

*An occurrence of phlogopite and its transformation
to vermiculite by weathering*

With Plate VII

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Summary. The characteristics of a phlogopite and its weathering product, vermiculite, have been studied by X-ray, infra-red, electron-microscope, and chemical methods. Inclusions within these minerals have been identified as thin films rich in iron oxide (which impart a schiller effect), spinel (which may exist in a preferred orientation), and serpentine. The orientation relationship between the fresh mica and its weathering product has been established by single-crystal methods. The mechanism of transformation from a 1-layer to a 2-layer polytype is discussed.

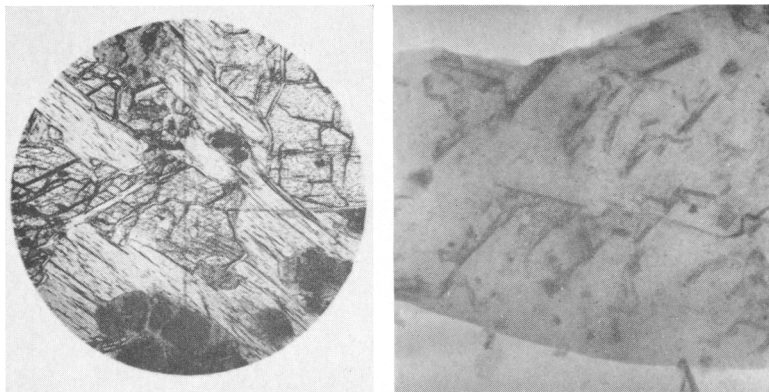
A SOIL sample containing the material used in the present investigation was submitted to the Macaulay Institute for identification of a gold-coloured mineral that was abundant both in the soil and on the surfaces of rock particles included in that soil. The sender did not include details of the exact locality of the occurrence but the parent rock has been identified as scyelite, an ultrabasic rock that was named by Judd (1885) after Loch Seye, situated about 5 miles west of Scotscaidder, Caithness.

Experimental procedure

A thin section was made of one of the freshest rock fragments and the rock type determined by optical examination. It was obvious, on inspection, that the micaceous component in the rock alters to the golden material, and an attempt was made to concentrate a pure sample of this mica.

Fresh cores of rock fragments were crushed and a 30-100-mesh size-fraction selected for separation according to specific gravity. Using bromoform and a mixture of bromoform and dimethyl sulphoxide the fraction of density 2.76-2.9 was found to contain most mica. About 100 mg of flakes that were free of inclusions were hand-picked under the binocular microscope. The material of density < 2.4 from the 30-100-mesh fraction of the soil appeared to consist almost entirely of golden weathered mica flakes.

Several micaceous crystals of maximum diameter 0.5 mm, taken from the fresh rock, its weathered surface, and the soil, were examined by X-ray diffraction using a Unicam single-crystal Weissenberg goniometer, S. 35. For each exposure with Co- $K\alpha$ radiation, a single film was used; no attempt was made to standardize exposure times (approx. 60 hours) or to correlate the intensities of reflections on one film with those on another. The axes for crystal rotation were chosen after inspection



FIGS. 1 and 2. FIG. 1 (left). Scyelite, with amphibole (centre) surrounded by phlogopite with inclusions of serpentine (bottom). $\times 42$. FIG. 2 (right). Electron micrograph showing extremely small inclusions of serpentine in phlogopite. Many of the crystallites exhibit preferred orientation. $\times 13\ 900$.

of interference figures using the conoscopic lens system of the optical microscope.

Samples of the selected fresh and weathered material were immersed in a little isopropyl alcohol and hand-ground in a mortar until fine enough to pass through a 200-mesh sieve. X-ray powder diffraction and infra-red analyses were carried out on these samples and the cation-exchange capacity determined on the weathered sample only.

In addition to these techniques, spectrochemical and electron-optical investigations were made to provide supplementary information.

Results

Optical examination of the scyelite has shown it to be characterized by large anhedral crystals of phlogopite and a colourless amphibole (fig. 1). Abundant serpentine pseudomorphs after olivine are enclosed poikilitically in these minerals and small euhedral crystals of spinel are

dispersed throughout the rock. The mineral assemblage was considered by Judd to be secondary in origin, the rock being derived from a mica-picrite. His description of scyelite is perfectly applicable to the rock sample under consideration here. The Geological Survey (1960) describe the rock as a biotite-hornblende-peridotite although in the present instance the mica has been identified as phlogopite.

TABLE I. Powder diffraction data (1) for phlogopite from scyelite, (2) by Smith and Yoder (1956), (3) for vermiculite from Scotscaider, (4) for West Chester vermiculite by Walker (Brown, 1961)

1M and 3T phlogopite				vermiculite			
1		2		3		4	
<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>
10.05 Å	10	10.05 Å	vs	14.5 Å	10	14.4 Å	vvs
5.05	2	5.02	w	7.2	1	7.2	vw
4.57	4	4.59	vw	4.8	2	4.79	vw
		4.55	vw	4.63	3	4.60	s
3.92	1	3.93	vvw	3.58	4	3.587	m
3.67	3	3.66	vw	2.87	5	2.869	m
3.36	9	3.39	m	2.65	2	2.657	mw
		3.35	vs	2.60	3	2.597	m
3.16	3	3.14	w	2.56	3	2.550	mw
2.93	3	2.92	w	2.40	4	2.392	ms
2.71	1	2.71	vw	2.275	1	2.266	vvw
2.61	7	2.64	vw	2.21	1	2.214	vw
		2.62	m	2.085	1	2.081	w
2.52	4	2.51	m	2.055	1	2.048	vw
2.43	5			2.025	1	2.011	vw
2.26	1			1.84	1	1.835	vvw
2.18	5			1.75	1	1.748	w
2.02	6			1.68	2	1.677	mw
1.99	2			1.58	1	1.574	vvw
1.90	1			1.545	3	1.537	ms
1.69	4						
1.53	5						

Unaltered mica. A preliminary examination of the mica and its weathered product was carried out using powder diffraction cameras. Several different small flakes of fresh mica were rotated in turn about an arbitrary axis within the cleavage plane and for each sample basal spacings of about 10 Å and at least 8 orders of this spacing were recorded. A weak 7 Å line was occasionally noted due to minor amounts of serpentine inclusions. A complete powder diffraction pattern of a ground sample of mica showed good agreement with the data for 1M or 3T phlogopite given by Smith and Yoder (1956) (table I). Infra-red studies on the phlogopite have shown that it is not quite normal in that the

frequency of the O-H stretching vibration is slightly higher than usual, i.e. 3718 cm^{-1} as against $3704\text{--}3709\text{ cm}^{-1}$. The presence of a very small amount of chloritic material and serpentine is also indicated by this technique and is not inconsistent with optical or electron-optical observations (fig. 2). A spectrographic investigation of the trace element content showed 0.2 % Cr, which is higher than normal but may be explained by the presence of small inclusions of the spinel, chromite.

Weathered mica. X-ray studies on the weathered mica flakes from the soil using powder diffraction cameras showed that the 10 \AA line and integral orders are in most cases replaced completely by a reflection at about 14 \AA and at least 12 orders of this basal spacing. Since this diffraction pattern could be characteristic of either chlorite or vermiculite, differentiation was attempted by observing the effects of heat treatment. Many different flakes were heated in air to 600° C , 650° C , and 700° C for several hours and with few exceptions a strong reflection remained at 14 \AA ; as little time as possible was lost between removing the flakes from the furnace and setting them in the powder cameras, which were immediately evacuated to reduce rehydration to a minimum; powdered samples and even flakes that had been exfoliated in a bunsen flame retained the 14 \AA spacing and it seemed at first that the weathered material was chlorite.

However, the powdered material did occasionally have a 12 or 10 \AA phase or both accompanying the 14 \AA phase after heating, and on this account it was decided to heat a sample to 600° C in a Pyrex glass tube, which was sealed immediately on removal from the furnace to prevent any possible rehydration. In this case, and for several other samples including a single crystal, a 10 \AA line was produced without any trace of a 14 \AA phase; displacement of the 14 \AA reflection to 10 \AA on heating is incompatible with a chlorite structure. Thus, the findings of Walker (Brown, 1961) are emphasized in that a false diagnosis of 'chlorite' may be made if heated samples are allowed to remain in a humid atmosphere for even very short periods before examination. If possible, a high-temperature camera should be used.

Contraction of the vermiculite lattice on NH_4^- or K-saturation has been studied by Barshad (1954), who concludes that for material with a cation-exchange capacity of 150 or more me per 100 g the basal spacing becomes 11.2 \AA for NH_4^- -saturation and 10.4 \AA for K-saturation. In further studies Weaver (1958) has found that the degree of contraction appears to be a function of the total charge and the mineral is important in that those derived from mica-type minerals tend to

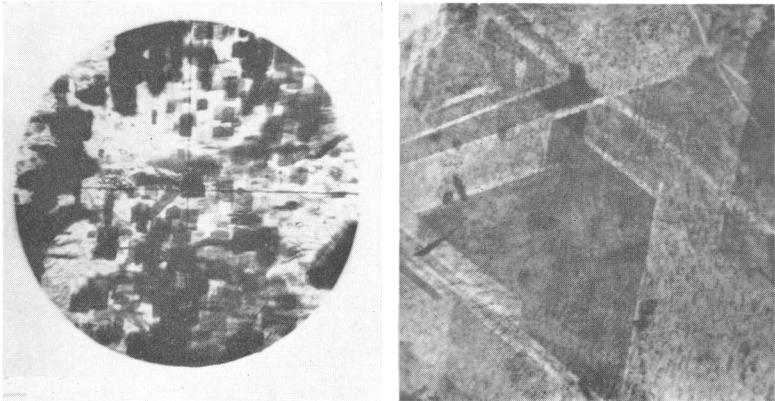
have a high tetrahedral charge and give more complete collapse than those minerals derived from non-micaceous material or chlorite. In the present study, the ground sample of the 14 Å weathered material was allowed to stand in 1N KOH solution for 18 hours after which the lattice showed partial collapse to 11.1 Å. According to Weaver, material derived from muscovite and some other micas should contract to about 10 Å after this treatment while those clays derived from biotite must be boiled in the K^+ solution for 15 minutes to 6 hours before contraction will take place. The latter, more drastic, treatment was applied for 6 hours to the vermiculite but again only partial collapse occurred with reflections at 14.3 Å and 11.6 Å. From these results it would appear that the original material ought to be non-micaceous in character although it is known to be phlogopite. Whether the phlogopite itself is primary in origin is, however, uncertain and this may have some effect upon the reaction.

The cation-exchange capacity of the weathered material was found to be about 150 me per 100 g, which is characteristic of vermiculite. A complete powder diffraction pattern of the ground sample (table I) shows good agreement with data by Walker (Brown, 1961) for vermiculite. The infra-red spectrum, however, suggests that the weathered material could be described as a hydrated phlogopite rather than as vermiculite since the pattern, though greatly altered compared with that of the unweathered material, shows a vibration with perpendicular orientation near 823 cm^{-1} as does phlogopite.

Schiller inclusions. A schiller effect is very marked in many of the mica crystals and seems to be caused by numerous inclusions lying parallel to the basal planes (fig. 3). They are retained in the weathered material and regardless of weathered state give the crystals a dark brown appearance. Their habit is tabular when viewed perpendicular to the main mica cleavage and they appear as thin parallel lines resembling brush marks in sections viewed parallel to the plane of the principal cleavage. These individual crystallite forms often show preferential orientation. According to Judd, 'the distribution of these inclusions is strikingly local, some parts of the mica crystals being almost entirely free from them, while in adjoining portions they have become so frequent as to entirely destroy the translucency of the crystal'.

For both phlogopite and vermiculite no significant difference could be readily detected between the powder diffraction or single-crystal photographs of samples with and without inclusions. It was hoped that the intensity ratio of the (004)/(005) phlogopite reflections, which is

sensitive to iron-magnesium substitution (Gower, 1957), might give some indication as to whether the inclusions are simply of iron-rich mica or not. Both powder diffraction and single-crystal photographs were used for this purpose. The relative intensities of adjacent 004 and 005 reflections were calculated from density measurements made with a recording microphotometer. Although the ratio consistently showed an increase for the material with inclusions, which is supposed



FIGS. 3 and 4. FIG. 3 (left). Inclusions in phlogopite that give rise to a schiller structure. Their preferential orientation is marked. $\times 197$. FIG. 4 (right). Electron-dense areas in phlogopite due to schiller inclusions showing a hexagonal form. $\times 13\ 900$.

to indicate an increase in iron content, the values obtained by the film method varied too greatly between the powder and single-crystal results to be considered reliable for direct comparison with those given by Gower, which were obtained using a diffractometer.

Ground flakes containing inclusions were examined by infra-red spectroscopy and showed no trace of biotite spectra. The pattern was on the whole similar to that for the fresh material without inclusions. In electron micrographs some flakes show electron-dense areas on the basal surfaces, which could represent the inclusion material in the sample, (fig. 4), but although these areas have crystallographically oriented boundaries no structural difference from the surrounding phlogopite was detectable in electron diffraction patterns.

Since X-ray, infra-red, and electron diffraction techniques yielded no structural information on the schiller inclusions it is concluded that the

difference is probably chemical and that the included material exists as thin films that show no evidence of individual crystallinity. A spectrochemical investigation of both major and minor trace elements showed that the only significant difference in the schillerized material lies in an increase in the iron content.

Single-crystal data. Having identified the starting material as phlogopite, and the weathered product as vermiculitic in character, by using single-crystal X-ray methods it was possible to make a more specific determination of the phlogopite polymorph form and to attempt to relate this to the structure of the secondary phase. Material selected from the weathered surface of the rock was also examined as it was assumed to represent an intermediate stage in the alteration process.

While setting up crystals of phlogopite for rotation about their *a*- or *b*-axes, it was found that at least 50 % of the specimens were anomalous in that the optic plane was normal to (010). The main polymorphic form was found to be 1M in type although some crystals did produce rather complex single-crystal patterns due to stacking faults or possibly twinning. The normal-beam equatorial Weissenberg photograph with rotation of the crystal about the *b*-axis is shown in plate VII, fig. *a*, and a list of reflections, as compiled from several photographs, and their observed intensities are given in table II. The relative intensities match those calculated values quoted from the results of Steinfink (1962) for a 1M phlogopite very well. Hendricks and Jefferson (1939) also observed a 1-layer phlogopite that was optically anomalous although Levinson and Heinrich (1954) have reported that this feature is more commonly characteristic of 2-layer types. Thus, although optical properties are useful for determining positions of principal axes, structural information is more diagnostic and fundamental.

In dealing with the weathered phlogopite flakes from the soil, it was found that the optic axial plane was usually normal to (010), an unusual feature for vermiculite and possibly related to the nature of the parent phlogopite. The normal-beam equatorial Weissenberg photograph for a *b*-axis rotation is shown in plate VII, fig. *b*; reflections and their observed intensities from several films are listed in table III. The relative intensities of $00l$, $20l$, and $40l$ reflections agree well with calculated data for Kenya vermiculite given by Mathieson and Walker (1954). The β angle is approximately 97° and since $l = 2n$ for $h0l$ reflections the cell contains a *c*-glide plane.

Examination of diffuse spectra with $02l$ indices was carried out on Weissenberg photographs of the equatorial layer rotated about the

TABLE II. Comparison of observed intensity distribution with calculated values (Steinfink, 1962) for 1M phlogopite

<i>hkl</i>	<i>I</i> _{est.}	<i>F</i> _{calc.}	<i>hkl</i>	<i>I</i> _{est.}	<i>F</i> _{calc.}	<i>hkl</i>	<i>I</i> _{est.}	<i>F</i> _{calc.}
001	—	-50	133	ms	80	336	w	-46
002	vw	-46	134	w	-39	337	w	47
003	vs	-120	135	m	115			
004	m	57	136	w	-43	331	vs	145
005	s	101				332	mw	-39
006	w	31	150	vvw	-23	333	—	—
007	m	-59	151	w	40	334	vvw	-34
008	ms	59	152	mw	62	335	w	59
009	vw	—				336	vvw	34
0.0.10	m	34	151	vvw	-23	337	—	0
			152	vvw	28	338	mw	-46
020	s	-58	153	vw	43	339	m	?
021	vvw	4						
022	vs	84	170	vvw	-24	350	—	-12
023	ms	58	171	—	—	351	w	26
024	—	—	172	vvw	29	352	vvw	26
025	—	-16						
026	vvw	21	171	—	—	351	vw	-25
027	w	34	172	w	42	352	vvw	17
			173	w	39	353	vw	51
040	ms	28				354	vw	27
041	ms	33	220	m	29			
042	w	31	221	mw	41			
043	vvw	-9	222	—	6	370	—	—
044	vw	-26	223	—	-29	371	vvw	25
045	vvw	11	224	—	—			
046	mw	48	225	vw	39	371	—	—
047	w	37	226	vw	51	372	—	—
						373	vvw	18
060	vs	144	221	m	26			
061	ms	-40	222	mw	37	200	vs	119
062	—	—				201	s	-97
063	w	-35	240	—	—	202	ms	84
064	ms	63	241	m	55	203	w	-38
065	w	32	242	mw	49	204	m	112
066	—	—				205	vw	-44
067	ms	-48	241	w	-28			
068	ms	46	242	—	6	201	m	57
			243	w	42	202	mw	-49
111	mw	35				203	mw	-60
112	m	87	260	s	83	204	m	73
113	vvw	32	261	ms	-72	205	vvw	48
114	—	-15	262	—	12	206	w	96
115	—	—	263	vvw	-25	207	mw	-112
116	vvw	30	264	mw	82	208	vvw	19
117	vvw	26				209	vvw	11
			261	m	53	2.0.10	vw	54
111	—	-25	262	—	—			
112	m	64	263	m	-62	400	—	—
113	m	78				401	vw	-56
			311	vvw	-26	402	—	23
130	mw	58	312	—	20	403	—	17
131	mw	-49	313	m	61	404	vvw	65
132	w	-63				405	vvw	-52
133	m	73	311	mw	38			
134	vvw	47				401	w	54
135	mw	9	330	mw	-42	402	mw	83
136	m	-111	331	—	—	403	mw	-68
137	vw	20	332	vvw	-31	404	—	14
			333	vw	61	405	vvw	-28
131	vs	121	334	vvw	32	406	w	82
132	s	-97	335	—	—			

TABLE III. Comparison of some observed intensity values with calculated values by Mathieson and Walker (1954) for Kenya vermiculite. * See text, p. 160.

<i>hkl</i>	<i>I</i> _{est.}	<i>F</i> _{calc.}	<i>hkl</i>	<i>I</i> _{est.}	<i>F</i> _{calc.}	<i>hkl</i>	<i>I</i> _{est.}	<i>F</i> _{calc.}
002	—	298	111	vs		130	—	18
004	—	78	113	w		132*	vs	169
006	ms	-93	115	ms		134	—	49
008	s	163	117	m		136	w	74
0.0.10	vs	336	119	—		138	mw	180
0.0.12	mw	21	1.1.11	w		1.3.10	—	40
0.0.14	m	-114				1.3.12	—	-40
0.0.16	w	-76	11 $\bar{1}$	s		1.3.14	m	157
0.0.18	vw	4	11 $\bar{5}$	ms		1.3.16	ms	196
0.0.20	s	224	117	w		1.3.18	s	271
0.0.22	w	69	119	w				
0.0.24	m	150				132*	s	130
0.0.26	vwv	41	311	w		134	vs	210
0.0.28	mw	88	313	mw		136	vs	298
0.0.30	mw	85				138	m	82
			31 $\bar{1}$	ms		1.3.10	m	187
200	ms	127	31 $\bar{3}$	ms		1.3.12	w	92
202	s	212	315	w		1.3.14	ms	259
204	vs	300	317	mw		1.3.16	m	171
206	m	81	319	—		1.3.18	—	24
208	ms	189	3.1. $\bar{1}\bar{1}$	vw		1.3.20*	vwv	-45
2.0.10	mw	87				1.3.22*	vwv	-58
2.0.12	ms	257	021	vs	54	1.3.24*	vw	109
2.0.14	mw	173	022	—	111	1.3.26*	w	89
2.0.16	—	23	023	vw	-25			
2.0.18	w	-48	024	—	56	330	s	140
2.0.20	vw	-57	025	m	-61	332	mw	104
2.0.22	w	109	026	—	-67	334	—	-8
2.0.24	mw	87	027	—	-11	336	vw	50
			028	—	-81	338	w	134
20 $\bar{2}$	—	20	029	w	43	3.3.10	vw	60
204	s	173	0.2.10	—	10	3.3.12	—	7
206	—	51	0.2.11	vw	30	3.3.14	vw	82
208	mw	72				3.3.16	—	19
2.0.10	m	175	221	mw		3.3.18	mw	119
2.0.12	vw	41	223	—		3.3.20	mw	115
2.0.14	vw	-40	225	m		3.3.22	w	75
2.0.16	mw	163	227	—				
2.0.18	m	203	229	mw		33 $\bar{2}$	vs	304
2.0.20	s	281	2.2.11	vwv		334*	s	140
2.0.22	vw	37	2.2.13	vw		336	mw	102
2.0.24	—	1	2.2.15	vw		338	—	-5
2.0.26	—	30	2.2.17	vwv		3.3.10	vwv	53
2.0.28	w	100	2.2.19	vwv		3.3.12*	mw	143
2.0.30	w	98				3.3.14	vwv	59
			22 $\bar{3}$	mw		3.3.16	—	4
400	m	-126	22 $\bar{5}$	vw		3.3.18	vwv	-88
402	w	94	227	w		3.3.20	—	24
404	s	134	229	vwv		3.3.22	mw	135
406	w	81	2.2.11	vw		3.3.24	mw	122
408	vwv	-33	2.2.13	—		3.3.26	w	79
4.0.10	—	-8	2.2.15	vwv				
4.0.12	mw	152	2.2.17	—		530	m	130
4.0.14	mw	143	2.2.19	vwv		532	m	134
4.0.16	mw	142				534	vwv	40
			421	w		536	—	27
40 $\bar{2}$	mw	-69	423	mw		538	w	51
404	m	122	425	mw		5.3.10	m	133
406	s	223	427	—				
408	ms	178	429	vwv		53 $\bar{2}$	w	34
4.0.10	—	-30				534	—	-4
4.0.12	w	73	42 $\bar{1}$	ms		536	vw	-47
4.0.14	m	132	42 $\bar{3}$	—		538	—	58
4.0.16	m	206	425	m		5.3.10	w	53
4.0.18	w	77	427	vwv		5.3.12	w	70

a -axis and the second layer rotated about the b -axis. Doubling of the c -axis value to approximately 29 Å was indicated as found earlier by Gruner (1934), Hendricks and Jefferson (1938), and Mathieson and Walker (1954) for their vermiculite structures. The diffraction maxima intensities for these reflections do not, however, agree with those recorded for Kenya vermiculite by Mathieson and Walker in that odd, instead of even, values of l give the most intense diffraction peaks. The stacking sequence of the silicate layers has an important effect upon these reflections and since a c -glide operation can be produced in four different ways, calculation of structural amplitudes is necessary to determine the exact sequence. Even then it may be difficult, as Mathieson and Walker could only reconcile their diffuse spectra by postulating the occurrence of two equally probable stacking orders.

Apart from differences in the intensities of the $02l$ reflections as compared with those values given by Mathieson and Walker, several reflections (marked in table III with an asterisk) showed better agreement with the observed intensities quoted by these authors than with their calculated figures. On the whole, however, the weathered material has a vermiculitic C -face centred structure.

X-ray examination of individual flakes of mica from the surface of the weathered rock fragments showed that they are intermediate in that they consist of various proportions of both 14 Å and 10 Å phases. In all the specimens examined the two components exist as discrete structural units with no evidence of interstratification either regular or random. The two phases exist in a fixed orientation with respect to each other as shown in plate VII, fig. c —a normal-beam equatorial Weissenberg photograph with b -axis rotation. The $+a$ - and $+b$ -axial directions are common, while there is a 3° angle between the $+c$ -axes due to the differences in β values— 100° for phlogopite and 97° for vermiculite. The weathering of trioctahedral mica to vermiculite-like products has been noted in the past by Lacroix (1940) and Walker (1949). The mechanism that produces the vermiculite phase involves the replacement of K^+ ions by water molecules and emplacement of Mg^{2+} ions in the interlayer positions. In order to reduce the attractive forces between the layers sufficiently to allow penetration of a double layer of water, changes in the lattice charges must be effected by processes such as lattice substitution and possibly oxidation of any iron present. According to Walker, there exists an intermediate stage in the weathering process when mixed-layer structures exist but in the present study diffraction maxima between 10 Å and 14 Å were never detected.

While examining some material containing schiller inclusions by single-crystal methods, an additional set of reflections other than those belonging to phlogopite were identified as belonging to the spinel, chromite. Although the presence of spinel was previously noted, the position of the reflections indicates a preferred structural orientation with respect to the mica. The relationship is illustrated by stereographic projection in fig. 5. The equivalent spacings of the two structures may be compared

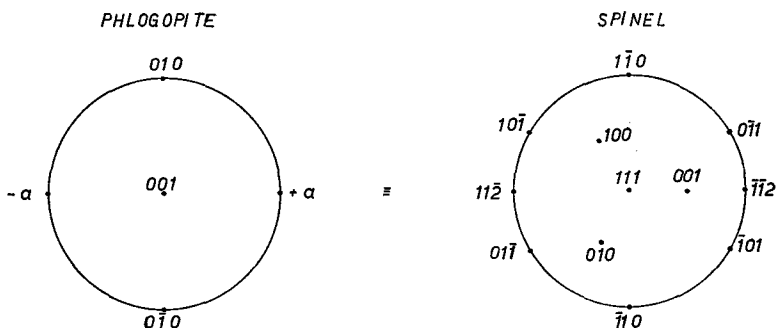


FIG. 5. Orientation relationship between phlogopite and spinel in stereographic projection.

as follows: phlogopite $d_{(001)} = 10.1 \text{ \AA}$, spinel $d_{(111)} = \frac{1}{2} \times 9.7 \text{ \AA}$; phlogopite $a = 5.3 \text{ \AA}$, spinel $d_{(224)} = \frac{1}{3} \times 5.1 \text{ \AA}$; phlogopite $d_{(010)} = 9.2 \text{ \AA}$, spinel $d_{(220)} = \frac{1}{3} \times 8.82 \text{ \AA}$.

For these minerals the planes of close-packed oxygens are in parallel alignment just as was found in the alteration of brucite to periclase by Ball and Taylor (1961), who postulated that in the transformation from hexagonal close-packing in brucite to cubic close-packing in periclase oxygen layers had to slip over each other. In the present instance, which also involves the same two types of close-packing, there is no evidence to suggest that any transformation process is involved in the orientation relationship, although it does seem that a preferential position is adopted by one phase with respect to another during crystallization.

Discussion

The mechanism of transition from phlogopite to vermiculite by weathering is of interest. The structures are similar in that they both have 'talc' sheets. However, in phlogopite, these sheets are bonded together by K atoms with twelve-fold coordination whereas in vermiculite there exists between the 'talc' layers a brucite-like structure 'with

incomplete occupancy of the brucite atomic positions by Mg and H_2O ' (Brown and Bailey, 1962). The change from a 1-layer to a 2-layer polymorph, such as occurs in the present instance, necessitates an alteration in the stacking arrangement of the layers. By employing the results of investigations by previous workers, with respect to possible stacking sequences in chlorites and vermiculites, it was hoped that more information could be gained on structural movements brought about during weathering.

The $h0l$ intensities of chlorite have been used by Brown and Bailey to determine the stacking relation between 'brucite' and 'talc' sheets of one unit cell in the structure. They give intensity distribution data for four main types of stacking produced in polytypes with $\beta = 97^\circ$ and have identified a number of vermiculite specimens as belonging to one of these types—Ia. Exact comparison of data is not feasible for the Scotscaider sample but, in general, closer agreement is found with the Ia stacking arrangement than with the other three. This means that the exchangeable cations of the 'brucite' layer are situated over hydroxyls of the nearest 'talc' silicate sheet while the water molecules of the lower part of the 'brucite' unit are placed directly over the octahedral cations of the 'talc' layer. Such an arrangement is the same as that for Kenya vermiculite determined by Mathieson and Walker.

The polytype of the complete structure is determined by the position of the repeating 'talc' sheet with respect to a fixed initial 'talc' sheet since it determines the direction of the c -axis of the resultant structure as well as the c repeat distance. Brown and Bailey have shown that for all 1-layer regular polymorphs of chlorite it is necessary that the directions of stagger at the octahedral junctions of the individual 'talc' layers are parallel to each other throughout the structure. Also, interlayer shifts of $a/3$ take place in one of six possible directions 60° apart with corresponding effects upon the space group symmetry.

For 2-layer polymorphs such as occur in the vermiculite structure it is not essential that the octahedral stagger should operate for each 'talc' layer in the same direction nor that the hexagon of surface oxygens of one 'talc' layer be superimposed exactly upon that of another as in 1M phlogopite, especially when the octahedral stagger is in the same direction for each layer, for if this were so there would be no c -glide operation and the resultant symmetry would be $C2/m$.

In Kenya vermiculite, a study of the intensity distribution of diffuse $02l$ reflections (Mathieson and Walker 1954) showed that the stacking of the 'talc' sheets with respect to each other corresponded to a sequence in

which the octahedral stagger alternated by 120° in direction between one unit and the next, i.e. M and N type layers according to the designation of Brindley, Oughton, and Robinson (1950). In addition, displacement of the N layers by $b/3$ and $2b/3$ with respect to M layers was indicated since the positional average of these displacements gave calculated F^2 values that agreed best with the observed intensities. The structure of Kenya vermiculite has been referred to by Brown and Bailey as a 'somewhat irregular alternation of Ia-4 and Ia-6 layers'.

For the Scotscladder sample, although no absolute measurement of intensities has been attempted for the diffuse 02l diffraction maxima, the observed values suggest that the stacking sequence of 'talc' sheets with respect to each other must differ from that of Kenya vermiculite. Since the Scotscladder vermiculite has been derived directly from a 1M phlogopite, one would expect a mechanism of weathering in which the movement of the 'talc' sheets away from the 1M type of stacking would be as little as possible. Layers in which the directions of octahedral stagger are retained in parallel orientation may be shifted $b/3$ or $2b/3$ with respect to each other, to accommodate the introduction of a 'brucite'-type sheet and at the same time introduce a c -glide operation. This 's' stacking sequence is one of those proposed as possible for vermiculite by Mathieson and Walker, but further detailed structural calculations would be required to find out whether the intensity distribution arising from such a structure would agree with that observed for the 02l reflections of the Scotscladder sample.

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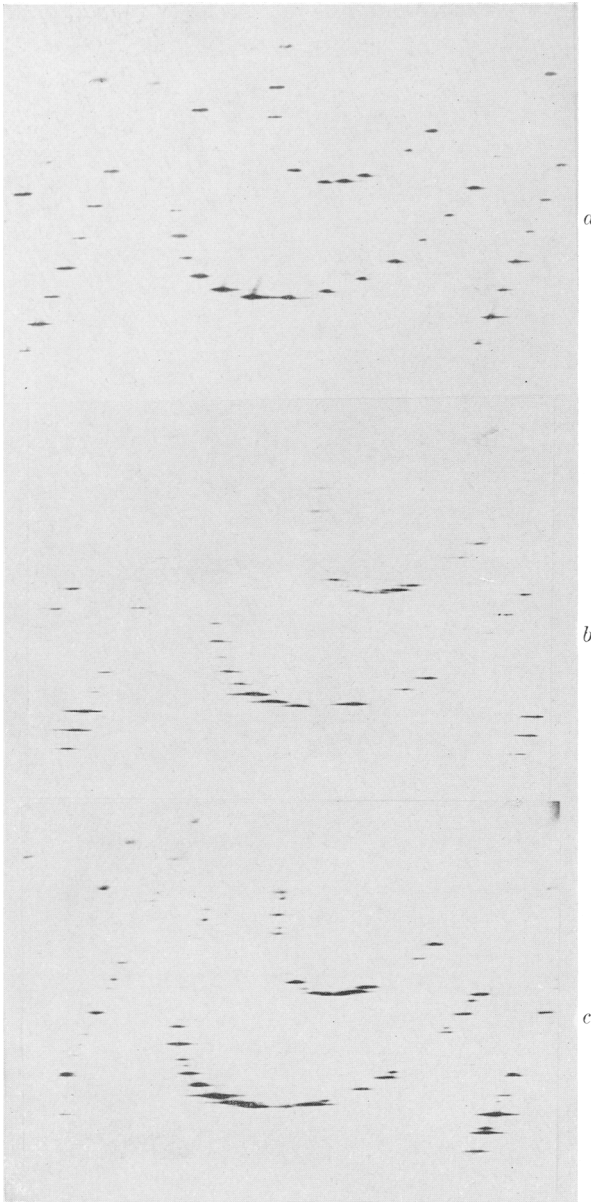
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EXPLANATION OF PLATE VII

- FIG. *a*. Normal-beam equatorial Weissenberg photograph of phlogopite, 1M polymorph, rotated about the *b*-axis.
- FIG. *b*. Normal-beam equatorial Weissenberg photograph of weathered phlogopite from the soil, rotated about the *b*-axis. The mineral is vermiculitic.
- FIG. *c*. Normal-beam equatorial Weissenberg photograph of weathered phlogopite from the rock, rotated about the *b*-axis. The orientation relationship between the phlogopite and vermiculite phases is shown.

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