# STUDIES IN THE MICA GROUP; SINGLE CRYSTAL DATA ON PHLOGOPITES, BIOTITES AND MANGANOPHYLLITES

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Abstract

Weissenberg studies of about 250 specimens of phlogopite, biotite and manganophyllite, of which 63 have been chemically analyzed, indicate: (1) in phlogopites there is no discernible relationship between structure and either composition or paragenesis; (2) in biotites there is no relationship between structure and composition, but geologic environment may be a factor in determining the polymorphic type; (3) the optic plane of all 2-layer polymorphs is normal to (010), as is also the case for the biotite variety, anomite; and (4) 1-layer polymorphs nearly always have the optic plane parallel with (010), which corresponds with the orientation of the biotite variety, meroxene.

## Introduction

Systematic variations between composition and the various polymorphs in the muscovite-lepidolite series have been demonstrated by Levinson (1953). On the basis of these results it seemed desirable to investigate the biotite-phlogopite series in a similar manner in the hope of gaining insight into causes of the complex polymorphism known to exist in these micas. The only other Weissenberg study of these micas known to the writers is in the work of Hendricks and Jefferson (1939), predominantly on unanalyzed specimens.

We have obtained over 60 analyzed specimens of biotite, phlogopite and manganophyllite (which we consider as manganoan phlogopite), most of which have been described by several investigators. In addition we have studied a suite of approximately 200 unanalyzed biotites and phlogopites from geologically well-studied occurrences. All specimens were studied by means of the Weissenberg method using copper radiation. In most cases only zero level, *a*-axis photographs were taken, as these are sufficient to determine the layer periodicity. In many cases, however, upper level photographs were taken, which confirmed the structure identification based on the zero level, *a*-axis photographs. In agreement with Hendricks and Jefferson (1939, p. 762), the 3-layer polymorph is considered to be hexagonal (actually trigonal) as a limiting case, even though some specimens are not truly uniaxial. Orientation of the flakes was secured by optical methods in those cases in which 2V was approximately 10° or greater. Laue photographs and percussion figures were

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TABLE 1. STRUCTURES OF ANALYZED PHLOGOPITES

| Number | Reference | Structure | Occurrence | FeO | $Fe_2O_3$ | MgO | $TiO_2$ | F |
|---|---|---|---|---|---|---|---|---|
| 1054 | Jakob (1938) | ? Very diffuse scattering | from Morawitza, Czechoslovakia | 0.64 | 1.05 | 26.77 | 0.10 | 0.00 |
| 1055 | Jakob (1931) No. 55 | 1-layer (mod. scattering) | mica peridotite, Italy | 2.52 | 0.00 | 25.45 | 2.80 | 0.00 |
| 1056 | Jakob and Parga-Pondal (1932) No. 73 | 1-layer | Burgess, Ontario[1] | 1.24 | 0.73 | 26.45 | 1.15 | 1.00 |
| 1057 | No. 70 | 1-layer | Rossie, N. Y.[2] | 4.79 | 0.00 | 22.30 | 4.07 | 0.12 |
| 1058 | No. 69 | 1-layer | Hull, Quebec[1] | 2.49 | 0.71 | 24.60 | 1.74 | 2.04 |
| 1059 | No. 68 | 1-layer | Burgess, Ontario[1] | 0.30 | 0.00 | 27.32 | 0.39 | 6.74 |
| 1060 | No. 67 | 1-layer | Burgess, Ontario[1] | 1.50 | 0.00 | 26.14 | 0.86 | 2.37 |
| 1061 | No. 66 | 1-layer | Isolo, Madagascar[3] | 3.28 | 0.00 | 25.29 | 2.19 | 1.15 |
| 1062 | No. 65 | 2-layer (mod. scattering) | Ampandrandara, Madagascar[3] | 2.96 | 1.18 | 23.40 | 1.69 | 0.68 |
| 1063 | No. 64 | 1-layer | Mandridano, Madag.[3] | 2.09 | 0.00 | 24.48 | 1.64 | 0.08 |
| 1064 | No. 63 | 2-layer | Saharakara, Madag.[3] | 2.79 | 1.68 | 23.78 | 1.11 | 0.56 |
| 1065 | No. 62 | 1-layer | Ambatoabo, Madag.[3] | 1.42 | 0.66 | 24.80 | 0.86 | 0.86 |
| 1066 | Jakob (1928) No. 19 | 1-layer (heavy scattering) | Simplon Tunnel, Switzerland | 0.00 | 1.71 | 24.79 | 0.39 | 0.00 |
| 1067 | Jakob (1928) No. 20 | 1-layer | dolomite-Tessin | 0.00 | 1.31 | 25.81 | 0.83 | 0.00 |
| 1068 | Jakob (1928) No. 22 | 1-layer | contact metamorphic S. W. Africa | 2.72 | 0.97 | 25.05 | 0.66 | 0.58 |
| 1069 | Jakob (1928) No. 23 | 1-layer | pneumatolytic, Vesuvius, Italy | 0.58 | 1.92 | 28.18 | 1.27 | — |
| 1071 | Jakob—not published | 1-layer | Skräbböle, Pargas, Finland[4] | 0.71 | 1.01 | 27.80 | 0.12 | 2.11 |
| 1072 | Jakob—not published | 1-layer | Pargas, Finland[4] | 5.59 | 1.54 | 22.00 | 1.41 | 2.80 |
| 1073 | Jakob—not published | 1-layer | Patteby, Pargas, Finland[4] | 1.47 | 1.29 | 26.16 | 0.33 | 1.87 |
| 1074 | Jakob—not published | 1-layer | Ontala, Pargas,[4] Finland | 1.68 | 1.87 | 25.91 | 0.68 | 1.30 |
| 1075 | Jakob—not published | 1-layer | Skräbböle, Pargas, Finland[4] | 0.59 | 1.75 | 27.22 | 0.10 | 0.48 |
| 949 | Dana (1892) Anal. 12 p. 633 | 1-layer | Rossie, N. Y.[2] | 7.62 | 1.12 | 21.47 | 1.16 | 4.00 |
| 1135 | Pagliani (1940) | 1-layer | marble, Italy | 1.55 | 2.65 | 27.62 | 2.83 | — |
| 1139 | Hutton and Seeyle (1947) | 1-layer | marble, New Zealand | 2.38 | 0.43 | 22.95 | 0.82 | 0.62 |
| 1252 | Pieruccini (1950) | 1-layer | pneumatolytic, Mt. Somma, Italy | 7.89 | tr. | 15.66 | 0.33 | 2.57 |
| 1261 | {(*a*) (*b*) (*c*) Mauguin (1928)} | {1-layer 3-layer} | {Ambatoabo, Madagascar.[3]} | 2.30 | n.d. | 24.42 | 0.74 | n.d. |
| 730 | Prider (1939) | 1-layer | leucite lamproite,[5] West. Australia | 3.75 | 2.18 | 19.66 | 8.97 | 0.66 |
| 1325 | Cross (1897) | 1-layer | in wyomingite,[5] Leucite Hills, Wyo. | 0.90 | 2.73 | 22.40 | 2.09 | 2.46 |

(1) The Quebec and Ontario phlogopites occur in pegmatitic bodies associated with pyroxenite and marble.
(2) The occurrences near Rossie, St. Lawrence, N. Y., are in marble.
(3) The Madagascar deposits are similar to the Canadian type.
(4) The occurrences at Pargas are in marble.
(5) Of pyrogenic origin.

used in the majority of the cases; for the most part the percussion figure method was found to be unreliable.

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## *X*-RAY STUDIES OF PHLOGOPITE

The structures of approximately 80 phlogopite crystals from about 50 different specimens (28 chemically analyzed) were identified. All but five have crystallized with the 1-layer monoclinic structure. Of these five, three have crystallized as the 2-layer monoclinic polymorph, whereas the remaining two have crystallized with the 3-layer structure.

2-layer phlogopites:
- No. 1062 (*a* and *b*)—Madagascar; see Table 1 for analysis and reference.
- No. 1064 (*a* and *b*)—Madagascar; see Table 1 for analysis and reference.
- No. 1220 (*a*) —Hull, Quebec; not analyzed.

3-layer phlogopites:
- No. 1261 (*b* and *c*)—Madagascar; see Table 1 for analysis and reference.
- No. 1227 (*a*) —Labelle Co., Quebec; not analyzed.

It is noteworthy that three of the non 1-layer types are from Madagascar, but four other specimens of Madagascar phlogopite crystallized with the common 1-layer monoclinic structure (Table 1). The analyzed specimen (No. 1261) from Prof. Mauguin consisted of two thin cleavage fragments; specimens No. 1261 (*b* and *c*) *x*-rayed from one of these have the 3-layer structure, whereas No. 1261 (*a*) from the second fragment has the 1-layer structure. The optical properties of the *x*-rayed crystals are similar (all essentially uniaxial), and there is probably little difference in their composition. This appears to be analogous to the association of 1- and 3-layer lepidolites as described by Levinson (1953). The specimens of known composition that have crystallized as the 2- and 3-layer forms do not appear to be markedly different, chemically, from the 1-layer types. In fact, the compositions of all the Madagascar varieties are practically identical. Optically, however, the 2-layer varieties differ in having their optic planes normal to (010); all 1-layer forms have their optic planes parallel with (010).

These findings are in qualitative agreement with those of Hendricks and Jefferson (1939), who found 14 phlogopites with the 1-layer monoclinic structure and one each with the 2- and 3-layer forms. No new polymorphs of phlogopite have been found in our investigations. Crystals reported by Pieruccini (1950) to be triclinic on the basis of goniometric measurements were isolated from a type specimen supplied by him. Weissenberg photographs indicate that the structure is the common 1-layer monoclinic form (Table 1). However, written communication with Professor Pieruccini indicated that two distinct varieties of phlogopite are to be found in his specimens, and it is possible, therefore, that we did not *x*-ray the crystals he believes to be triclinic.

The unanalyzed specimens are from widely scattered localities but are predominantly from pegmatites in Canada, and from marbles or contact metamorphic deposits in New York and to a lesser extent in Montana and Finland.

## Relation of Chemistry, Structure and Paragenesis

On the basis of the data in Table 1 an attempt was made to correlate polymorphism with chemistry and paragenesis in phlogopite but the results were negative. The fact that all three polymorphic modifications of phlogopite occur among the Madagascar specimens, may well rule out the possibility of temperature or pressure as a major factor in determining lattice type.

## *X*-ray Studies of Manganophyllite

The only *x*-ray work reported for manganophyllite has been that of Hendricks and Jefferson (1939), who note a manganophyllite from Rajsberg, Sweden, with a 2-layer monoclinic structure. We have found that all the Swedish manganophyllites described by Jakob (1925) have the 1-layer monoclinic structure. Of the six unanalyzed specimens studied, only one from Franklin, N. J. (No. 1358) has a 2-layer structure; we found no examples of the 3-layer structure. The Swedish manganophyllites that have a very small 2V (particularly Jakob, 1925, Nos. 1 and 5, which are from Varmland) appear to approach the 3-layer hexagonal structure, owing to an intimate "twinning" on (001) of thin sheets, similar to that described for uniaxial lepidolites (Levinson, 1953). From the chemical analyses of these micas there is no correlation between composition and polymorphism. Manganophyllites with the 1-layer form show extreme ranges in Fe and Mn (for example, $Fe_2O_3 = 0.00$ to 16.94%, see Table 2).

TABLE 2. STRUCTURES OF ANALYZED MANGOPHYLLITES

| Michigan Number | Reference: Jakob (1925) | FeO | $Fe_2O_3$ | MgO | MnO | $Mn_2O_3$ | $TiO_2$ |
|---|---|---|---|---|---|---|---|
| 1076 | No. 8 | 0.00 | 16.94 | 26.79 | 4.52 | 0.00 | 0.00 |
| 1077 | No. 7 | 2.54 | 0.00 | 29.22 | Tr. | Tr. | 0.00 |
| 1078 | No. 6 | 0.00 | 3.95 | 22.60 | 0.00 | 8.30 | Tr. |
| 1079 | No. 5 | 0.00 | 4.68 | 21.18 | 9.25 | 2.96 | 0.00 |
| 1080 | No. 4 | 0.00 | 2.97 | 24.80 | 0.00 | 5.77 | 0.55 |
| 1081 | No. 3 | 0.00 | 2.68 | 26.65 | 0.00 | 4.07 | 0.41 |
| 1082 | No. 2 | 0.00 | 0.91 | 29.28 | 0.00 | 3.92 | 0.00 |
| 1083 | No. 1 | 0.00 | 2.81 | 27.87 | 0.00 | 4.93 | 0.22 |

All have the 1-layer structure and are from Långban and Varmland, Sweden, but apparently are of several different paragenetic types.

The most significant observation concerning the optical properties of the studied manganophyllites is the fact that several of Jakob's (1925) specimens (Numbers 3, 4, and 6) have the optic plane normal to (010). Nearly all micas crystallizing as the 1-layer polymorph have the optic plane parallel with (010). The orientations of these micas were determined by *x*-ray methods; the cause of the anomalies has not been determined.

## X-RAY STUDIES OF BIOTITE

The results of the *x*-ray studies of analyzed biotites are in Table 3. Among the biotites there is a significantly larger number of specimens with multiple layer periodicities. During this study, however, neither the 6- nor 24-layer triclinic biotites described by Hendricks and Jefferson (1939) have been found. Zero-level Weissenberg photographs taken about one of the pseudo *a*-axes of the 2-layer monoclinic polymorph appear to be identical with the zero-level, *a*-axis photograph of the 6-layer triclinic biotite illustrated by Hendricks and Jefferson (1939, p. 745). Likewise, zero-level pseudo *a*-axis photographs of the 1-layer monoclinic polymorph strongly resemble zero-level, *b*-axis photographs of the 6-layer monoclinic polymorph reported only in lepidolites. Therefore extreme caution had to be exercised in deciding the structure of these micas from zero-level photographs. Diffuse scattering, particularly among the biotites, also made structure determinations difficult in several instances.

The structures of several uncommon biotite varieties (not analyzed) also were determined (Table 4).

## RELATIONSHIP BETWEEN CHEMISTRY, STRUCTURE AND PARAGENESIS

As is the case of the phlogopites, attempts to correlate structure with

TABLE 3. STRUCTURES OF ANALYZED BIOTITES

| Number | Reference | Structure | Paragenesis | FeO | $Fe_2O_3$ | MgO | $TiO_2$ |
|---|---|---|---|---|---|---|---|
| 728 | Glass (1935) | 3-layer | pegmatite, Amelia, Va. | 26.72 | 2.87 | n.d. | 3.60 |
| 729 | Stevens (1946) | 1-layer | pegmatite, Ridgeway, Va. | 8.96 | 3.31 | 16.15 | 1.11 |
| 797 | Grout (1924) No. 1 | 1-layer (+3-layer ?) | basic segreg. in Kekequabic granite, Minn. | 7.72 | 7.44 | 16.55 | 1.67 |
| 798 | Grout (1924) No. 2 | 3-layer (mod. scattering) | Vermillion granite, Minn. | 14.80 | 4.05 | 10.21 | 2.23 |
| 799 | Grout (1924) No. 3 | 2-layer | granite, Mora, Minn. | 23.23 | 3.03 | 9.24 | 3.32 |
| 800 | Grout (1924) No. 4 | 1-layer | granite, Rockville, Minn. | 23.75 | 1.14 | 6.16 | 2.73 |
| 801 | Grout (1924) No. 5 | 1-layer | peridotite, base Duluth gabbro, Minn. | 12.96 | 8.63 | n.d. | 4.34 |
| 994 | van Biljon (1940) | 2-layer | granite, Namaqualand, S. Af. | 13.58 | 4.23 | 12.16 | 3.89 |
| 960 | Pagliani (1949) | 2-layer | mica schist, Beura, Italy | 9.10 | 5.10 | 10.44 | 0.56 |
| 1001 | Inoue (1950) | 1-layer | nepheline syenite* | 19.9 | 4.53 | 6.26 | 1.10 |
| 1002 | Inoue (1950) | 1-layer (very heavy scatt.) | nepheline syenite* | 21.94 | 8.53 | 5.32 | 1.97 |
| 1003 | Inoue (1950) | 1-layer | nepheline syenite* | 21.02 | 12.45 | 4.19 | 1.04 |
| 1004 | Inoue (1950) | 1-layer (mod. scattering) | cancrinite syenite pegmatite* | 16.03 | 20.22 | 1.37 | 0.70 |
| 1010 | Kawano (1933) | 1-layer | metamorphosed xenolith, Minederayama, Japan | 16.38 | 3.28 | 8.99 | 2.45 |
| 1011 | Kawano (1942) | 1-layer | lepidomelane-quartz-fels, Minederayama, Japan | 23.27 | 7.81 | 4.32 | 2.42 |
| 1084 | Jakob (1931) No. 57 | 2-layer (weak scattering) | 2-mica pegmatite, Monti di Daro, Bellinzona, Switz. | 10.47 | 4.09 | 13.19 | 2.06 |
| 1085 | Jakob (1931) No. 58 | 2-layer | 2-mica pegmatite, Monti di Darò, Bellinzona, Switz. | 9.72 | 2.24 | 14.22 | 1.64 |
| 1086 | Jakob (1931) No. 59 | (3-layer ?) | 2-mica pegmatite, Claro, Tessin, Switz. | 16.26 | 4.03 | 8.46 | 3.16 |
| 1087 | Jakob (1931) No. 60 | 2-layer (weak scattering) | 2-mica pegmatite Claro, Tessin, Switz. | 16.85 | 4.08 | 8.06 | 2.71 |
| 1088 | Jakob (1931) No. 61 | 1-layer | lamprophyre, Gotthard, Switz. | 15.84 | 5.03 | 11.17 | 1.95 |
| 1089 | Jakob (1937) | 1-layer | feldspar pegmatite, Derome, Halland, Sweden | 28.61 | 0.00 | 6.72 | 1.99 |
| 1117 | Coats and Fahey (1944) | 1-layer (mod. scattering) | (siderophyllite) pegmatite, Brooks Mtn., Alaska | 30.16 | Tr. | 0.22 | 0.02 |
| 1140 | C. O. Hutton (not published) | 2-layer | from sands derived from granitic rocks, Calif. | 14.49 | 9.30 | 5.80 | 3.47 |
| 1145 | Hallimond (1947) | 1-layer | marble, Tiree, Hebrides, Gr. Brit. | 6.8 | 1.1 | 18.7 | 1.9 |
| 1257 | Hutton and Seeyle (1947) | 2-layer | pegmatite-like lens in gneiss, Old Pt., Charles Sound, New Zealand | 14.41 | 3.92 | 11.11 | 3.02 |
| 1262 | Mauguin (1928) | 1-layer (heavy scattering) | from Tschebarkul, Ural Mtns., U.S.S.R. | 12.77 | 7.09 | 13.30 | 1.61 |
| 1350 | Yamada and Sugiura (1950) | 1-layer (weak scattering) | pegmatite, Motomiya, Korea | 3.49 | 0.67 | 24.24 | 0.64 |

*Fuku-Shinzan, Korea

TABLE 4. STRUCTURES OF BIOTITE VARIETIES

| Number | Varietal name | Locality | Structure |
|---|---|---|---|
| 732(*a*) | waddoite | Isle of Waddo | 1-layer |
| 746(*a*) | lepidomelane (pterolite) | Brevig, Norway | 2-layer |
| 752(*a*) | meroxene | Mt. Vesuvius | 1-layer |
| 753(*a*) | meroxene (colorless) | Mt. Vesuvius | 3-layer |
| 759(*a*) | meroxene | Mendham, N. J. | 1-layer |
| 754(*a*) | calciobiotite | Italy | 1-layer |
| 770(*a*) | annite | Rockport, Mass. | 1-layer |
| 1116(*a*) | annite | Rockport, Mass. | 1-layer |
| 775(*a*) | siderophyllite | Pikes Peak, Colo. | 1-layer |
| 1117(*a*) | siderophyllite | Brooks Mt., Alaska | 1-layer (mod. scattering) |
| 783(*a*) | cryophyllite | Rockport, Mass. | 1-layer |
| 1112(*a*) | monrepite | Vibora, Finland | 1-layer |
| 1113(*a*) | eukamptite | Presburg, Hungary | 1-layer |
| 1114(*a*) | bastonite | Bastogne, Belgium | 2-layer |

chemistry have been unsuccessful and for the most part structural-paragenesis correlations are likewise not evident. However, some generalizations are available with regard to the geographic and geologic distribution of various polymorphs in selected pegmatite districts.

About 80 biotite specimens from about 15 pegmatite deposits in the southeastern United States were studied. Fifty have crystallized as the 2-layer monoclinic polymorph, 15 as the 3-layer hexagonal polymorph, but only one as the 1-layer monoclinic polymorph. Diffuse scattering, which is characteristic of biotites from this area, and the presence of mixed structures prevented accurate structural determination of the remainder. The results of studies from other scattered pegmatite districts are:

Southern Norway—predominantly 1-layer monoclinic forms, but 2-layer forms common.
Canada: Bancroft and Wilberforce Districts—predominantly 1-layer forms.
Colorado: Guffey District—predominantly 1-layer forms.

Geological environment may play an important role in guiding the crystallization of biotite micas. Pegmatites of the southeastern United States (North Carolina, particularly) are chiefly of quartz dioritic or granodioritic composition, whereas those of the Bancroft area, for example, are nepheline syenitic. It seems possible that biotites crystallizing from magmas of such widely different compositions might have structures related to their particular crystallization environments or source magmas, but further investigation is required to test this hypothesis. As mentioned above, these factors do not themselves uniquely account for the layer types. It was hoped that a knowledge of the temperature of crystallization of these micas might be useful and for this reason a suite of biotite phenocrysts from hypabyssal acid dike rocks from the Guffey District, Colorado, was studied. However, both 1- and 2-layer polymorphs were found in these biotites.

## Position of Optic Plane

Among the studied 2-layer biotites and phlogopites the position of the optic plane is invariably normal to (010). In this respect the 2-layer octophyllite type micas are similar to the 2-layer muscovite types, which have been shown by Hendricks and Jefferson (1939) and Levinson (1953) to have their optic planes normal to (010). All the investigated dark colored micas that have crystallized as the 1-layer polymorph, with the exception of the three anomalous manganophyllites described above, have their optic planes parallel with (010). Hendricks and Jefferson (1939) also observed a 1-layer phlogopite similarly anomalous, i.e., with its optic plane normal to (010).

There has been considerable discussion as to the origin and abundance of the biotite varieties, meroxene (optic plane parallel with (010)) and anomite (optic plane normal to (010)). It has been noted that anomite is relatively rare, whereas meroxene is more common (Winchell and Winchell, 1951). These observations appear to be correct, i.e., the 2-layer polymorph with optic plane normal to (010) (anomite) is less common than 1-layer forms, but it is by no means rare. The cause of this difference in optic orientation is inherent in the various polymorphs, so that the term anomite is synonymous with 2-layer biotite and meroxene with 1-layer biotite. Since the structural characteristics are more fundamental than the optical characteristics, the varietal terms anomite and meroxene probably should be discarded. Furthermore, most of the older descriptions of anomite and meroxene were based on orientation by means of the longest ray of percussion figures. In this study, after the position of (010) has been determined by $x$-ray method, we found that the percussion orientation method was unreliable in about one-third of the trials.

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