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NATURAL MICA STUDIES

By

E. WM. HEINRICH

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INTRODUCTION

For the past nine months emphasis in natural mica studies has been on investigation of the crystal structures of muscovites and lepidolites. Answers to many unsolved problems concerning the micas are considered to be directly related to polymorphic variations with changes in chemical composition. In the past quarter, work has progressed further along this line, but has been concerned to a larger extent with the determination of the structures of the chemically "aberrant" muscovite varieties, e.g., roscoelite, ocellacherite, mangan-muscovite, etc. In addition the structure of a newly acquired uniaxial muscovite has been determined, and a restudy of the rose muscovite problem completed.

Since the initiation of the mica study, many other methods and lines of investigation in addition to x-ray techniques have been followed simultaneously, but these have only been mentioned in previous quarterly reports. These fields include:

1. Optical studies
2. Chemical history of the micas
3. Compilation of complete synonymy of muscovite and lepidolite.
4. Extensive research on the paragenesis of the micas.

The complete results of these and other correlative areas of investigation will be incorporated in the final report.

Approximately 130 specimens of mica, most of which have been chemically analyzed, have been received from about 15 investigators. These include mainly muscovites, phlogopites, and lepidolites. To date little attention has been granted to the study of phlogopite, but during the coming quarter it is planned to investigate this species in detail. The extensive phlogopite collections now contain about 30 analyzed specimens.

UNIAXIAL MUSCOVITE

In Quarterly Report No. 1 (p. 4-5) reference was made to the muscovite described by Axelrod and Grimaldi (1949) whose $2V$ is small ($3-15^\circ$) and whose structure deviates from that of all other reported muscovites. This mica is said by Axelrod and Grimaldi (1949) to contain three layers in the unit cell and to have crystallized in the monoclinic space group, Cm. This mica was re-examined, and at that time there was noted a striking similarity between Weissenberg photographs obtained from this three-layered monoclinic muscovite and those obtained from three-layered rhombohedral micas described by Hendricks and Jefferson (1939). Axelrod and Grimaldi (1949), chose the monoclinic space group owing to a combination of factors: (1) A $2V$ of as much as 15° , which they chose not to ascribe to strain, and (2) diffuse scattering along certain reciprocal lattice lines. The restudy of this type of material substantiated the existence of a new muscovite type but left the space group assignment in doubt. The differences in diffuse scattering in the Weissenberg photographs obtained in the Michigan Mineralogical Laboratory are not considered to be of sufficient magnitude to warrant classing the structure as monoclinic. The $2V$ of as much as 15° , however, presented another problem, for it was also thought (as did Axelrod and Grimaldi) that such a large $2V$ could not be ascribed to strain. Thus, no evidence disagreeing with the conclusions of Axelrod and Grimaldi (1949) was obtained, although some of their arguments are not completely convincing.

In the meantime, numerous mica specimens analyzed by Professor J. Jakob, of the Technical University of Zürich, Switzerland, during the years 1925-1937 were made available for study. Included in this suite are many muscovites, one of which (No. 1041) is predominately uniaxial. This is Jakob's (1929) specimen 43.

Megascopically, specimen 1041 has the appearance of a normal, non-pegmatitic muscovite, very similar to specimens of the three-layered monoclinic muscovite supplied by Dr. Axelrod. It is silvery gray in color, and the flakes obtained are as large as 5 mm in diameter. Microscopic studies, however, reveal that a large majority of the flakes are uniaxial.

A few small areas in a few flakes show birefringence and have moderate $2V$ (approximately 40°). One such biaxial piece (1041b) that isolated proved to have the normal two-layered muscovite structure. Several uniaxial flakes have been studied by the Weissenberg method, and all possess the rhombohedral structure, similar to that reported by Hendricks and Jefferson (1939) for various other micas. Weissenberg photographs are identical with those obtained from the muscovite described by Axelrod and Grimaldi (1949), except that a few additional weak reflections were obtained along the $(hk\ell)$ $k=3n$ reciprocal lattice lines and there was no diffuse scattering. Weissenberg 0-, 1-, and 2-level photographs have been taken about all a- and b-axes of specimen 1041c ($2V = 0^\circ$). Corresponding photographs obtained from each of the axes showed absolutely no diffuse scattering, as was noted by Axelrod and Grimaldi (1949); all photographs had clear, sharp reflections. With $2V = 0^\circ$, and with identical photographs obtained from each of the a- and b-axes, a rhombohedral space group assignment becomes valid. The possible rhombohedral space groups would be either $C3_112$ or $C3_212$, the same as those found by Hendricks and Jefferson (1939) for lepidolite, phlogopite, and other micas. From these data it seems that the monoclinic description given to the Sultan Basin muscovite by Axelrod and Grimaldi (1949) is probably not advisable. There is no doubt that the rhombohedral description is accurate as a limiting case ($2V = 0^\circ$).

Strain or some other type of macrophysical disorder may be responsible for the $2V$ as large as 15° in the muscovite described by Axelrod and Grimaldi (1949).

Dimensions of the three-layered rhombohedral muscovite polymorph (specimen 1041) are approximately:

$$a_0 = 5.2 \text{ \AA}$$

$$b_0 = 9.0 \text{ \AA}$$

$$c_0 = 20.0 \text{ \AA}$$

$$\beta = 90^\circ$$

Information on the paragenesis of this specimen is scanty. Jakob (1929) merely mentioned that it occurs in a cleft vein in eclogite. It is interesting to note that both of these uniaxial muscovites apparently are hydrothermal low-temperature micas.

It cannot be denied that both the rhombohedral muscovite described in this report and the mica described by Axelrod and Grimaldi (1949) are true muscovites. The chemical analyses of these specimens are apparently trustworthy and normal in every respect. Henceforth, micas of this

character will be referred to as rhombohedral muscovite in keeping with an attempt to develop a classification based on both structure and chemistry. Other muscovite types on these bases are normal muscovite and "lithium muscovite", both of which have been discussed in detail in previous reports. There is some reason to believe that the rhombohedral symmetry of this type of muscovite may be explained on the basis of twinning. Work is progressing on this hypothesis.

ROSCOELITE

During the past quarter emphasis also has been placed on the study of the so-called muscovite varieties, particularly roscoelite, ocellacherite or barium muscovite, and mangan-muscovite. The purpose of this line of investigation was twofold: to determine the exact structural relationships of these varieties to true muscovite, and to study the limits of isomorphous substitution in the muscovite structure.

Roscoelite has been generally accepted as a vanadiferous muscovite in which vanadium has replaced much of the aluminum. This conclusion was reached by Clarke based on the chemical analyses of Hillebrand (Hillebrand, Turner, and Clarke, 1899). A detailed study of the optical properties of roscoelite was made by Wright (1914) in which the similarity to muscovite, such as the optic plane normal to (010), was noted. Atomic ratios calculated by Wells and Brannock (1946) from ten chemical analyses (nine from the literature) show that vanadium and aluminum combined (with the exception of the aluminum required to fill tetrahedral positions) have a total of two atoms with octahedral coordination in the mica sheet. This is the theoretical number for a heptaphyllite mica. The isomorphous replacement also seems probable on the basis of ionic radii: 0.57 for Al^{3+} and 0.65 for V^{3+} (Rankama and Sahama, 1950, p. 794).

Therefore, roscoelite was investigated by means of x-rays to determine the effect of substitution on the reflection intensities. In the University of Michigan collections only a few very fine-grained samples of roscoelite were available, so that the study was begun by means of the powder method. Powder photographs were taken of specimens 627, 724, and 987, all from California localities, all of which proved to have spacings identical with the one-layered monoclinic mica polymorph. Another type roscoelite from the Stockslager mine, was obtained from the U. S. National Museum and Harvard University. Powder photographs of this type material (Specimen 1091) also gave the one-layer polymorph pattern. On a crystal suitable for Weissenberg study from specimen 1091, a 0-level a-axis photograph indicated the presence of one-layered monoclinic structure with

the optic plane parallel to the side pinacoid. Samples of two recently analyzed roscoelites, Nos. 1014 from Paradox Valley, Colorado, described by Wells and Brannock (1946), and 1015 from the Placerville District, Colorado, described by Fischer, Hoff, and Rominger (1947) have been obtained from Dr. Axelrod of the U. S. Geological Survey. Powder photographs of these specimens likewise gave one-layer patterns. These results prove that roscoelite does not have a muscovite structure, despite the chemical similarity between it and normal muscovite. It seems most probable, therefore, that we are dealing here with an example of polymorphism related to the isomorphism, V-Al. The exact structural effect of as much as 20% V_2O_5 on the muscovite structure remains to be determined. However, the conclusion is that roscoelite is a true mica but not a vanadiferous muscovite.

OELLACHERITE

The term oellacherite was first used by Dana (1867) for the barium-containing mica from Pfitschthal, Tyrol, originally called margarite by Oellacher (1860). Chemical analyses of oellacherite are reported by Rammelsberg (1862, 1879) and Sandberger (1875). A specimen of oellacherite (No. 1092), originally from the Karabacek collection, from the type locality has been borrowed from Harvard University (Harvard No. 93774). The oellacherite is pseudomorphous after a curved, bladed mineral, probably kyanite. X-ray studies of the mica show that it has crystallized with the normal muscovite structure.

The only other similar mica reported is a barium muscovite, with 9.89% BaO, from Franklin, New Jersey, described by Bauer and Berman (1933). Specimens of this material have been obtained, Nos. 540 and 1095 from Harvard University and No. 1127 from the U. S. National Museum (No. 105848). All specimens are massive and pink in color, not unlike some so-called oncosines. Very little could be learned from an optical study of this material owing to its very fine-grained character. X-ray powder photographs were taken of the three specimens, and almost identical patterns were secured. Very small differences probably are due to impurities. The pattern is unique and is unlike that of any other mica polymorph. Therefore, the position of barium muscovite in the mica series cannot be determined at present. If it is a mica at all, it has crystallized in an unreported structure.

From the preceding discussion it can be seen that there is considerable confusion as to the authenticity and structure of barium-containing natural micas. Most of the limited number of analyses are old.

Further work on this interesting problem awaits the acquisition of additional cellacherite specimens.

MANGAN-MUSCOVITE

Eskola (1914), in describing the pegmatites of southwestern Finland, notes (p.37);

"A particular manganese-mineral, an intensely blue-coloured, fine-crystalline manganese-muscovite containing 2.30% MnO, was found at Mattkärr."

A specimen of this material, No. 1093, also has been borrowed from Harvard University (No. 87441) and studied. It, like the barium-muscovite from Franklin, New Jersey, is extremely fine-grained and little could be learned from optical studies. Powder x-ray analysis of this specimen also revealed a pattern totally different from any known mica polymorph, including the barium muscovites, and decidedly different from normal muscovite. This mineral should be studied further before it may definitely be called a mica. It is noteworthy that muscovites with more MnO have been reported, and that these have not been designated as mangan-muscovite.

ROSE MUSCOVITE

Owing to their striking color the rose muscovites have attracted the attention of mineralogists for well over a century. It was only natural that these micas were first considered to be lepidolites because of color similarity. In 1857, the true chemical nature of the rose muscovite was discovered by Mallet (1857). Controversy has existed since then on the cause of the color, the relationship of these micas to lepidolite, and their paragenetic significance. A restudy of this problem, assisted by five new chemical analyses financed by the Faculty Research Fund of the University of Michigan, has shed much light both on the crystal chemistry of the rose muscovites and their paragenesis.

Prior to 1854, rose mica was always considered lepidolite. At that time, however, Dana observed that the rose mica from Goshen, Massachusetts, apparently had a low Li_2O content and thus might not be a lepidolite. He notes (p. 227):

"In the United States, a rose mica at the albite vein in Chesterfield, Mass.; and at Goshen, Mass.; granular variety at Paris, Maine, with red tourmalines; also near Middletown, Conn. The Goshen mineral is of difficult fusibility and slight lithia reaction, and may not be of this species."

It remained for Mallet (1857), who determined that $\text{Li}_2\text{O} = 0.64\%$ and $\text{F} =$ approximately 1.89% in a specimen from Goshen, to establish the fact that the rose-colored mica was actually a muscovite. He states (p. 180):

"This small percentage of fluorine, and the very small amount of lithia found, both go to prove that the Goshen mica does not belong to the species Lepidolite, but rather to that of muscovite or common oblique mica."

Mallet (1857) noted that a yellow-green mica also was found in the same vein at Goshen, and concluded (p. 180):

"...there can I think be little doubt that both are essentially potash-micas of the species Muscovite, the rose-colored being probably tinged by oxyd of manganese."

The first complete chemical analysis of a rose muscovite (from Goshen) was made by Rammelsberg (1875) in which the low Li_2O and F contents reported by Mallet (1857) were confirmed. Rammelsberg determined $\text{Li}_2\text{O} + \text{Na}_2\text{O} = 0.30\%$ $\text{F} = 0.52\%$. Bowman (1902), in describing the minerals at Haddam Neck, Connecticut, presented a chemical analysis of a pink fibrous muscovite and noted (p.99):

"The MnO is sufficient to give the fused carbonates a pale bluish-green tinge, and is probably the cause of the pink colour of the mineral, but it does not appear to be present in weighable amount."

Schaller and Henderson (1926) made a detailed study of "purple" muscovite from the Harding and Pilar pegmatites in northern New Mexico and presented chemical analyses of the rose muscovite from these pegmatites. They conclude (p. 11):

"In the chemical composition there is very little variation in these micas from that of normal muscovite. Small quantities of iron and manganese are present as inherent parts of the mineral, probably replacing alumina, and their combined color effect produces the abnormal color and pleochroism of the mineral."

With regard to paragenesis Schaller and Henderson (1926) state (p. 9):

"Muscovite definitely replaces microcline and spodumene and probably also albite and quartz and together with these minerals is itself later probably replaced by lepidolite."

Hess (1925) disagrees with this interpretation and notes that rose muscovite at the Harding Mine replaces spodumene and (p. 294) "practically all other minerals."

Stevens and Schaller (1942) believe that rose muscovites form when muscovite is transformed into lepidolite. They state (p. 533):

"In the full development of the lithium stage, the mica formed is essentially lepidolite. No new muscovite is formed in this stage. The muscovite now present in specimens from the lithium phase is residual muscovite from the preceding sodium phase, as evidenced by the many examples of muscovite completely or partially changed to lepidolite, by the pink color of this muscovite with percentages of Li_2O intermediate between those of greenish muscovites of the albite phase and those of lepidolite, and by the border of lepidolite formed around the muscovite, which is often colored pink adjacent to the lepidolite border."

Objections to this idea may be summarized as follows:

1. There is very little paragenetic evidence that indicates that lepidolite replaces muscovite to a large extent.
2. Many overgrowths of lepidolite on muscovite have abrupt boundaries; the pink muscovite adjacent to the lepidolite border has not been confirmed by investigations in the Mineralogical Laboratory.
3. From the standpoint of crystal structure, under the normal conditions of replacement it is difficult to imagine the muscovite structure being transformed to one of the lepidolite polymorphs.
4. New data suggest that pink muscovite may be later than lepidolite and therefore does not occupy an intermediate paragenetic position between muscovite and lepidolite.
5. The lithium content of pink muscovite is not intermediate between that of greenish muscovite and lepidolite; in many instances it is less than that of normal muscovites.

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Approximately 50 specimens of rose muscovite from ten localities are in the University of Michigan collections. The colors vary from almost purple to a pale pink; it seems best to use the general term rose to designate the general tint. The colors vary, of course, considerably with thickness, and thin flakes appear colorless. The typical color of rose muscovite in hand specimens from several localities may be related to color plates in Ridgeway's (1912) Color Standards and Color Nomenclature as follows:

<u>Number</u>	<u>Locality</u>	<u>Plate number</u>	<u>Color</u>
554	Iveland, Norway	XXVI	Cameo Pink
555	Tördal, Norway	XXXVIII	Pale Rhodonite Pink
558	Apache, New Mexico	XII	Rose Pink
559	Newry, Maine	XXXVIII	Pale Persian Lilac
563	Harding, New Mexico	XII	Deep Rose Pink
572	Brown Derby, Colorado	XXVII	Pale Vinaceous
600	Bolton, Massachusetts	XXVI	Cameo Pink

In this study pink oncosines such as are described by Berggren (1940) and an apparently pale rose muscovite described by Jakob (1925) are not considered rose muscovite. A specimen of the analyzed material received from Jakob (his No. 9) would probably be described by most investigators as ruby muscovite. On the same grounds several other so-called rose muscovites will be eliminated from consideration in this report.

The structures of about 25 specimens of rose muscovite from the ten localities in our collections were determined by means of the Weissenberg x-ray method. All samples have a normal two-layered monoclinic muscovite structure with the same unit cell dimensions as described by Hendricks and Jefferson (1939). The distinguishing feature of the muscovite structure, i.e., the presence of $06l$ reflections with l odd, was observed on all photographs. Powder x-ray photographs were also taken of many of the samples before they were sent for chemical analysis. In no case were any extraneous lines observed; thus impurities were absent.

TABLE 1

Optical Constants of Rose Muscovite

Number	Locality	α	β	γ	2V	2E
554	Iveland, Norway	1.554	1.586	1.590	45°	74-1/2°
555	Tordal, Norway	1.553	1.585	1.590	45°	74-1/2°
557	Setesdal, Norway	1.555	1.586	1.592	46-1/2°	77-1/2°
558	Apache, New Mexico	1.555	1.586	1.592	43-1/2°	72°
559	Newry, Maine	1.555	1.587	1.592	46-1/2°	77-1/2°
568	Harding, New Mexico	1.555	1.587	1.592	45°	75°
572	Brown Derby, Colorado (coarse-grained)	1.553	1.584	1.587	45°	74-1/2°
*584	Brown Derby, Colorado (fine-grained)	1.555	1.588	1.591	45°	75°
591	White Spar No. 1 Colorado	1.553	1.586	1.590	45 to 46-1/2°	74-1/2 to 77-1/2°
597	Bolton, Massachusetts	1.556	1.590	1.595	45°	75°

*(\pm 0.002)

The optical properties of the rose muscovites from nine localities are presented in Table 1. All three indices were measured on an Abbé refractometer, and β and γ were also determined by the immersion method. In all cases, except No. 584, the values obtained are within 0.001. In the case of No. 584 the maximum error is \pm 0.002. 2V was determined by the Mallard method and is probably accurate to within 2°. A study of the values in Table 1 shows that the optical constants of the rose muscovites are remarkably similar for deposits throughout the United States and Norway. Actually, there appears to be more variation in different types of rose muscovites, within a single pegmatite (compare 572 and 584) than in the world-wide occurrences. Averages of the values presented in Table 1 are:

$$\alpha = 1.555^-$$

$$\beta = 1.586^+$$

$$\gamma = 1.592$$

$$2V = 45^\circ$$

The indices of rose muscovites are slightly lower, approximately 0.01 to 0.02, than the average obtained in this laboratory for about 250 other specimens of pegmatitic muscovite. Also, the values of $2V$ are larger than those obtained for the other types of pegmatitic muscovites. These results agree very favorably with the conclusions presented in Quarterly Report No. 1 (page 3) based on optical studies of muscovite. It was pointed out at that time that those muscovites with a relatively large $2E$, whether green or brown, generally have lower indices than muscovites with a relatively small or moderate size $2E$. Also it was noted that if both green and brown muscovites occur within the same pegmatite, the green variety will consistently have the larger $2E$ (and thus lower indices). Inasmuch as the rose muscovites have lower indices and higher $2E$ than any other type of pegmatitic muscovite, the previous conclusion is substantiated.

Some well-known field relations are of interest at this point. Many mica concentrations are confined to specific parts, zones, or other structural units of differentiated pegmatites. Brownish books generally are nearer the walls. Likewise, green books are rare in pegmatites with interior concentrations of brown mica. A pegmatite containing both brown and green muscovite has the brown farther from the core. Since green muscovites have larger $2E$ values than brown muscovites, $2E$ increases with decreasing relative age. Color and other optical variation are, in the main in the micas, a function of composition and probably iron in particular. From theoretical considerations, based on ionic radius comparisons and analogy to the crystal fractionation in the magnetic series, gabbro-granite, it can be demonstrated that early pegmatite minerals, including micas, contain more iron than later ones. Thus brown wall-zone mica should be relatively rich in Fe^{2+} , green core-margin mica should have an intermediate iron content, and rose muscovite, which occurs in replacement units that formed very late, should be lower in iron.

This hypothesis is substantiated by the results of five new wet-method quantitative analyses of rose muscovites. The analyses are in all major elements normal for muscovite. Of striking significance, however, are the total iron contents, which are decidedly lower than those of other pegmatitic muscovites. Most pegmatitic muscovites are estimated to contain about 3% of combined $FeO + Fe_2O_3$. The manganese content in the rose muscovites is not unlike that reported for most muscovites.

If the electrical properties of unstained, structurally perfect muscovite are a function of the $\text{FeO} + \text{Fe}_2\text{O}_3$ content (which remains to be decided from analyses and work now in progress), the above discussed variation leads to the interesting suggestion that on theoretical grounds and with other factors equal, green muscovite, against whose employment the mica industry has been decidedly prejudiced, may actually be more suitable than the "ruby" or brown types. In this connection it is well to note that synthetic ferro-ferri micas are semiconductors rather than insulators.

Final conclusions as to the interrelation of color and composition await the results of spectrochemical analyses.

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