curves of dark-green mica (A) and light-green mica (B) from Wyszonowice.
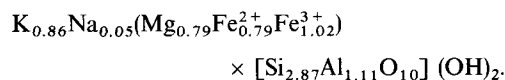

Fig. 8. Fe, Al, Si, Ti and K distribution in a dark-green mica flake from kaolin Wyszonowice, along the line marked on the electron micrograph (Fig. 9).
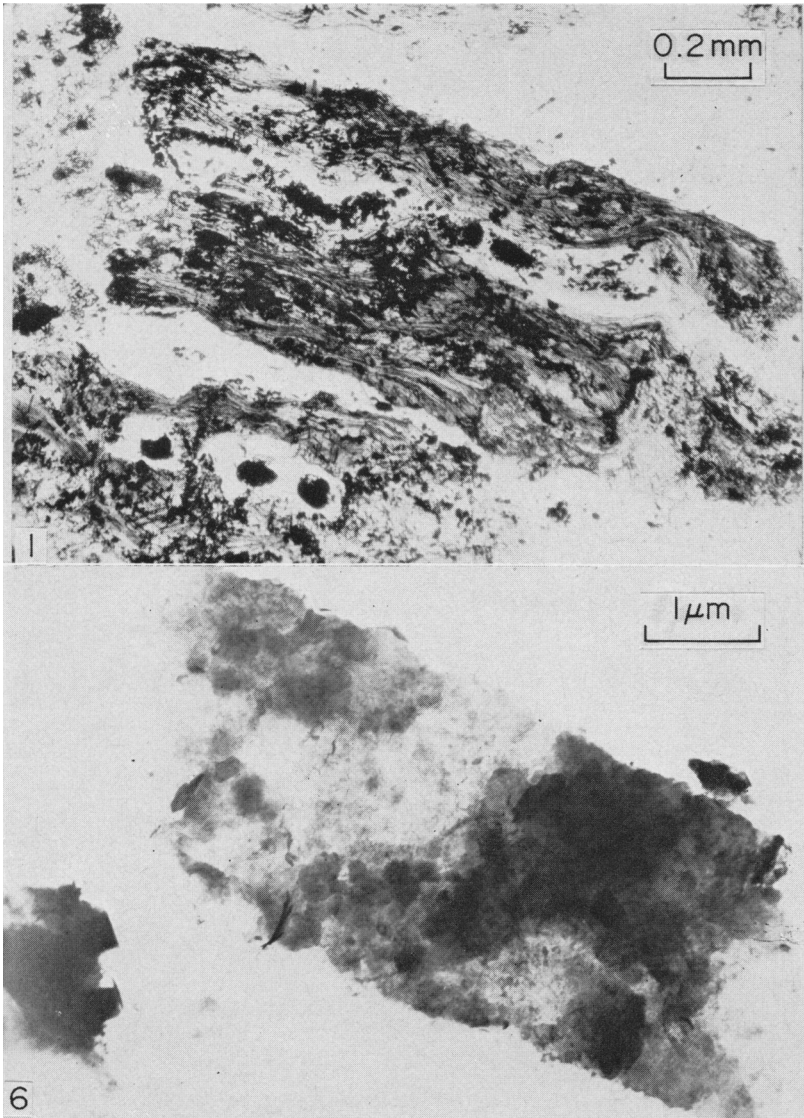
Fig. 1. Dark-green mica with precipitates of iron oxides. Parallel light.
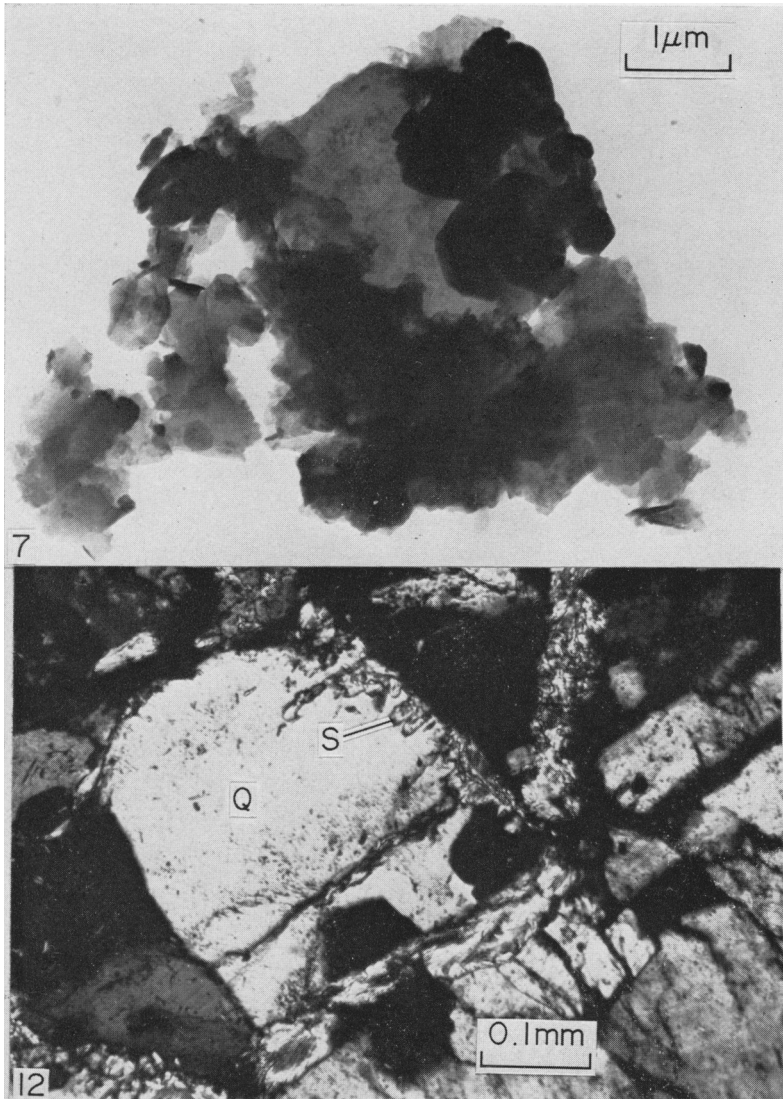Fig. 6. Electron micrograph of plate of dark-green mica.

Fig. 7. Hexagonal kaolinite plates formed within the plate of dark-green mica.

Fig. 11. Sericite formation on the surface of quartz grain in kaolin from Boleslawice; *s*—sericite, *q*—quartz.
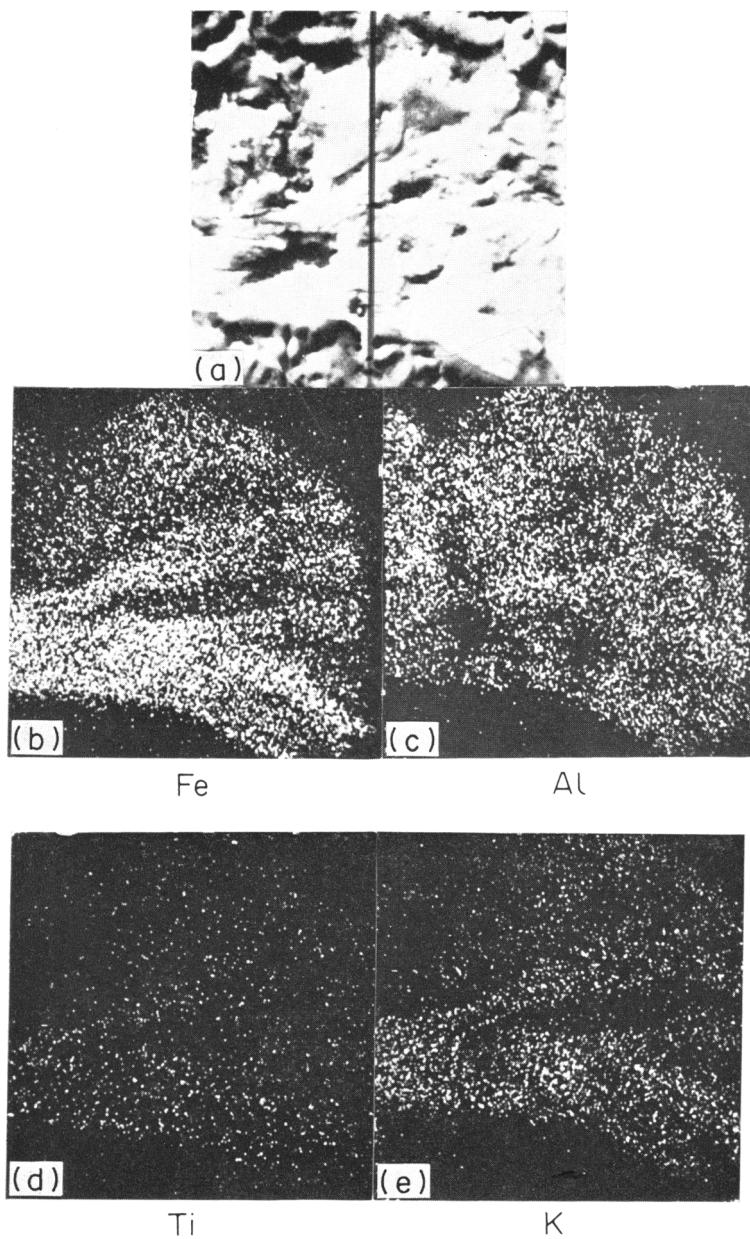
Fig. 9. Fe, Al, Ti and K distribution in a dark-green mica plate from kaolin from Wyszonowice.

Table 1. Chemical composition of micas

	Dark-green mica	Light-green mica	Silver-white mica
SiO_2	39.88	48.49	44.85
AL_2O_3	26.53	33.13	36.67
TiO_2	1.54	0.36	0.27
Fe_2O_3	7.72	1.71	1.35
FeO	4.95	0.48	—
CaO	—	—	0.41
MgO	2.78	0.81	0.56
K_2O	3.63	3.59	0.88
Na_2O	0.08	0.18	0.06
$+H_2O$	9.38	8.92	11.37
$-H_2O$	2.79	1.87	2.04
Total	99.28	99.54	98.46

The content of kaolinite and mica was determined from the amount of structurally bound water. It was assumed that kaolinite has the theoretical chemical composition. The presence of mica–vermiculite and dioctahedral mica was neglected since these components occurred in minor quantities. On the basis of the calculated formula, the mica phase in dark-green mica can be determined (after Foster, 1960) as Fe–biotite in which a part of Fe^{2+} is oxidized into Fe^{3+}.

Light-green mica

The light-green mica also represents intergrowths of mica and kaolinite, It is formed from the dark-green mica and constitutes a later stage in the weathering of biotite. Many plates of this mica possess well-preserved fragments of dark-green mica. In contrast to the dark-green mica, the mica phase in intergrowths is dioctahedral with a low iron content ($d_{060} = 1.498$). This is accompanied by trioctahedral mica ($d_{060} = 1.520$ Å) occurring in minor quantities (Fig. 3).

The average chemical composition of the mica phase of the intergrowths calculated from chemical analysis (Table 1), after taking into account the presence of kaolinite, is given by the formula:

$$K_{0.58}Na_{0.03}(Al_{1.71}Fe^{3+}_{0.012}Fe^{2+}_{0.005}Mg_{0.15})$$
$$\times [Si_{3.46}Al_{0.54}O_{10}] (OH)_2.$$
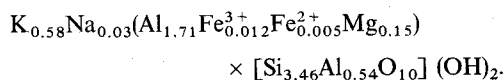
Silver-white mica

In one of the kaolin deposits formed by weathering of a gneiss a silver-white mica was found for which the X-ray analysis shows a swelling phase in addition to lines of kaolinite and mica. The swelling phase gives a line at 14.7 Å which, after saturation with glycerol, is shifted to 17.7 Å. The position of the line at 14.7 Å depends on the type of exchangable cations (Fig. 10). After saturation with K^+, the 14.7 line is shifted to 10 Å and the sample loses its swelling properties. When heated to 550°C the swelling structure collapses.

Irreversible collapse of the structure, after saturation with K^+, indicates a higher-than-usual layer charge of the montmorillonite. It also differs from montmorillonite by the sharp profile of the 001 line (Fig. 10).

The X-ray diagram of the silver-white mica also shows weak lines, at 24 and 12 Å, of a regular mixed-layer mica/montmorillonite. It appears that this phase is an intermediate form in the transformation of mica into dioctahedral montmorillonite, which in the next stages transforms into kaolinite.

The following formula was calculated on the basis of chemical analysis of the mica (Table 1) after substracting the participation of kaolinite. It corresponds to the average chemical composition of the mica and montmorillonite phases:

$$K_{0.35}Na_{0.04}Ca_{0.11}Mg_{0.25}Al_{1.79}Fe_{0.17}$$
$$\times [Si_{3.07}Al_{0.93}O_{10}] (OH)_2 \cdot nH_2O.$$
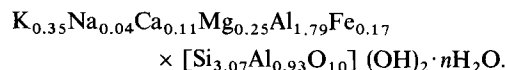
Illite

In fractions $< 2\ \mu m$ separated from the material of the weathering crust a fine-grained secondary mica has been found. It constitutes up to 30 weight % of this fraction. This is dioctahedral mica (Md) of illite type, containing a relatively high amount of iron (6.5% Fe_2O_3). The 001 line of this mica shows a
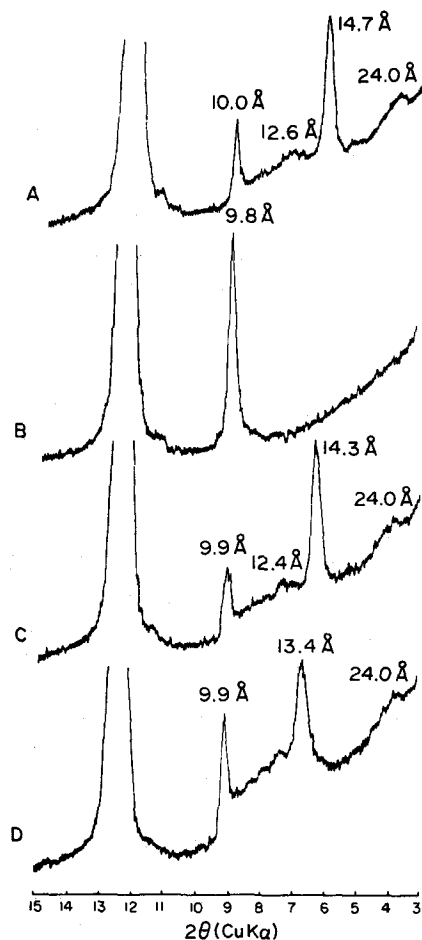

Fig. 10. 001 lines of silver-white mica (A) and that saturated with K^+ (B), Mg^{2+} (C) and Al^{3+} (D).

characteristic widening caused by the presence of irregular interlayering of montmorillonite layers.

FORMATION AND DEGRADATION OF MICAS IN
WEATHERING CRUSTS

During the weathering of crystalline rocks, complex physico-chemical processes occur which lead to the formation of new minerals. In the weathering crusts on granites and gneisses of Lower Silesia the most important mineral-forming processes were incongruent dissolution of rock-forming minerals of the crystalline rocks, and structural transformation of layer silicates (Sikora and Stoch, 1972).

The formation of micas generally is observed within the zone of slightly changed parent rock and locally in the kaolinite–mica zone. Microscope observations show that incongruent dissolution of feldspars in these zones leads to the formation on their surface of mica* (sericite and/or illite). This process takes place under alkaline conditions and with appropriate activity of K^+ ions in the surrounding pore solution. The conditions under which this process is possible have been determined by studies on the solubility of potassium feldspars (Garrels and Howard, 1959) and also by thermodynamic calculations which provide appropriate diagrams of mineral equilibria (Hess, 1966; Tardy, 1970). In the zone of slightly changed original rock, local formation of sericite is observed at the borders and along cracks of quartz grains (Fig. 11). This sericite is formed probably by dissolution of quartz in solutions containing ions of K^+ and Al^{3+}, the latter being provided by minerals remaining with them in equilibrium.

At the same time large scale processes of transformation of original micas (biotite, muscovite) and of secondary ones (sericite or illite) take place in the weathering crust. Structural transformation of layer silicates during weathering consists in reorganization of their structure, accompanied by a change of chemical composition by exchange of some components with their surroundings (H_2O, K^+, Mg^{2+}, Si^{4+}, Al^{3+}, Fe^{2+}, Fe^{3+}). Transformation of biotite takes place in a different manner from that of muscovite which is also observable in the studied weathering crusts. The micas described above represent different stages of this transformation and make it possible to draw conclusions about its mechanism.

Transformation of muscovite

The final effect of degradation of muscovite in the weathering crust is the formation of kaolinite, which is frequently observed although under continental conditions of weathering. Formation of kaolinite from muscovite has been known for a long time, although

many authors do not take this into account and confine the transformation to similar structures of the 2:1 type. Some authors believe that the muscovite–kaolinite transition takes place through the stage of hydromuscovite (Petrov, 1967). Studies of kaolinitized muscovite from kaolinite clays have shown that the muscovite–kaolinite transition may take place directly. The stage described as hydromuscovite is in fact an intergrowth of two phases, kaolinite and muscovite, co-existing within the original plate of mica. The formation of structures containing molecular water between the layers and representing the intermediate stages of transformation have not been found (Stoch, 1960).

The finding of dioctahedral montmorillonite and regular mixed-layer mica–montmorillonite in one of the products of weathering of muscovite suggests that the transformation into kaolinite may take the following course:

muscovite → muscovite/montmorillonite

→ montmorillonite → kaolinite.
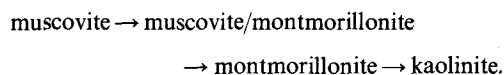
In the products of muscovite weathering, however, montmorillonite only rarely is observed and it may be assumed that it constitutes an unstable intermediate phase.

To form kaolinite from muscovite it is necessary to remove potassium and to bring from the surroundings water and protons which are transformed into OH groups, coordinating aluminum in accordance with the equation:

$$2KAl_2[Si_3AlO_{10}](OH)_2 + 2H^+ + 3H_2O$$
$$= 1.5Al_4[Si_4O_{10}](OH)_8 + 2K^+.$$
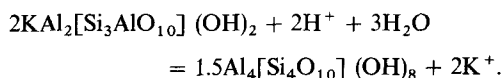
Rebuilding of a 2:1 mica layer into a 1:1 kaolinite layer requires displacement of part of the Si^{4+} and Al^{3+} ions. The mechanism of this process has not been elucidated as yet although an attempted explanation was presented by Kukovsky (1969). It may be expected that the process of formation of kaolinite commences by the shift of Al^{3+} ions from tetrahedral positions to interlayer space with a simultaneous change of coordination from IV into VI and the development of the new octahedral layer. At the same time, K^+ is removed and protons and molecules of water are incorporated for the formation of the full coordination of aluminium cations. Then there takes place a replacement of Si^{4+} ions of one of the tetrahedral sheets of the mica layer towards forming a new octahedral sheet. This is equivalent to an inversion of SiO_4 tetrahedra of this sheet. A similar inversion of tetrahedra is observed in other silicate structures (Taylor, 1960). In this way three layers of kaolinite are formed from two layers of mica. These become domains within the structure of mica, growing gradually as the process proceeds.

It can be expected that with appropriate physico-chemical weathering conditions and in particular at a sufficient pH, the aluminium ions placed between

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* Fine-grained secondary dioctahedral mica $2M_1$, having optical properties like muscovite. It differs from mica termed illite, among other things, by the sharp 001 X-ray line.

layers of mica in the first stage of kaolinization may be coordinated by molecules of water. This is equivalent to the formation of a montmorillonite structure. In other conditions, perhaps at higher activity of hydrogen ions, aluminium is coordinated solely by OH groups and kaolinite is formed directly with omission of the montmorillonite phase.

Transformation of biotite

Weathering of biotite in the studied crusts consists in a gradual structural transformation of mica terminating with formation of kaolinite, or a direct formation of kaolinite, according to the schemes:

(a) biotite → dioctahedral mica → kaolinite

(b) biotite → kaolinite.

Both processes are accompanied by oxidation and isolation of iron in the form of oxides or hydroxides. In the first case it proceeds gradually, in the second case the process is rapid.

Weathering takes place according to the first or the second scheme depending on the concentration of K^+, H^+, Al^{3+}, Mg^{2+} ions in the immediate surroundings, as it follows from the phase diagram of the minerals (Fig. 12) made on the basis of thermodynamic calculations. The values of free energy used were taken from works by Rossini *et al.* (1952), Barany (1964), Garrels and Christ (1965) and Reesman and Keller (1968). The value of free energy of phlogopite instead of that of biotite has been taken in the thermodynamic calculations.

Transformation of trioctahedral into dioctahedral mica may take place at appropriately high concentration of Al^{3+} when $lg[K^+]/[H^+] \geqslant 6$. In view of the small value of the ratio $[K^+]/[H^+]$ trioctahedral mica passes directly into kaolinite. For this reason the first of the processes takes place in the lower part of the weathering profile in the neighborhood of potassium feldspars. Transformation of biotite into dioctahedral mica (muscovite) during kaolinitization

of granites has been noted by Konta *et al.* (1972) who proposed the mechanism of this process.

Chemical composition of the studied micas, constituting the successive stages of transformation of biotite indicates that transformation of trioctahedral into dioctahedral mica may take place by exchange of cations of the octahedral layer by aluminum according to the equation:

$$KMg_3[AlSi_3O_{10}] (OH)_2 + 2Al^{3+}$$
$$= KAl_2[AlSi_3O_{10}] (OH)_2 + 3Mg^{2+}.$$
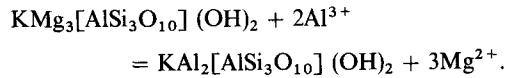
The changes occurring in the octahedral layer may not take away cations of inter-layer micas. Investigations by Farmer *et al.* (1971) have shown, for instance that oxidation and removal of part of the iron from biotite takes place without change of K^+ content; the presence of trivalent iron causes an increase of the strength of potassium bonding in the structure (Barshad and Kishk, 1968).

Iron removed from biotite in the initial stages of the weathering is concentrated along the planes of cleavage (Fig. 1). This suggests that diffusion of iron ions from the inside of layers occurs normal to the plane 001 due to the filling of the interlayer space by potassium ions. Magnesium may be removed in a similar way. Migration of these cations, mobile in the weathering conditions, takes place following the gradient of concentration from their high concentration in the bulk of the structure to the low concentration in the pore solutions filling the weathering crust. Loss of magnesium and iron may be equilibrated by the flow of aluminum ions in the opposite direction. Stability of potassium in the structure is secured by its high concentration in the surroundings maintained by the weathering feldspars. Interlayer potassium most probably stabilizes aluminum in the tetrahedral layer. Feldspars also may be the source of aluminum ions necessary for the transformation of trioctahedral into dioctahedral mica. From experimental studies on the weathering of feldspars, not all of the aluminum is bonded in the structure of the solid products. A certain amount of it passes into solution.

Direct formation of kaolinite from biotite may be considered as either incongruent dissolution or the result of transformation of mica structure. The first of these processes occurs when the kaolinite plates have an orientation incompatible with the 001 planes of mica. Their position is random, or one observes epitaxial growth of kaolinite on biotite. In the latter case, kaolinite forms columnar aggregates developed perpendicular to the biotite surface.

Transformation of the mica structure with the participation of Al^{3+} ions from the outside may account for the formation of (pseudomorphs) of kaolinite after biotite with orientation and shape the same as that of the mica plates. The fact that the volume of the pseudomorphs is equal to or slightly higher than the volume of the mica plate proves that kaolinite is
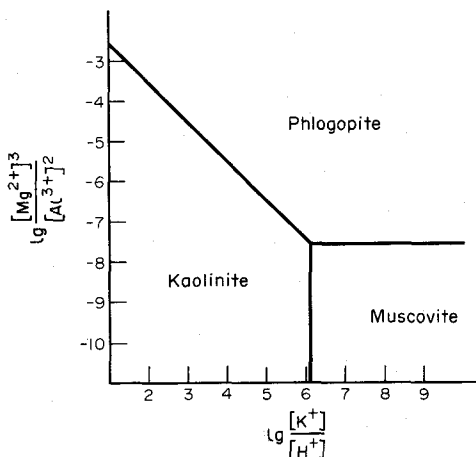

Fig. 12. Stability relations among kaolinite, muscovite and phlogopite.

formed with participation of Al^{3+} ions from outside. If kaolinite was formed solely from the aluminum of biotite, then the theoretical volume of the kaolinite formed would be about 67% smaller than the mica volume.

The changing physico-chemical conditions in the weathering crust cause the second weathering processes to be superimposed on the first and direct formation of kaolinite from biotite is observed at each stage of weathering. It is formed from dark green, trioctahedral mica as well as from dioctahedral mica.

SUMMARY

During the process of kaolinitization of acidic magmatic rocks, the structure of muscovite usually undergoes direct transformation into kaolinite, without a stage of illite or hydromica phase. Gradual transformation of the muscovite structure according to the scheme: muscovite → mica/montmorillonite → montmorillonite → kaolinite, is also observed. Biotite passes directly into kaolinite, the ratio of concentrations K^+/H^+ is low in the weathering medium. At high concentrations of K^+ and Al^{3+}, one observes the following sequence of biotite degradation: biotite → dioctahedral mica rich in Fe → dioctahedral mica poor in Fe → kaolinite. This process consists in exchange of Mg^{2+} and then Fe^{2+} of the octahedral layers of micas into less mobile Al^{3+} in the weathering conditions. Finally K^+ is removed in the stage of kaolinite formation.

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