ENGINEERING RESEARCH INSTITUTE
UNIVERSITY OF MICHIGAN
ANN ARBOR

STUDIES IN THE NATURAL HISTORY OF MICAS

SEPTEMBER 1951 - AUGUST 1953

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ABSTRACT

The micas, a group of sheet-structure alumino-silicates, are composed mainly of K,Mg,Fe^2,Fe^3,Mn and Li with OH and F but also contain varying small amounts of Na,Ba,Rb,V,Cr, and Ti as well as minute amounts of a large variety of trace elements. The main species are muscovite, lepidolite, phlogopite, biotite, zinnwaldite, paragonite, roscoelite, and taeniolite. Most micas are monoclinic; some are hexagonal; a few may be triclinic. In muscovite the 2-layer form is essentially invariant. Lepidolites crystallize in 1-layer, 3-layer and 6-layer forms which can be correlated with the Li content. Phlogopites, biotites, and zinnwaldites also show considerable polymorphic variation but correlations with composition or occurrence are not yet possible.

The micas of commercial significance are muscovite and phlogopite. Economically significant deposits of sheet-size, high-quality muscovite are confined to pegmatites, principally in wall zones and core-margin units. Muscovites from different zones in individual pegmatites are characterized by distinct differences in color, structural defects, and composition. Similar variations occur among the micas of different districts.

The pegmatitic micas show systematic differences in composition, mainly in Fe^3,Fe^2,Mg,Li, and F, which reflect the stage of differentiation of the pegmatite magma at the time each type crystallized.

PART I. GENERAL INFORMATION

I. INTRODUCTION

A. General Statement

The micas constitute a closely related group of minerals with both common and rare representatives. This report gives the results of research on this group of minerals undertaken mainly as Project M978, Natural Mica Studies, of the University of Michigan Engineering Research Institute, sponsored by the United States Army Signal Corps. The purpose of the project was "To conduct investigations on physical properties, crystal structure, and composition of natural micas as related to geological occurrences and genesis." Thus we have attempted to learn as much as possible concerning the natural history of micas.

A study of the mica group is of significance for two reasons: (1) Certain members of the group are widely distributed rock-forming minerals and therefore a study of these species leads not only to information regarding the conditions of their own formation, but also helps in establishing the environmental conditions under which the enclosing rocks were developed. (2) Several of the micas are of crucial and strategic significance, either as raw materials themselves or as a source for rare metals.

We have attempted to concentrate our work mainly on those micas that are of some economic significance; these include muscovite, phlogopite, and lepidolite. But because of the difficulty of defining the limits of the various species, and also because of the structural and chemical relationships between the economically significant micas and others of lesser or no commercial importance, some of our work has necessarily included micas of no direct economic value.

Members of the mica group are widespread in igneous rocks and mineral deposits of magmatic derivation; for example, muscovite in certain granites and particularly in granitic pegmatites; phlogopite in certain peridotites; biotite in gabbros, norites, diorites, and granites as well in many granitic pegmatites and other types of pegmatites; and lepidolite and

zinnwaldite, mainly in certain rare pegmatites and some high-temperature veins. Muscovite and its fine-grained variety, sericite, are widespread essential constituents and accessory minerals of hydrothermal veins and replacement deposits. Among the metamorphic rocks, biotite and/or muscovite are common and widely distributed in many types of slates, phyllites, schists, and gneisses, and phlogopite is a common constituent of some marbles. In sedimentary rocks, fine-grained detrital muscovite may be an important mineral in clastic rocks and authigenic muscovite may also become a significant constituent in some fine-grained clastics. We have not attempted to study the micaceous minerals that lie between the true micas and the clay-minerals group, namely, members of the illite group of minerals of which the most widespread representative is hydromuscovite or hydromica.

The micas are also of scientific significance in that they are the most outstanding representatives of a type of silicate structure, namely, the sheet structure or disilicate type, in which the silica tetrahedra all are placed in one plane with each tetrahedron being joined to others by the three oxygen atoms that lie within the common plane. A continuous extension of such a linkage provides a hexagonal network within the plane. Other mineral groups that are characterized by this basic sheet-type structure include the chlorites, brittle micas, and vermiculites. By studying representatives of the mica group it is possible to obtain more information on the fundamental properties of silicates showing this type of linkage.

The micas are of scientific significance also because of their highly diversified composition, being hydrous or fluosilicates of alkali metals, principally potassium, and of divalent as well as trivalent metals. Consequently, a great many isomorphous substitutions are possible within the group.

From the economic standpoint, muscovite and phlogopite represent substances that require an essentially unique type of mining and recovery, for they are desirable not because of their composition, but mainly because of their unusual combination of physical properties and because of the size of the crystals in which they occur. Consequently, the mining of muscovite and phlogopite demands that the crystals as they occur in the deposit be removed without damage or without diminution of grain size. In this respect the micas are similar to quartz crystals in the requirements of their mining. The unique combination of properties possessed by muscovite and phlogopite which makes them economically desirable is: (1) their perfect basal cleavage, (2) their flexibility and elasticity, (3) their extremely low electrical conductivity and high dielectric property, (4) their very low heat conductivity, and (5) their occurrence in large, relatively perfect, single crystals.

Lepidolite, the other mica of commercial value, is not mined because of its properties as a mica, but because it contains the element lithium which is extracted from it. Thus it is unimportant whether it occurs in large single crystals or relatively free of mineralogical impurities.

The importance of the mica studies is therefore two-fold; scientific, leading to information on mineral and rock genesis, silicate structures, and isomorphism; and economic, leading to data on occurrences, recovery, and effective utilization.

B. History and Personnel of Project

The investigations of Heinrich on micas began in 1941 at the suggestion of the late Professor Harry Berman of the Department of Mineralogy and Petrography of Harvard University. It was originally intended to attempt a study of the micas for a doctoral dissertation, but due to the death of Professor Berman and various other factors, this was abandoned. However, this preliminary work led directly to the compilation of a relatively complete collection of analyses of micas and a bibliography of articles relating to all aspects of mica research. From this work, one paper (Heinrich, 1946) resulted. During the period 1942 to 1947 Heinrich, as a member of the United States Geological Survey, was assigned during much of this time to the investigation and mapping of pegmatite mineral deposits, particularly mica pegmatites, and participated in such investigations in the southeastern mica districts of North Carolina, Georgia and Alabama as well as those of Idaho, Colorado and New Mexico. Subsequently, he has had opportunity to study mica deposits in New England, Montana and other western states, in Canada, and in various European countries, particularly those of Scandinavia and Germany. Most of the mica specimens which have been studied as part of this investigation have been personally collected by Heinrich and were carefully labeled and studied as to their detailed geological occurrence within their deposits. One group of Colorado specimens was collected by Levinson and the remaining studied specimens are analyzed micas which have been obtained from numerous investigators both within the United States and abroad. In all cases, however, we have attempted to confine our studies to specimens whose detailed paragenesis either was known to us from our own field work or whose geological occurrence was a matter of record.

General mica investigations were resumed in 1950 by Heinrich and Levinson in an attempt to learn more regarding the variation in optical and physical properties of muscovites within individual pegmatites and individual pegmatite districts. Attention was soon directed toward the widespread polymorphism existing in the mica group, and it became clear that in order to learn more regarding the genesis of pegmatitic muscovites, basic information on structural and compositional variations was required. In September, 1951, a large part of the investigation was transferred to Project M978 of the Engineering Research Institute of the University of Michigan sponsored and supported by the U. S. Army Signal Corps, without whose assistance the wide scope of our work would have been impossible.

At the Mineralogical Laboratory of the University of Michigan, many techniques were utilized in the quest for new data on natural micas. Foremost of these were X-ray techniques, by means of which the structures of over 1000 specimens of mica were identified. The refractive indices of about 500 specimens, particularly muscovites, were determined by means of the Abbé refractometer and immersion liquids, and 2V was measured by means of the Fuess axial-angle apparatus and the Mallard method. Extensive surveys of the literature were undertaken in connection with such problems as the study of overgrowths, zoning, paragenesis, and trace elements, in an attempt to correlate and interpret the older data on these subjects with information secured from our measurements and from the large number of new chemical analyses. A number of apparently significant Russian papers were translated in their entirety.

The project during the two years of its sponsorship by the U. S. Army Signal Corps has been under the administrative supervision of Dr. S. Benedict Levin, Deputy Chief, Chemical-Physics Branch, Squier Signal Laboratory. The project as a whole has been guided and supervised by Heinrich. Levinson has personally carried out nearly all of the X-ray work. The other two writers, Levandowski and Hewitt, have been employed as general assistants in varied aspects of the research work, but have also contributed specifically to the writing of certain sections of this report.

C. Acknowledgments

We are extremely grateful for the assistance rendered by many individuals and organizations.

Technical assistance and advice were given by Dr. S. B. Levin, Dr. H. Kedesdy (Squier Signal Laboratory); Mr. C. E. Harvey (Amer. Spectrographic Laboratories); Dr. R. A. Hatch, Dr. J. A. Kohn (U. S. Bur. of Mines); Prof. L. S. Ramsdell, Prof. R. M. Denning, Prof. C. B. Slawson, Prof. G. B. B. M. Sutherland (Univ. of Michigan); Dr. H. S. Yoder (Geophysical Lab.); Dr. W. T. Schaller, Dr. M. Fleischer (U. S. Geological Survey); and Prof. S. Goldich (Univ. of Minnesota).

Through the courtesy of the following investigators we have been fortunate in obtaining samples of about 200 different micas, of which approximately 150 have been analyzed and described in the literature:

(1)   Dr. J. M. Axelrod (U.S.G.S., Washington)
(2)   Dr. F. A. Bannister (Brit. Mus. Nat. History, London)
(3)   Prof. L. G. Berry (Queens Univ., Kingston, Ontario)
(4)   Mr. J. E. Bever (Univ. of Michigan)
(5)   Dr. Harald Björlykke (Norwegian Geol. Survey)
(6)   Dr. H. V. Ellsworth (Geol. Survey of Canada, Ottawa)
(7)   Mr. R. C. Erd (U.S.G.S., Washington)

(8) Prof. H. W. Fairbairn (Mass. Inst. Techn.)
(9) Prof. R. R. Franco (Univ. of Sao Paulo, Brazil)
(10) Prof. C. Frondel (Harvard Univ.)
(11) Prof. E. N. Goddard (Univ. of Michigan)
(12) Dr. C. H. Greene (Corning Glass Works, Corning, N.Y.)
(13) Mr. W. R. Griffitts (U.S.G.S., Washington)
(14) Dr. E. Grip (Boliden, Sweden)
(15) Dr. A. F. Hallimond (London, England)
(16) Dr. S. B. Hendricks (U. S. Dept. of Agriculture, Washington)
(17) Dr. P. Hidnert (Nat. Bur. of Standards, Washington)
(18) Prof. W. F. Hunt (Univ. of Michigan)
(19) Prof. C. O. Hutton (Stanford Univ., California)
(20) Mr. R. Jacobson (Geol. Surv. of Nigeria, Kaduna Junction)
(21) Prof. J. Jakob (Tech. Univ. of Zurich, Switzerland)
(22) Dr. Y. Kawano (Geol. Surv. of Japan)
(23) Dr. A. Knopf (Yale University, New Haven, Conn.)
(24) Prof. L. Lokka (Helsinki, Finland)
(25) Mr. A. M. Macgregor (Geol. Survey of Sourthern Rhodesia)
(26) Prof. C. Mauguin (Univ. of Paris, France)
(27) Dr. V. B. Meen (Royal Ontario Museum, Toronto)
(28) Dr. and Mrs. A. Miyashiro (Geol. Surv. of Japan, Tokyo)
(29) Prof. J. Murdoch (Univ. of California, Los Angeles)
(30) Prof. T. Noda (Nagoya Univ., Japan)
(31) Prof. E. W. Nuffield (Univ. of Toronto, Canada)
(32) Dr. O. H. Ödman (Royal Inst. of Technol., Stockholm)
(33) Prof. I. Oftedal (Univ. of Oslo, Norway)
(34) Prof. G. P. Pagliani (Univ. of Milan, Italy)
(35) Prof. R. L. Parker (Techn. Univ. of Zurich, Switzerland)
(36) Prof. G. Pehrman (Abo Academy, Abo, Finland)
(37) Prof. R. Pieruccini (Univ. of Florence, Italy)
(38) Dr. F. H. Pough (Amer. Mus. Nat. Hist., New York)
(39) Prof. R. T. Prider (Univ. of Western Australia, Perth)
(40) Prof. Percy Quensel (Univ. of Stockholm)
(41) Mr. A. H. Roberson (U. S. Bur. of Mines, Albany, Oregon)
(42) Dr. H. P. Rowledge (Govt. Chem. Laboratories, Perth, West. Australia)
(43) Dr. A. Schiener (Mus. Nat. Hist., Vienna, Austria).
(44) Dr. F. T. Seeyle (Dominion Laboratories, Wellington, New Zealand).
(45) Dr. K. Sugiura (Toyko Inst. of Technology, Japan).
(46) Dr. G. Switzer (Smithsonian Institution, Washington)
(47) Prof. S. Tyler (Univ. of Wisconsin, Madison, Wis.)
(48) Dr. S. van Biljon (Heidelberg, Transvaal, South Africa)
(49) Dr. F. Wickman (Royal Mus. of Stockholm, Sweden)
(50) Prof. H. Winchell (Yale Univ., New Haven, Conn.)
(51) Dr. H. Yamada (Tokyo Inst. of Tech., Japan)

II. CRYSTAL CHEMISTRY OF MICAS

Mauguin (1927, 1928A, 1928B) was the first to apply X-ray analysis to the study of micas. He determined their unit cells and symmetry and discussed the types of isomorphous replacement. Knowing the cell dimensions, the density and composition of micas, and the weight of each atom, he calculated the average number of atoms of each kind in the unit cell. One of the main points noted was that the total number of oxygen and fluorine atoms in the structural unit is always 12. It is upon this value that modern formulas of the micas are based.

Pauling (1930) advanced the general concept of the structure of mica and other layered silicates by proposing on the basis of his coordination theory, an outline of the general structural scheme for these minerals. Pauling's (1930) work gives, in particular, the sequence of atomic planes parallel to the cleavage plane. The first complete analysis of a mica structure on this basis was made independently by Jackson and West (1930, 1933). Their X-ray work on muscovite showed conclusively that the ideas proposed by Pauling (1930) were correct.

The basis of the mica structure, which has been described in detail by Bragg (1937), Hendricks and Jefferson (1939), and others, is the silica tetrahedron (SiO_4) arranged in sheets to form a hexagonal network (Fig. 1). Silicon has a coordination number of four. In the tetrahedron, aluminum may replace one-fourth of the silicon. Inasmuch as the silicon-oxygen sheet, by sharing oxygen atoms of adjacent tetrahedra, has the composition (Si_4O_{10}), the substitution referred to may result in the silicon-oxygen sheet having the formula ($AlSi_3O_{10}$). Two of these sheets are placed together with the vertices of the tetrahedra pointing toward each other. In muscovite these sheets are held together by aluminum atoms which have octahedral coordination with respect to the neighboring hydroxyl groups and oxygen atoms. The hydroxyl groups are situated at the centers of the plane hexagons formed by the oxygen atoms of the silica tetrahedra. The octrahedral groups may also be built around magnesium as in phlogopite, lithium as in lepidolite, and so forth. This constitutes one firmly bound double-sheet. The structure of the micas is completed by joining the above-mentioned double-sheets with potassium atoms which have twelve-fold coordination with respect to oxygen atoms. The perfect cleavage of the micas lies between these double-sheets.

With the possible exception of titanium, the position occupied by the many elements reported in all good analyses is no longer in doubt. Berman (1937) indicates the general formula for the mica group as:

—2K⁺
—6 O²
—3Si⁴⁺, 1Al³⁺

—4 O⁻, 2OH⁻

—4Al³⁺
—4 O²⁻, 2OH⁻

—3Si⁴⁺, 1Al³⁺
—6 O²⁻

—2K⁺

—6 O²⁻

—3Si⁴⁺, 1Al³⁺

—4 O²⁻, 2OH⁻

—4Al³⁺

—4 O²⁻, 2OH⁻

—3Si⁴⁺, 1Al³⁺
—6O²⁻

—2K⁺
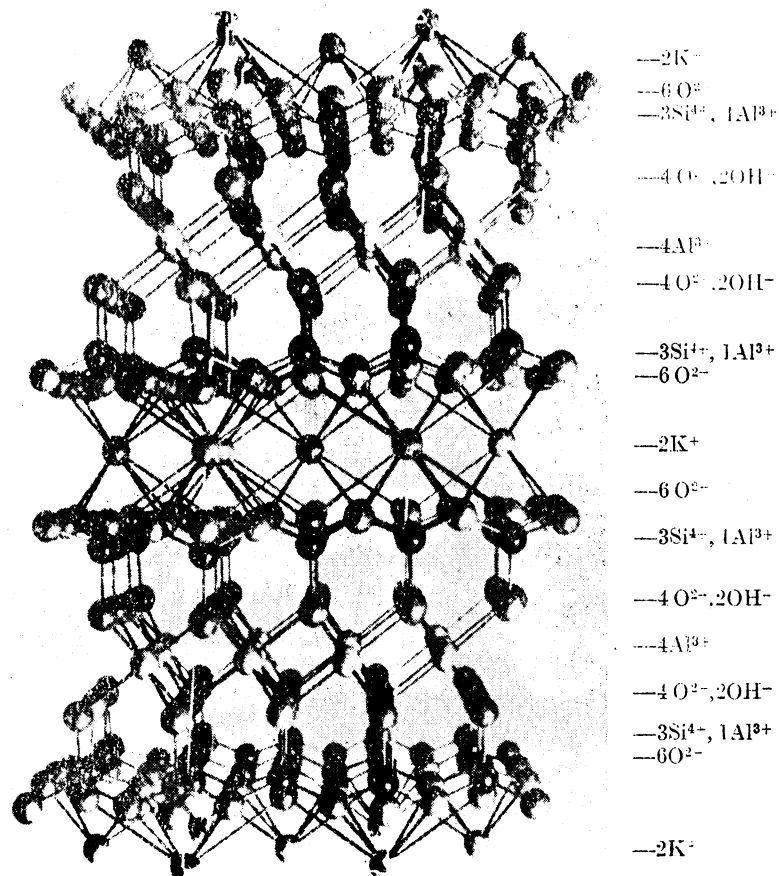
Fig. 1. Muscovite structure after Grim and Bradley (1951).

$$W(X,Y)_{2-3}Z_4O_{10}(O,OH,F)_2$$
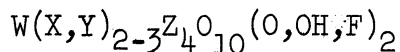
in which

W = K predominantly; Na, Ba, Ca, subordinate to rare;

X = Mg, Fe^2, Mn^2, Li;

Y = Al, Fe^3, Ti subordinate, Cr, Mn^3, V rarely;

Z = Si:Al 5:3 to 7:1

Atoms represented by W have twelve-fold coordination, those represented by X and Y have octahedral coordination, and those represented by Z have tetrahedral coordination.

Mauguin (1927, 1928A, 1928B) noted in his original X-ray studies that the c-axis of biotite was apparently only half as long as that of muscovite and he was thus the first to encounter polymorphism in the micas. It remained for Hendricks and Jefferson (1939) to give a full account of the scope and magnitude of polymorphism in this complex group. In all, among 100 different specimens, they discovered seven polymorphic variations which embrace three different crystal systems. They described the seven different structures (hexagonal, monoclinic, and triclinic) in detail and showed how they might be derived from the single-layer form by the application of various combinations of symmetry operations. Hendricks and Jefferson (1939) pointed out that muscovite crystallized in only the two-layer muscovite structure, but Axelrod and Grimaldi (1949) have reported a three-layer monoclinic muscovite (see page 47). Levinson (1953), as a result of work carried out on this project, has described a new variation of the muscovite structure for which the term "lithian muscovite" has been proposed. These are the only X-ray studies on micas since the classic contribution of Hendricks and Jefferson (1939).

III. X-RAY TECHNIQUES FOR IDENTIFICATION OF POLYMORPHS

It has been necessary to do a considerable amount of X-ray work, consisting mainly of identifying the structures of as many micas as possible. Whenever possible, Weissenberg photographs were taken about the 0-level a-axis, as this type of photograph is unique for the different polymorphs. In many cases, however, upper level photographs were taken as further verification of the structural interpretation.

In determining the orientation of the flakes used in the single crystal work, an optical procedure was used whenever practicable. Lines corresponding to the desired crystallographic axes were scratched upon the cleavage flakes with a razor. The exact positions of these axes were determined from interference figures and from extinction positions in those micas which have observable birefringence on cleavage planes. For all practical purposes, the crystallographic axes may be assumed to correspond with the extinction postion. However, many micas, particularly biotites and phlogopites, are uniaxial or have small 2V. In this case the orientation had to be determined from Laue photographs or by means of percussion figures.

In several instances crystal development was poor, crystals were minute, or extinctions were poor. This made single-crystal methods difficult if not impossible, and left powder X-ray methods as the only study technique. Unfiltered copper radiation was used in most single-crystal work and a nickel filter (with copper radiation) was used in powder studies. Iron radiation was used whenever powder photographs of biotites were taken. Only a large diameter (114.6 mm) powder camera is suitable for the mica studies. Polymorphs of muscovite and lepidolite can be distinguished by means of X-ray powder photographs but those of biotite and phlogopite cannot be differentiated by this method.

IV. NOMENCLATURE

In every complex mineral group there exists a large number of var-
ietal names based on major or minor chemical variations, optical distinctions,
grain size, occurrences, localities, and misidentification. The mica group
is no exception for it is burdened with a surplus of trivial names. The list
that follows defines the varietal names that have been employed in the series.
Most of these are listed in Hey (1950), Chemical Index of Minerals, but the
following references also were consulted in constructing the complete synonymy.

1. Dana (1892), A System of Mineralogy.

2. Chester (1896), A Dictionary of the Names of Minerals.

3. Winchell and Winchell (1951), Elements of Optical Mineralogy,
   Part II, Description of Minerals.

The following scheme is employed to differentiate the various cat-
egories:

Synonyms and varieties: lower case (e.g., adamsite)
End-members: first letter capitalized (e.g., Ferri-
        muscovite)
Pseudomorphs: capital delta preceding name (e.g.,
        Δachlusite
Doubtful species: underlined (e.g., anhydromuscovite)
Valid species, chem. varieties: capitals (e.g., MUSCOVITE)
Terms which are useful or sufficiently well defined, in the
opinion of the writers, and should therefore be retained:
asterisk preceding name (e.g., *MUSCOVITE).

Δachlusite: a green alteration product of topaz, resembling steatite, but
        near soda mica in composition; pseudomorphous after topaz.

adamsite: syn. of muscovite.

agalmatolite: syn. of pinite, pyrophyllite or talc.

alurgite: var. of muscovite near $K_2(Mg,Al)_{4-5}(Al,Si)_8O_{20}(OH)_4$, also contains
        about 1% Mn oxides.

ammochrysos: syn. of muscovite.

amphilogite: syn. of didymite.

anhydrobiotite: artificially dehydrated biotite, nature unknown.

anhydromuscovite: artificially dehydrated muscovite, nature uncertain.

annite: var. of lepidomelane; syn. of hydroxyl-annite.

anomite: var. of biotite with optic plane normal to b-axis (see meroxene).

anthrophyllite: "A mica (?)" Hey (1950).

Δaspasiolite: aluminosilicate of Mg, an altered cordierite.

aspidolite: near $(Na,K)_2(Mg,Fe^{+2})_6Al_2Si_6O_{20}(OH)_4$
            with $Na:K \approx 3$ and $Mg:Fe2 \approx 5$; near phlogopite.

avalite: near $K(Al,Cr)_4(Al,Si)_8O_{20}(OH)_4$; probably a mixture (Chester, 1896).

baddeckite: a mixture of hematite and clay, originally considered to be iron
            mica related to muscovite (Schaller and Henderson, 1926).

barium muscovite: syn. of oellacherite.

barium-phlogopite: var. of phlogopite with 1% BaO.

barytbiotite: var. of phlogopite; $2\left[(K,Ba)_2(Mg,Al)_{4-6}(Al,Si)_8O_{20}(OH)_4\right]$
            containing 6.84% BaO and also some Fe. Analysis in Dana (1892)
            shows it to be a phlogopite rather than biotite (Hey, 1950, p. 160).

basonite: near a hydrobiotite or a vermiculite; near $K_{1/2}(Mg,Fe^3,Fe^2Al)_{5-1/2}$
            $(SiAl)_8O_{20}(OH)_4 \cdot 1\text{-}1/2H_2O$ with Mg and Fe ≈ 2 and Si $\approx 5\text{-}1/2$.

bauerite: SiO_2 pseudomorphs after mica; the nature of the material is unknown.
            (Hey, 1950).

biaxial mica: syn. of muscovite.

bildstein: syn. of agalmatolite.

*BIOTITE: $K_2(Fe^2,Mg)_{6-4}(Fe^3,Al,Ti)_{0-2}(Si_{6-5}Al_{2-3})O_{20-22}(OH,F)_{4-2}$.

Δbonsdorffite: aluminosilicate of Fe^2 and Mg, an altered cordierite, pinite
            group.

brammallite: var. of illite, aluminosilicate of Na and K; syn. is sodium illite.

buldymite: aluminosilicate of Mg, Fe^2, Fe^3, and K; probably a hydrobiotite.

caesium-biotite: var. of biotite with as much as 3% Cs_2O and 1-1/2% Rb_2O (Hess and Fahay, 1932).

calciobiotite: var. of biotite containing as much as 14% CaO.

Δcataspilite: aluminosilicate of Mg,Ca,Na, and K, pseudomorphous after cordierite.

cat gold: syn. of muscovite; an early popular name for gold-colored mica.

catlinite: a mixture of sericite, pyrophyllite, etc.

cat silver: syn. of muscovite.

chrome mica: (1) syn. of fuchsite (Dana, 1892, p. 614); (2) syn. of fuchsite and mariposite (Whitmore et. al., 1946).

chromglimmer: syn. of chrome-mica and fuchsite.

chromglimmer (of Schafhäutl): in part var. of biotite, near $K_2(Mg,,Fe,Cr,Al)_4(AlSi)_8O_{20}(OH)_4$; the rest fuchsite (Hey 1950).

*CHROMIAN MUSCOVITE: name proposed for fuchsite since fuchsite has the typical muscovite structure (Whitmore, et. al., 1946).

*CHROMIAN PHENGITE: name proposed for mariposite as it has a high silica content and a small (less than 1%) Cr_2O_3 content. (Whitmore, et al., 1946).

chromochre: appears to be similar to muscovite but has more Cr_2O_3 and has a very small optic angle; it may be a chromian phengite (Winchell and Winchell, 1951).

colomite: syn. of roscoelite.

common mica: syn. of muscovite.

Δcordierite-pinite: pinitic pseudomorph after cordierite.

cryophyllite: var. of zinnwaldite often with some deficiency in (Li,Al,Fe) group; typically $Li \approx 2$-$1/2$, $(Fe^2, Fe^3) \approx 1$, and $Si \approx 7$. Also the name has been used for 2 hypothetical end-members; $K_2Li_2FeAl_2Si_7O_{18}(OH)_4$ and $K_2Li_2FeAl_2Si_6O_{16}(OH)_4$ (Hey, 1950).

cymatolite: muscovite and albite mixture, perhaps pseudomorphous after spodumene.

damourite: var. of muscovite; an altered muscovite, usually contains more water or more easily removed water (Winchell and Winchell, 1951). Folia less elastic; includes most hydro-mica in general; does not necessarily contain more water; they may, however, give it off more easily (Dana, 1922).

didrimite: syn. of didymite.

didymite: syn. of muscovite.

Δdysyntribite: syn. are dysintribite, dyssintribite, dyssyntribit; aluminosilicate of K and Na; pseudomorphous substance of pinite group.

Eastonite (of Winchell): theoretical end member, $K_2Mg_5Al_4Si_5O_{20}(OH)_4$; some natural micas apparantly approach this composition closely.

epileucite: pseudomorphs of orthoclase and muscovite after leucite.

epi-sericite: syn. of sericite; sericite formed in epi (upper) zone of metamorphism.

euchlorite (of Shephard): var. of biotite.

eukamptite: aluminosilicate of Mg and K; a mica or chlorite, not well defined.

euphyllite: near $(Na,K)Al_3Si_3O_{10}(OH)_2$; may be a mica intermediate between muscovite and paragonite, or a brittle mica, or perhaps a mixture.

Δfahlunite: aluminosilicate of Mg, Fe, and K near $(Mg,Fe^2)\ Al_2Si_3O_{10}\cdot 2H_2O$; pinite group.

ferribiotite: syn. of lepidomelane.

Ferri-muscovite: hypothetical end-member used by Winchell (1951) $(K_2Fe^3_2Al_2(OH)_4Si_6Al_2O_{20})$; also by Wahl (1925)$(K\ Fe^3_3Si_3O_{10}(OH)_2)$.

ferrititanbiotite: syn. of ferriwotanite.

ferriwotanite: var. of biotite high in Fe^3 and Ti (see wodanite).

ferro-ferri-muscovite: syn. of monrepite.

Ferrophengite: hypothetical end-member used by Winchell and Winchell (1951) $(K_2Fe^2Al_3(OH)_4Si_7AlO_{20})$.

Fluor-annite: var. of lepidomelane; the mica end-member $KFe^2_3AlSi_3O_{10}F_2$.

Fluor-biotite: var. of biotite; the mica end-member $K(Mg,Fe^2)AlSi_3O_{10}F_2$.

Fluor-lepidomelane: var. of lepidomelane; end-member $K(Mg,Fe^2)_3(Al,Fe^3)Si_3O_{10}F_2$.

Fluor-meroxene: var. of biotite; the end-member $KMg_3(Al,Fe^3)Si_3O_{10}F_2$.

Fluor-phlogopite: var. of phlogopite; the fluorine end-member $KMg_3AlSi_3O_{10}F_2$. Also the most abundant synthetic phlogopite.

Fluor-siderophyllite: var. of biotite; the end-member $KFe^2_3(AlFe^3)Si_3O_{10}F_2$.

frauenglas: syn. of muscovite.

fuchsite: var. of muscovite with as much as 4.81% Cr_2O_3. X-ray studies by Whitmore, Berry, and Hawley (1946) have shown that fuchsite has the muscovite structure. They propose that chromian muscovite be adopted in place of fuchsite which has no structural significance. This proposal is entirely justified. It follows, therefore, that if mariposite is also shown to have structures characteristic of phengite, the term chromian phengite or hexagonal chromian phengite would be appropriate (owing to a high silica content).

gaebhardite: syn. of fuchsite.

Δgieseckite: aluminosilicate of Mg and K; sometimes appreciable FeO; a pinite type pseudomorph.

gigantolite: a mixture of muscovite and biotite; pinite group.

gilbertite: var. of muscovite.

glimmer: syn. of mica.

goeschwitzite: syn. of illite.

Δgongylite: aluminosilicate of Mg,Fe^2, and K; a pinitic pseudomorph.

grundite: syn. of illite; trade name for illite clay.

gumbellite: var. of hydromuscovite (Hey, 1950); a hydrous silicate of aluminum, near pyrophyllite (Chester, 1896).

hallerite: a lithium bearing mica (0.5 to 1.5% Li), but it is not clear whether it should be classed as a variety of paragonite or of muscovite.

haughtonite: var. of biotite rich in Fe^2 and Fe^3.

helvetan: aluminosilicate of Fe^2,Mg,Ca and alkalis; probably an impure muscovite.

*heptaphyllite: group name for those micas which contain 7 other atoms to 12 (O,OH,F) as distinguished from octophyllite micas which contain 8 atoms to 12 (O,OH,F); originally applied to muscovite and lepidolite but now applies to muscovite only (and paragonite). The term heptaphyllite is structurally correct, and is thus considered valuable.

heterophyllite: var. of biotite; a mica between siderophyllite and annite; somewhat lower in Fe^2 than siderophyllite; the name seems of doubtful value.

hexagonal mica: syn. of biotite.

hydrobiotite: a hydrated biotite low in K, Mg, etc. and OH in place of the O. Also classed as a vermiculite.

hydromica: "Used as a term to designate a micaceous clay mineral of common occurrence resembling sericite but having weaker double refraction. Galpin (1912) indicates that kaolinite alters through metamorphism into hydromica, thence to muscovite. ... The group of hydrous micas, illite, hydromuscovite or glimmerton has been studied with particular care by Grim, Bray and Bradley (1937) who proposed the name illite. ... The material has less K_2O and more water than muscovite." (Kerr and Hamilton, 1949 p. 33)

*hydromuscovite: (1) var. of muscovite higher in OH, lower K, or K and Al than muscovite (Hey, 1950);
(2) general name for various hydromicas derived from muscovite (Chester, 1896).
(3) This mineral seems to lie between muscovite and illite both as regards potassium content and perfection of crystallinity (Brammall, Leach and Bannister, 1937).

hydrophlogopite: a var. of phlogopite high in H_2O.

hydrous mica: syn. of hydromica.

Hydroxyl-annite: var. of lepidomelane; the end-member $K_2Fe^2_6(Al_2Si_6)O_{20}(OH)_4$ with little Mg (Grigoriev, 1935). Also a syn. of annite. (Winchell and Winchell, 1951).

Hydroxyl-biotite: syn. of normal biotite so named to distinguish it from fluor-biotite.

Hydroxyl-lepidomelane: syn. of lepidomelane, in particular the F-free end-member.

Hydroxyl-meroxene: var. of biotite: the hydroxyl end-member to be distinguished from fluor-meroxene.

Hydroxyl-phlogopite: syn. of normal phlogopite so named to distinguish it from fluor-phlogopite.

Hydroxyl-siderophyllite: syn. of normal siderophyllite so named to distinguish between it and fluorsiderophyllite.

Δhygrophyllite: aluminosilicate of Fe^2,Mg,Ca and alkalis; derived in part, at least, from feldspars; pinite group.

iberite: (of Svanberg, 1844) syn. of gigantolite.

*illite: "It is not proposed as a specific mineral name, but as a general term for clay mineral constituents of argillaceous sediment belonging to the mica group." (Grim, Bray and Bradley, 1937, p. 816).

iron mica: syn. of lepidomelane and biotite.

irvingite: lepidolite near $(Na,K)_3(Li,Al)_5(Si,Al)_8(O,OH,F)_{24}$ with $Li \approx 2$-1/2 and $(OH,F) \approx 3$.

isinglass: syn. of mica, particularly muscovite and phlogopite.

ivigtite: aluminosilicate of Fe and Na(?); analysis imperfect (Hey, 1950); may be near gilbertite (Chester, 1896).

kaliglimmer: syn. of muscovite.

Δkillinite: aluminosilicate of K; pseudomorphous after spodumene; probably identical with muscovite; pinite group.

kryptotile: $Al_3(SiO_4)_3H_3$, extreme hydrogen end of muscovite series (Clarke, 1914).

lardite: syn. of steatite and of agalmatolite; pinite group.

*LEPIDOLITE: $K_2(Li,Al)_{5-6}(Si_{6-7},Al_{2-1})O_{20-21}(F,OH)_{3-4}$.

lepidomelane: a term commonly employed for iron-rich biotite. The term has been used for biotites rich in Fe^3, those rich in Fe^2, and those with relatively large amounts of both Fe^2 and Fe^3 (Heinrich, 1946).

lepidomorphite: (1) var. of phengite (Hey, 1950);
(2) a fine, scaly mica; the result of the alteration of oligoclase (Chester, 1896).

leucophyllite: a high silica var. of muscovite; similar to phengite; original analysis possibly on impure material.

leverrierite: may be the same as kryptotile but of different genesis (Clarke, 1914).

Λliebenerite: aluminosilicate of alkalis of Fe and Ca; a pinite group pseudomorph.

lilalite: syn. of lepidolite.

lilalith: syn. of lepidolite.

lithia mica: syn. of lepidolite.

lithioneisenglimmer: syn. of zinnwaldite.

lithionglimmer: syn. of lepidolite.

lithionit: syn. of lepidolite.

lithionite: syn. of lepidolite.

lithionitesilicat: syn. of lepidolite.

Lithium muscovite: a hypothetical mica end-member:
(1) Stevens (1938), $K_4Li_6Al_6Si_{12}O_{40}(OH,F)_8$;
(2) Berggren (1941), $K_4Li_6Al_{10}Si_{12}O_{41}F_5OH$.

lithium (Li)-phengite: contains 3.02% Li_2O, 47.44% SiO_2, 1.41% MgO and 1.87% total Fe. These values do not permit it to be classed as a phengite. Perhaps somewhere between muscovite and lepidolite; described by Shibata (1952A and 1952B).

lythrodes: an altered nepheline; pinite group.

macrolepidolite: var. of lepidolite; distinguished from microlepidolite by larger 2V.

magnesia mica: syn. of phlogopite.

mahadevite: a mica supposedly between muscovite and phlogopite in composition; near $(K,Na)_{0.97}(Al,Fe,Mg)_{2.66}(Si,Al)_4(O,OH)_{12}$; should be reanalyzed.

manandonite: a basic boro-silicate of lithium and aluminum closest to lepidolite in composition $Li_4Al_{14}B_4Si_6O_{29}(OH)_{24}$ (?); should be studied further; possibly not a mica.

manganese mica: syn. of manganophyllite.

manganese muscovite: syn. of mangan-muscovite.

mangan-muscovite: var. of muscovite with about 2% MnO.

manganophyll: syn. of manganophyllite.

manganophyllite (of Igelstrom, 1872): an Mn var. of phlogopite; $K_2(Mg_{5-4}Mn_{2-1}Fe^2_{0-0.5}Fe^3_{0-1}Mn^3_{0-1})(Si_6Al_2)O_{20}(OH)_4$; also considered var. of biotite (Hey, 1950).

Manganophyllite (of Yoshimura, 1939): hypothetical mica end-member $K_2Mn_5Al_4Si_5O_{20}(OH)_4$.

margarodit: syn. of margarodite.

margarodite: syn. of damourite.

marienglas: syn. of muscovite.

mariposite: var. of phengite (Whitmore, et al., 1946) with high silica and as much as 1% Cr_2O_3; see discussion of fuchsite.

meroxene: var. of biotite which has optic plane parallel with b-axis (perpendicular to 010) as contrasted with anomite. Most biotite was considered of this type (Dana, 1892).

metabiotite: syn. of bauerite.

metasericite: var. of muscovite; a hydrous mica classed under damourite.

micarel: mica pseudomorphous after scapolite; pinitic.

micarelle: syn. of micarel.

microlepidolite: var. of lepidolite distinguished from macrolepidolite by its small optic angle.

minguetite: near $(K,Na)_{0.8}(Fe^2,Mg,Fe^3,Al)_{5.2}(SiAl)_8O_{20}(OH)_4$ with (Fe,Mg)
≈ 3.2 and $Si \approx 6.8$; belongs either to chlorite or mica groups;
nature uncertain.

monrepite: $[KFe''_3Fe'''Si_3O_{10}(OH)_2]$; a ferro-ferrimuscovite; should be
studied further.

*MUSCOVITE: $[KAl_2(Si,Al)_4O_{10}(OH)_2]$.

nacrite (of Thomson, 1836): syn. of muscovite.

natronbiotite: var. of biotite containing appreciable Na.

natronphlogopite: var. of phlogopite containing appreciable Na.

oblique mica: syn. of muscovite.

*octophyllite: group name for those micas which contain 8 other atoms to 12
(O,OH,F) atoms. Biotite, phlogopite and lepidolite generally classed
as octophyllites (see heptaphyllite).

odenite: syn. of odinite, odite, and oderite. var. of biotite (Hey, 1950)
and var. of muscovite (Chester, 1896).

oellacherite: muscovite containing barium.

oncophyllite: muscovite pseudomorphous after feldspar.

oncosine: (1) aluminosilicate of Mg and K; a pinite group
pseudomorph; probably a mixture of muscovite, quartz, etc.
(Hey, 1950);
(2) a cryptocrystalline mica classed under damourite (Ford, 1932).

onkophyllit: syn. of oncophyllite.

onkosin: syn. of oncosine.

Δoosite: aluminosilicate of Fe^2 and alkalis; a pinitic pseudomorph.

pagodite: syn. of agalmatolite.

*PARAGONITE: $(Na,K)_2Al_4(Si_6Al_2)O_{20}(OH)_4$.

parophite: a pinitic rock.

Paucilithionite: a hypothetical mica end-member $(K_2Li_3Al_5Si_6O_{20}F_4)$.

*PHENGITE: var. of muscovite higher in silica than typical muscovite, and with Mg,Fe in place of some Al; $K_2(Mg,Fe,Al)_4(Si_7Al_1)O_{20}(OH)_4$.

*PHLOGOPITE: $K_2(MgFe^2)_6(Si_6Al_2)O_{20}(OH)_4$.

pholidolite: aluminosilicate of Mg, Fe^2 and K; perhaps a fluorine-free, Al-poor phlogopite; nature uncertain.

phyllite: (1) (of Thiébaut, 1925) syn. of pholidoide (group name for aluminous glauconites);
          (2) a general term for micas, chlorites, clays and vermiculites used by French writers;
          (3) (of Thomson, 1828) usually regarded as identical with ottrelite.

Picrophengite: a hypothetical mica end-member $K_2MgAl_3(Si_7Al)O_{20}(OH)_4$.

*pinite: a group of pseudomorphs, mostly of mica, after cordierite, nepheline, or scapolite; Heinrich (1950, p. 183), after a study of cordierite in pegmatite concludes, "Cordierite that crystallized in the magmatic pegmatite stage is not in equilibrium during the hydrothermal stage and is altered to pinite, which consists chiefly of muscovite, chlorite, and biotite in varying proportions. This change requires chiefly the introduction of potassium and hydroxyl by pegmatite solutions."

Δpinitoid: aluminosilicate of Fe^2 and alkalis; a pinitic pseudomorph.

Δpolyargite: aluminosilicate of Ca and K; a pinitic group pseudomorph.

poly-irvingite: a var. of irvingite high in Si.

Polylithionite: a hypothetical mica end-member.

polylithionite: a lithium bearing mica (lepdiolite) with high (as much as 9% Li_2O) lithium content.

potash mica: syn. of muscovite.

Protolithionite: a hypothetical mica end-member (Kunitz, 1924); $K_2Fe_3AlSi_3O_{10}(OH)_4$.

protolithionite (of Sandberger): var. of zinnwaldite.

pseudobiotite: an altered biotite high in H_2O; probably a hydrobiotite.

pterolite: a mixture of aegirine and lepidomelane.

pycnophyllite: syn. of pyknophyllite.

pyknophyllite: syn. of sericite.

Δpyrargillite: aluminosilicate of Fe^2,Mg, and alkalis; pinite group.

Δpyrrholite: aluminosilicate of Ca and K; a pinite group pseudomorph after anorthite.

rabenglimmer: var. of zinnwaldite high in Fe^3.

rastolyte: a hydrated biotite similar to voigtite.

rhombenglimmer: syn. of biotite.

rhombic mica: syn. of phlogopite.

*ROSCOELITE: has been regarded as a vanadium muscovite with as much as approximately 20% = V_2O_5; $K_2(V,Al)_4(Si_6Al_2)O_{20}(OH)_4$; new data is presented in this report (p. 212).

rosellan: syn. of rosite.

rosellite: syn. of rosellan.

Δrosite: aluminosilicate of Ca,Mg, and K; a pinitic pseudomorph.

rubellan: aluminosilicate of Mg,Fe, and K; probably an altered biotite.

sandbergerite:syn. of oellacherite.

sarospatakite: syn. of illite.

scale stone: syn. of lepidolite.

schernikite: pink, fibrous var. of muscovite occurring intergrown with lepidolite.

schuppenstein: syn. of lepidolite.

*sericite: (1) var. of muscovite; also includes much pinite (Hey, 1950);
           (2) fine scaly muscovite united in fibrous aggregates and char-
               acterized by its silky luster (Ford, 1932);
           (3) the name usually confined to white mica which is secondary,
               often the result of alteration of feldspar (it has been
               suggested that it contains less potash and more water)
               (Winchell and Winchell, 1951). Sericite as used by Winchell

and Winchell (1951) is so well established in the field of petrography that any attempt to dislodge or replace it will be futile. Gruner (1948, p. 684) notes, "The exceedingly fine-grained micas, which go under such names as sericite, illite, and hydromuscovite, are in a class by themselves. They are defect structures <u>principally</u> with regard to missing alkali cations. What takes the place of these ions? Analyses show definitely that as K decreases H_2O increases."

shilkinite: described originally by Merkulova (1939) as a new mineral but shown by Tchirvinskii (1948) to be identical with sericite.

siderischer-fels-glimmer: syn. of lepidolite.

siderophyllite: var. of biotite high in Fe^2 with little Fe^3.

sodium-illite: syn. for brammallite.

sterlingite: var. of damourite.

taeniolite: syn. of tainiolite.

*TAINIOLITE: $K_2Mg_4Li_2Si_8O_{20}(F)_4$; the only member of the mica group without essential aluminum. Classed under phlogopite by Dana (1926) and under lepidolite by Winchell and Winchell (1951).

talcite: syn. of damourite.

Δterenite: aluminosilicate of Mg and K; an altered scapolite; a pinitic pseudomorph.

titanbiotite: syn. of wodanite.

titanglimmer: original form of titanmica.

titanmica: group name for titaniferous micas.

titanobiotite: syn. of wodanite.

triclasite: syn. of fahlunite (pinitic pseudomorph).

uniaxial mica: syn. of biotite.

vanadinglimmer: syn. of vanadium mica.

vanadium mica: syn. of roscoelite.

verdite: impure fuchsite used as an ornamental stone.

voigtite: near $K(Fe^2MgFe^3)_6(SiAl)_8O_{20}(OH)_4 \cdot 3\text{-}1/2\ H_2O$ with $Fe^2:Mg:Fe^3 \approx 3:2:1$ and $Si \approx 5\text{-}1/2$; probably a hydrobiotite.

waddoite: a specimen labelled waddoite in the University of Michigan collections appears as typical biotite. Hey (1950) notes that it is incompletely described.

Δwilsonite: aluminosilicate of Mg and K; an altered scapolite; a pinite.

wodanite: a var. of biotite containing appreciable quantities of TiO_2 (as much as 12.5%).

wotanite: syn. of wodanite.

*ZINNWALDITE: $K_2(Fe^2{}_{1\text{-}2}Li_{2\text{-}3}Al_2)(Si_{6\text{-}7}Al_{2\text{-}1})O_{20}(F_{3\text{-}2}OH_{1\text{-}2})$. There seems to be some confusion as to whether zinnwaldite is more closely related to biotite or lepidolite.

zweiaxiger glimmer: syn. of muscovite.

The preceding classification is in general agreement with Hey (1950); a few points of difference, however, should be discussed. Hey considers brammallite, for example, a valid species when actually it is no more than a sodium-bearing illite. His opinion on varietal names is expressed as follows: (p. XI):

> "In the matter of varietal names, the chemical prefixes of
> W. T. Schaller (for example, Plumbian aragonite for Tar-
> nowitzite) have not been adopted; certainly a great mul-
> tiplication of varietal names based on small differences
> in chemical composition is to be deprecated, but it seems
> quite unnecessary to discard such well-known names as
> Freibergite, Pisanite, or Ceylonite; moreover, it is not
> really possible to draw a definite line between species
> and varieties."

Many objections to Hey's position on this matter have been raised and are clearly stated by Heinrich (1951, p. 634):

> "It is unfortunate that the compiler has seen fit to reject
> the Schaller System of adjectal modifiers which is rapidly
> obtaining general adoption by mineralogists in all parts of
> the world as a satisfactory scheme for freeing mineralogical
> nomenclature of its present unwieldy and largely meaningless
> agglomeration of varietal names. It is only by applying the

system of adjectal modifiers that mineralogists can remain
consonant with a modern conception of minerals as consti-
tuting chemical series."

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V. CLASSIFICATION

A. History of Mica Studies

Hypotheses advanced prior to 1924 to explain the chemistry of the micas and the other silicates are interesting today only from a historical point of view but a brief résumé of them is useful in giving an idea of the development of some modern theories.

Silicates as a group had been recognized about 1780 by Bergmann. He included in this division minerals of the mica, amphibole, garnet, feldspar, and clay groups, as well as individual species. As the discovery of new silicate species continued, several theories were evolved to explain their complex chemistry. Dana's fifth edition of A System of Mineralogy (1868) presented one of the most complete and widely accepted presentations in English up to that time.

Dana classified all silicates into two large groups--the anhydrous silicates and the hydrous silicates, each of which had its own subdivisions. The subdivision of the anhydrous silicates was as follows:

1. Bisilicates: oxygen ratio for the bases and silica 1:2

2. Unisilicates: oxygen ratio for the bases and silica 1:1

3. Subsilicates: oxygen ratio for the bases in silica 1: less than 1; mostly 1:2/3; but also 1:1/2 and 1:3/4

The following was the general subdivision of the hydrous silicates.

1. The general section of hydrous silicates. Includes all hydrous silicates except the zeolites and margarophyllites.

    a. Bisilicates.
    b. Unisilicates.
    c. Subsilicates.

2. Zeolite section.

3. Margarophyllite section.

The bases, with reference to the oyxgen ratio (defined by Dana, 1875, p. xi as "the ratio between the number of atoms of oxygen in the different oxygen compounds present") for bases and silica, comprise such elements as potassium, sodium, lithium, magnesium, manganese, aluminum, etc. In relation to the element silicon, which was considered so strongly negative, the oxides of the above named elements were relatively basic. For all practical purposes, the bases were considered mutually replaceable, eight to ten often occurring in the same compound combined in either simple or intermediate ratios.

Most of the micas were assumed to be unisilicates of the anhydrous silicate group, for the tersilicate subdivision, in which Dana placed them in his fourth edition (1854), was proved to have no existence. A few rare varieties such as margarodite, pinite, paragonite, oellacherite and euphyllite were placed in the margarophyllite section along with talc, serpentine and other hydrous micaceous minerals.

Ratios other than for bases and silica formed the foundation for further subdivision within the anhydrous unisilicate group. Chief of these were the RO and R_2O_3 ratio, as follows:

1. Oxygen ratio for bases and silica, 1:1;
    Phlogopite - oxygen ratio for RO, R_2O_3, between 2:1
             and 5:3;
    Biotite - oxygen ratio for RO, R_2O_3, about 1:1;
    Lepidomelane - oxygen ratio for RO, R_2O_3, about 1:3;
    Annite - oxygen ratio for RO, R_2O_3, 1:2.

2. Oxygen ratio for bases and silica, 1:1-1/4 to 1:2;
    Muscovite - oxygen ratio for RO, R_2O_3, 1:6 to 1:12
             and for RO + R_2O_3, SiO_2, mostly 1:1-1/4;
    Lepidolite - oxygen ratio for RO + R_2O_3, SiO_2 mostly
             1:1-1/2;
    Cryophyllite - oxygen ratio for RO + R_2O_3, SiO_2 1:2.

It is interesting to note that several relatively recent investigators have used similar ratios as the basis for their discussion of mica chemistry.

A systematic attempt to explain the chemistry of the micas was made by Tschermak (1878). In brief, Tschermak regarded the micas as consisting of four fundamental molecules to which the following formulas were assigned:

1. $R'_6Al_6Si_6O_{24}$ R' = K,Na,Li or H

2. $Mg_{12}Si_6O_{24}$

3. $H_8Si_{10}O_{24}$

4. $F_{24}Si_{10}O_8$
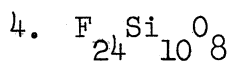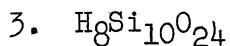
Aluminum may be replaced by ferric iron and chromium; magnesium is equivalent to ferrous iron and manganese.

The first of these formulas represents fairly well the composition of muscovite except for Tschermak's (1878) use of the double form of the molecule. Followers of Tschermak used the following formulas to represent the individual molecules:

1. $R'_3Al_3Si_3O_{12}$

2. $Mg_6Si_3O_{12}$

3. $H_4Si_5O_{12}$

4. $F_{12}Si_5O_4$
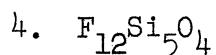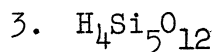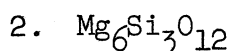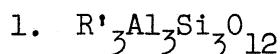
$H_2KAl_3Si_3O_{12}$ was given to represent the formula of common muscovite, which is similar to $KAl_2(Si,Al)_4O_{10}(OH)_2$, the one used today. Other ratios between K and H, e.g., $HK_2Al_3Si_3O_{12}$, were also hypothesized. The chief disadvantage in the former notation is that the true relationship between the various atoms, e.g., substitution of aluminum for silicon or indivuality of the hydroxyl radical, is not recognized.

The other Tschermak molecules represent hypothetical compounds. The second is a hypothetical polymer of chrysolite and the third is a hypothetical silicon hydroxide which may also take the form of the fourth, $F_{12}Si_5O_4$. The last two were not analogous to any known substance, and as pointed out by Clarke (1889, p. 384), were chemically improbable. Tschermak believed that the micas were isomorphous mixtures of these molecules.

According to Dana (1892, p. 612), Rammelsberg (1878, 1879)

"Regards the micas as containing the three silicates R_2SiO_3, R_4SiO_4, R_6SiO_5 in various molecular relation, e.g., Muscovite is $R_4SiO_4 + Al_4Si_3O_{12}$; the more acidic kinds are $R_{14}Si_4O_{15} = R_2SiO_3 + 3R_4SiO_4$, which is further written $mR_{14}Si_4O_{15} + nR_7Si_4O_{15} + pR_{14}Si_{12}O_{45}$, in which m:n:p = 5:1:5, 7:1:7, 9:1:9 in different cases. Similarly the other micas are resolved into the same three silicates, and the ratios in which they enter are calculated. That this method of calculation is applicable to any silicate, however complex, is obvious, but it is difficult to believe that the results reached really give the true constitution of the compounds."

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Tschermak's theory of the mica group remained generally in vogue and unchallenged until Clarke (1889) proposed an alternative. His hypothesis was considered by many to be superior. Dana (1892, p. 613) remarks, "The view of Clarke has the advantage that it assumes only one hypothetical molecule, which, moreover, is analogous to known compounds which play an important part in the Feldspar Group."

All orthosilicates (SiO_4 radical) were considered substitution derivatives of the normal salt $Al_4(SiO_4)_3$ in which the aluminum is replaced in varying amounts by univalent elements such as hydrogen, potassium, sodium, lithium, or univalent groups as MgF, AlO, and AlF_2. They are represented in the following formulas by R:

$$
Al
\begin{cases}
SiO_4 \equiv R_3 \\
-SiO_4 \equiv Al \\
SiO_4 \equiv Al
\end{cases}
\qquad 1. \quad R_3Al_3(SiO_4)_3
$$

$$
Al
\begin{cases}
SiO_4 \equiv R_3 \\
-SiO_4 \equiv R_3 \\
SiO_4 \equiv Al
\end{cases}
\qquad 2. \quad R_6Al_2(SiO_4)_3
$$

$$
Al
\begin{cases}
SiO_4 \equiv R_3 \\
-SiO_4 \equiv R_3 \\
SiO_4 \equiv R_3
\end{cases}
\qquad 3. \quad R_9Al(SiO_4)_3
$$
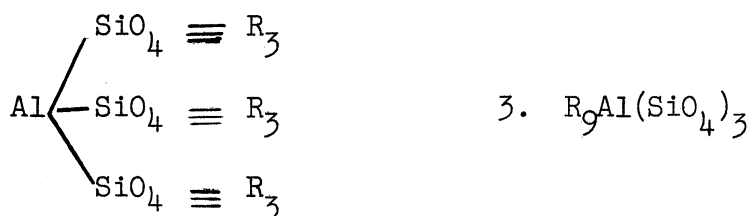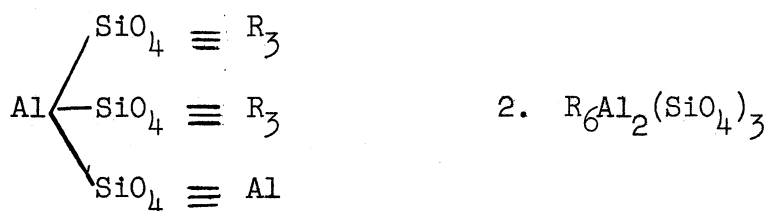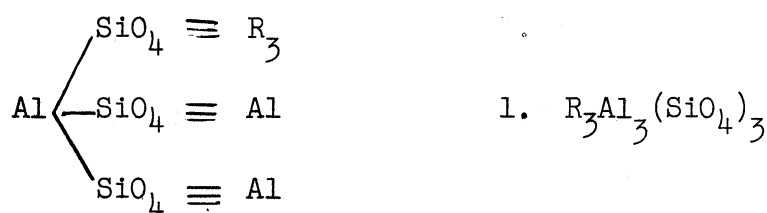
The second of these formulas is not essential as it can be regarded merely as a mixture of equal molecules of the first and third types. Derivatives of the first type include $R'R''$ $Al_3(SiO_4)_3$ or $R''_3Al_6(SiO_4)_6$. R'' represents magnesium, iron, manganese, etc. Likewise, derivatives of the second and third types may be obtained.

All the orthosilicates may be explained by means of the first and third derivatives. Those micas with fluorine are represented by the univalent groups MgF or AlF_2. Excess oxygen is represented by the univalent AlO group.

In the case of some muscovites, phlogopites, and many lepidolites, the oxygen-silicon ratio is low, which indicates a more acid condition. These micas are therefore considered to contain, in part, polysilicic acid, $H_4Si_3O_8$, which, like orthosilicic acid, H_4SiO_4, is tetrabasic. This line

of reasoning, which is similar to the one used at that time to explain the plagioclase series, necessitates isomorphous miscilibity between orthosilicates and polysilicates $(Si_3O_8$ radical).

Thus the general formulas for all micas may be obtained from the theoretical initial units $Al_4(SiO_4)_3$ and $Al_4(Si_3O_8)_3$. Examples of the initial points of the orthosilicates and corresponding polysilicates are given below:

1. $mR_3Al_3(SiO_4)_3$

   $nR_3Al_3(Si_3O_8)_3$

2. $mR_6Al_2(SiO_4)_3$

   $nR_6Al_2(Si_3O_8)_3$

3. $mR_9Al(SiO_4)_3$

   $nR_9Al(Si_3O_8)_3$

Clarke (1889) notes that if SiO_4 and Si_3O_8 were represented by X, then all micas fall within the limits $R_3Al_3X_3$ and R_9AlX_3.

Ordinary muscovite was represented by the first type, $R_3Al_3(SiO_4)_3$, in which $R_3 = H_2K$, giving $H_2KAl_3(SiO_4)_3$. Phengite, the high silica muscovite, was regarded as an isomorphous mixture of normal muscovite and the polysilicate $H_2KAl_3(Si_3O_8)_3$; other micas, such as lepidolite, are correspondingly much more complicated.

Kunitz (1924) made the first noteworthy modern attempt to correlate the chemical composition of the micas with physical and optical constants. In the course of his work he made 32 chemical analyses of various micas. Three main isomorphous groups of the micas were advocated by Kunitz:

1. Alumina micas, or the muscovite series, in which aluminum is replaceable by as much as 10% of Fe^3.

2. Magnesia-iron micas, or the biotite series, in which magnesium and ferrous iron are completely replaceable.

3. Lithia micas, or lithionites: Here Kunitz assumes a special trivalent complex group of elements consisting of lithium and silicon in variable proportions (Generally 2Li, Si), to which he assigns the symbol Le. This group is completely replaceable by ferrous iron. Lithia micas, therefore, may be written as $KH_2Al_2Le(SiO_4)_3$.

Kunitz (1924) points out that members of the lithionite series that are rich in lithia contain little alumina, and vice versa, and recognizes that lepidolite can be derived from muscovite by the substitution of Li_2O for Al_2O_3. He considers that there is a certain kind of isomorphous relationship between magnesium (and ferrous iron) and lithium. Partial isomorphous replacements also occur in the case of Si and Ti; Si and Al; Al, Cr, and Mn; K and Na; but not between K and H, or Al and Mg. His results show also that with increasing iron there is a corresponding, often linear, increase in the specific gravity and in the refractive indices. The refractive indices are to some extent modified by isomorphous replacement of fluorine and hydroxyl.

In 1925, Hallimond (1925, 1926, 1927) and Winchell (1925, 1927) presented almost simultaneously two new theories regarding the chemistry of the micas. In his discussion Hallimond (1925, 1926, 1927) emphasizes two main points, namely, that RO can replace R_2O_3, and that the invariant ratio of $K_2O:SiO_2$ is in the proportion of 1:6. On the basis of these assumptions, calculations are made and ideal formulas for various species and end-members are given. The micas are divided into two main sections; the acidic, which is further divided into the potash and lithia micas, and the basic, which comprises the biotites and phlogopites.

That RO can replace R_2O_3, Kunitz (1924 p. 389) denies on valency and theoretical grounds. Winchell (1925, 1927), however, believes that the $RO:R_2O_3$ ratio is variable and that K_2O is constant, but cannot agree with Hallimond's (1925, 1926) interpretation of either of these facts. Winchell (1925, 1927) observes that the $RO:R_2O_3$ ratio varies because Al_2O_3 replaces $MgSiO_3$ ($MgOSiO_2$) and vice versa. Winchell (1925, 1927) further regards potash as being constant and contends that not only does the $K_2O:SiO_2$ ratio vary, but that it varies in a regular manner from 1:6 in muscovite and phlogopite to 1:5 in protolithionite and siderophyllite and to 1:7 in phengite. One of the other points stressed by Winchell (1925, 1927) in his early studies of the micas is the belief that isomorphism depends on atomic volumes rather than on the valency of the elements which may proxy for one another (contrary to the opinion of Kunitz, 1924). In the micas, fluorine and hydroxyl may proxy for oxygen; potassium does not proxy for hydrogen, nor titanium for silicon. Winchell (1925, 1927) used many analyses to calculate end-members (many of which were revised in later works) of the biotite-phlogopite and muscovite-lepidolite series. In these contributions Winchell first used the terms heptaphyllite, (muscovite and lepidolite systems) and octophyllite (biotite system) for the two main groups of micas between which there is no isomorphism. This is the result of the observation that micas fall into the two distinct classes, the fundamental units of which contain seven and eight positive atoms respectively. These terms, somewhat modified, have now been generally accepted.

In a series of ten papers with the general title, "Contributions on the Chemical Constitution of Micas," Jakob (1925A,B; 1926; 1927; 1928; 1929A B,C,D; 1931; 1932) took an entirely different approach to the chemical and structural problems of the micas. The method is based on his theoretical and highly speculative theory of the constitution of the silicates (1920, 1921). A basic part of the theory is that certain atoms or groups of atoms are more closely united than others in a central atom, and may lie in an inner sphere of attraction while others are separately ionized and are possibly more remote. Jakob (1920, 1921) concludes that groups in the outer sphere have little or no influence on the capacity of two minerals to form mixed crystals. The constitution of the silicates he refers to certain prototypes $[SiO_4]R'_4$, $[SiO_5]R'_6$ and $[SiO_6]R'_8$ with or without added SiO_2 groups. It now appears that this theory has no basis either structurally or chemically.

With the application of X-ray diffraction technique to the study of natural micas between 1927 and 1932 (see section II of this part), new data was brought to light and with these new facts in mind Winchell (1932, 1935) revised some of his previous studies on the lepidolites and biotites. Lepidolites which had been classed as heptaphyllites are now, on the basis of Mauguin's (1927, 1928A, 1928B) X-ray results, regarded as octophyllite micas along with the biotites and phlogopites. New end-members in the biotite system were given as: annite $5(H_4K_2Fe_6Al_2Si_6O_{24})$, siderophyllite $6(H_4K_2Fe_5Al_4 Si_5O_{24})$, eastonite $6(H_4K_2Mg_5Al_4Si_5O_{24})$, and phlogopite $5(H_4K_2Mg_6Al_2Si_6O_{24})$. The end-members suggested by Winchell (1932, 1935) in these later investiations are in essential agreement with the conclusions of Berman (1937).

In his final paper on the micas, Winchell (1942) made his fourth attempt (others 1925, 1927, 1932) to correlate the chemical composition and optical properties of the lithia micas in which he took cognizance of the polymorphism established by Hendricks and Jefferson (1939). Newly calculated end-members include polylithionite $(K_2Li_4Al_2Si_8O_{20}F_4)$, protolithionite $(K_2LiFe_4Al_3Si_6O_{20}F_4)$, and paucilithionite $(K_2Li_3Al_5Si_6O_{20}F_4)$. Winchell (1942) has also noted that in most analyses of lepidolite the Li_2O content is deficient by almost one percent. This, he considers is due to the presence of some interlayered, but not isomorphously combined, muscovite.

The work of Stevens (1938) is the only other important contribution to the chemistry of the micas before it was realized that the micas crystallize in many polymorphic forms. This work on the lepidolites resulted in the interpretation of seventeen new analyses on the basis of isomorphous mixtures between four arbitrarily chosen end-members. They are polylithionite $(KLi_2AlSi_4O_{10}F_2)$, biotite $K(Mn,Fe,Mg)_3AlSi_3O_{10}(F,OH)_2$, lithium muscovite $(K_4Li_6Al_6Al_4Si_{12}O_{40}(F,OH)_8)$, and muscovite $(KAl_2AlSi_3O_{10}(OH,F)_2)$.

Volk (1939) studied the optical properties and chemistry of muscovite and suggested that the variations in composition and correlations with optical data may be interpreted with reference to the three end-members:

potassium muscovite, $H_4K_2Al_6Si_6O_{24}$; phengite $H_6K_2(Fe,Mg)_2Al_4Si_6O_{24}$; and ferric iron muscovite $H_4K_2Fe^3_2Al_4Si_6O_{24}$. The formula proposed for phengite is new and was used in order to get satisfactory correlation with the chemical analyses. The ratio of $R_2O:SiO_2$ remains almost constant in all three members.

The chemical constitution and classification of micas has also been discussed by Serdiuchenko (1948A). He proposed the idea that an almost continuous isomorphous series between muscovite and phlogopite is characterized by the replacement R_3^2--$R_2^3(Mg_3$--$Al_2)$ in the octahedral layers. The composition of biotite is considered intermediate between these two micas, i.e., isomorphism exists between heptaphyllites and octophyllites. This idea stems from the theory that in the biotite-phlogopite series, isomorphism takes place not only between magnesium and ferrous iron, or aluminum, ferric iron and chromium, but also between R_3^2 and R_2^3. This latter type "characterizes a limited isomorphism in the alternation ... $6Mg(OH)_2$ ... and ... $4Al(OH)_3$ ... layers in the lattices of the minerals." (In translation.)

The concept of alternating layers with octahedral coordination having different chemistry is similar, in a way, to that of Holzner (1936), who believed that biotite is intermediate in composition between phlogopite and muscovite. Furthermore, he suggested that the crystal structure of biotite is that of alternating layers of these two micas. Chemically he considered biotite as having the approximate composition of two phlogopites to one muscovite, which is $KMg_2Al_{0.667}(AlSi_3O_{10}OH)_2$, with the usual isomorphous replacements.

Serdiuchenko (1948A), moreover, suggests broad isomorphous replacements in the tetrahedral layers after noting that the Si:Al ratio fluctuates widely from 2.4:1.6 to 3.6:0.4. Silica rich micas such as phengite and alurgite are explained on the basis of the relatively large silicon content in these sheets. Selected biotites are cited as examples of micas intermediate in composition between biotite and muscovite. Serdiuchenko (1948A) also states (in translation),

> "In connection with the presence of polymorphic modification in the group of micas, the points of the heterostructural micas may be ranked on the line of our diagram."

This is interpreted as implying that the various polymorphic forms of biotite, for example, are direct consequences of their chemical position between muscovite and phlogopite.

From this discussion of the chemical history of the micas it is clear that the end-member concept is firmly engraved on the minds of many mineralogists. The requirements necessary for end-members are indicated by Winchell (1925, 1942) and Stevens (1938, 1946), and a method for calculating the ratios of end-members in the micas is well explained and illustrated by Stevens (1946). Heinrich (1946, p. 846), however, has pointed out that

"The theoretical 'end-molecules' used by Kunitz (1924) and
Winchell (1935) have little significance in the structure
of the biotite molecule, nor can variation in chemical
composition of the series be expressed adequately in terms
of them."

Rabbitt (1948, p. 284), in his work on the anthrophyllite series,
concurs and notes that the molecular end-member concept is founded on false
premises:

"No such end-members in the amphiboles are known in nature.
No such mixing on a molecular basis occurs. However convenient
it may be to plot the different properties in that way it must
be admitted that it engenders and perpetuates a false notion
of the variations in a mineral series."

B. Chemical-Structural Systematization of the Natural Micas

A workable systematization of the micas has long been one of the
aims of the project. A classification based on structural and chemical data,
along with an account of the very voluminous synonomy of the micas, has been
completed. In the following pages this classification is presented.

The numerous attempts at mica classification in the past have been
based almost entirely on their chemistry and fall generally into two cate-
gories: (1) those employing complex quasistructural formulas (e.g., Clarke,
1889; Jacob, 1925); and (2) those involving calculation of theoretical end-
member molecules (e.g., Tschermak, 1878; Kunitz, 1924; Stevens, 1946).
Berman (1937) approached the problem in more realistic fashion by correlating
mica formulas with unit cell contents and expressed the chemical variation
by means of atomic ratios of different elements occupying equivalent struc-
tural positions. However, the work of Hendricks and Jefferson (1939),
Levinson (1953), and others has demonstrated the complex polymorphism in the
mica group and the necessity for both chemical and X-ray studies of micas as
a basis for a sound classification.

Because of the complex chemistry of the group, the numerous tex-
tural and other minor varieties, and the repeated misapplication of names,
the nomenclature and synonomy of the micas have been ponderous and involved.
The purpose of this work is threefold: (1) to present a reasonable and usable
subdivision of the micas into naturally occurring species and major varieties
on the basis of both chemistry and structure; (2) to review the synonomy of
the group and thus assign minor varietal names or duplications to the species
or major variety to which they correspond; and (3) to list those micas that
are of indeterminate status owing to incomplete studies.

In the annotated classification that follows we have used for chemical varieties the prefixes of Schaller (1930). Wherever possible for each entry we list: (1) a simplified formula based on 24 negative ions in the unit cell; (2) the major isomorphous substitutions; (3) the structure; (4) notes; and (5) synonymy.

For the structural varieties, the method of polymorphic notation suggested by Ramsdell (1947) is adopted. The symbols and their meanings used in this work are as follows:

1M	1-layer monoclinic structure
2M muscovite	2-layer monoclinic muscovite structure
2M	2-layer monoclinic octophyllite mica structure
3M	3-layer monoclinic structure*
3H	3-layer hexagonal structure
6M	6-layer monoclinic structure
6T	6-layer triclinic structure
24T	24-layer triclinic structure
18T	18-layer triclinic structure (Amelinckx, 1952B)

The chemical-structural classification of the micas is as follows:

Species 1. <u>Muscovite</u>, $K_2Al_4(Si_6Al_2)O_{20}(OH)_4$. Minor Na,Rb,Cs,Ba, and Ca for K; minor Mg,Fe^2,Mn^2 and Li; Minor Fe^3,Ti, and Cr for Al; minor F for OH; maximum Li_2O = 3.30%, occupying vacant octahedral positions. Structure: 2M muscovite.

Synonyms: adamsite, ammochrysos, amphilogite, biaxial mica, cat gold, cat silver, common mica, damourite (in part), didrimite, didymite, ferro-ferri-muscovite, frauenglas, helvetan, heptaphyllite, isinglass, kaliglimmer, marienglass, monrepite, nacrite, oblique mica, potash mica, schernikite, zweiaxiger glimmer.

Hypothetical end-members: ferri-muscovite, kryptotile, leverrierite, lithium muscovite.

Varieties.   a. <u>Barian muscovite</u>. Ba with reported maximum of 5.91% BaO (Doelter, 1914) for K. Structure: probably 2M muscovite, material labeled oellacherite from Tyrol has the 2M muscovite structure.

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*There is reason for believing that the 3-layer monoclinic structure reported by Axelrod and Grimaldi (1949) is not truly monoclinic but just a distorted 3-layer hexagonal structure. This point is explained in further detail on page 47.

The barium-muscovite from Franklin Furnace, New Jersey, described by Bauer and Berman (1933), was examined by us by means of the X-ray powder method. The resultant pattern is not that of a muscovite, nor even that of a mica. The material is too fine-grained for detailed optical studies (See p. 68).

Synonyms: barium muscovite, oellacherite, sandbergerite.

b. <u>Manganian-muscovite</u>. Reported maximum MnO = 2.32% (Ellsworth, 1932); usually some Li_2O present. Structure: 2M muscovite.

The very fine-grained, deep purple mangan-muscovite of Eskola (1914) with 2.3% MnO has been checked by means of X-ray powder photographs, and the pattern does not correspond to any known mica structure. The photograph shows some quartz lines; see page 69.

Synonyms: mangan-muscovite, manganese muscovite.

c. <u>Ferrian muscovite</u>. Fe^{+3} with maximum of 5.70% Fe_2O_3 (Tschermak, 1878) reported, for Al. Structure: probably $2M^3$ muscovite, for the "alurgite" from Cajon Pass (Webb, 1939) with 5.32% Fe_2O_3 has been shown to have this structure.

d. <u>Ferroan muscovite</u>. Fe^{+2}, with maximum of 6.55% FeO (Wülfing, 1886) reported. Structure: probably 2M muscovite.

e. <u>Chromian muscovite</u>. Reported maximum of 4.81% Cr_2O_3 (Whitmore, <u>et al.</u>, 1946), for Al. Structure: 2M muscovite.

This follows the usage of Whitmore <u>et al</u>. (1946).

Synonyms: chromglimmer, chrome glimmer, fuchsite, gaebhardite, verdite.

f. <u>Lithian muscovite</u>. $K_2(Al,Li)$ ca. 5.0 (Si_{6-7},Al_{2-1}) $O_{20}(OH + F)_4$. Li_2O, at least 3.30% occupying vacant octahedral positions. Usually small amounts of F for OH. Structure: modified 2M muscovite (Levinson, 1953).

g. <u>Phengite</u>. $K_2(Mg,FeAl)_4(Si_7,Al_1)O_{20}(OH)_4$. High-silica muscovite with considerable MgO (7.96% Pagliani, 1937) and in some cases FeO; some Fe^3 for Al. Structure: 2M muscovite.

In addition to the 2M muscovite and 3H types of phengites, Dr. H. S. Yoder has informed us that some of the sericites from Amelia, Va. described by Glass (1935) have the 1M structure.

The nomenclature of the silica-rich end of the muscovite series as proposed by Schaller (1950) is regarded by us as unsatisfactory because: (1) there is some evidence that the original leucophyllite (Starkl, 1883) is a mixture, and in any event a new analysis and an X-ray study of the type material are needed to check its validity; and (2) the term, alurgite, which Schaller (1950) suggests as a substitute for leucophyllite in the event that the latter should prove untenable, also has been erroneously employed to indicate a normal, i.e., low-silica, ferrian muscovite (Webb, 1939; Ödman, 1950). X-ray studies, however, indicate that not all true alurgite is two-layer monoclinic in structure; some is three-layer hexagonal. Thus the term alurgite has at various times been used for:

(i) a red high-silica muscovite with minor Fe^3 and Mn (Penfield, 1893);

(ii) a red ferrian low-silica muscovite (Webb, 1939 and Ödman, 1950);

(iii) a manganian high-silica muscovite (Winchell and Winchell, 1951); and

(iv) a three-layer hexagonal polymorph of (i), discovered by Hendricks and Jefferson (1939) and verified by us on material from St. Marcel, Italy.

Because of this confusion, the use of the term alurgite for the high-silica end-member of the muscovite series is also undesirable. Less confusion accompanies the term phengite, which has been generally employed to mean high-silica muscovite.

Hypothetical end-members: ferrophengite, picrophengite.

h. Hexagonal phengite (3H phengite). Differs from 2M phengite (1g) in having the three-layer hexagonal structure (3H) and 2V = 0° - low. No well authenticated analysis of all-uniaxial material is available.

i. Chromian phengite. Cr, with maximum of 0.78% Cr_2O_3 (Whitmore, et al., 1946), for Al. Structure: 2M muscovite. This follows the usage of Whitmore et al. (1946).

Synonyms: chromochre, mariposite.

j. <u>Hexagonal chromian phengite</u>. (3H chromian phengite). Presumably chemically similar to 1i, but with the three-layer hexagonal structure and $2V = 0°$ - low. No analysis of all-uniaxial material available.

k. <u>Sericite</u>. Fine-grained muscovite. Structure: 2M muscovite.

The term-sericite has been used for:

(i) fine-grained muscovite, either primary or secondary;
(ii) fine-grained phengite; and
(iii) hydromuscovite.

Thus it cannot be defined exactly on a chemical basis. It remains, however, highly useful as a general, nonspecific term for fine-grained muscovite whose exact chemical nature is unknown.

Synonyms: (including pinitic pseudomorphs). achlusite, agalmatolite, aspasiolite, avalite, bildstein, bonsdorffite, catalinite, cataspilite, cordierite-pinite, cymatolite, damourite (in part), dysyntribite, epileucite, epi-sericite, fahlunite, giesickite, gigantolite, gilbertite, glimmer, gongylite, helvetan, hygrophyllite, iberite, ivgitite, killinite, lardite, lepidomorphite, liebenerite, lythrodes, margarodite, micarel, micarelle, oncophyllite, oncosine, onkophyllit, onkosin, oosite, pagodite, parophite, pinite, pinitoid, polyargite, pyknophyllite, pyrargillite, pyrrholite, rosellan, rosellite, rosite, shilkinite, sterlingite, talcite, terenite, triclasite, wilsonite.

<u>Species 2.</u> <u>Paragonite</u> $(Na,K)_2Al_4(Si_6,Al_2)O_{20}(OH)_4$. Structure: 2M; probably 2M muscovite. Schaller and Stevens (1941) have pointed out that the series muscovite-paragonite is not completely represented in nature. If intermediate types are discovered, it might be better to regard paragonite as sodian muscovite and reduce it to varietal status.

Synonyms: hallerite, natronglimmer, pregrattit, soda mica.

<u>Species 3.</u> <u>Roscoelite</u>. $K_2(V,Al)_4(Si_6Al_2)O_{20}(OH)_4$. Maximum $V_2O_5 = $ (ca.) 20%. Structure: 1M.

Although roscoelite has previously been regarded as a vanidiferous muscovite, it deserves full species rank because it is structurally distinct (see p. 212).

Synonyms: colomite, vanadinglimmer, vanadium muscovite.

Species 4. <u>Lepidolite</u>. $K_2(Li,Al)_{5-6}(Si_{6-7},Al_{2-1})O_{20-21}(F,OH)_{3-4}$. Rb and Cs replace K, in some types in considerable amounts $(Rb_2O = 3.2\%$, $Cs_2O = 1.90\%$; Lundblad, 1942). Small amounts of Mn,Mg,Fe^2,Fe^3 are normally present. The OH:F ratio varies considerably, and OH may become negligible.

Synonyms: irvingite, lilalith, lilalite, Li-phengite, lithia mica, lithionglimmer, lithionit, lithionite, lithionitesilicat, macrolepidolite, microlepidolite, poly-irvingite, scale stone, siderischer-fels-glimmer.

Hypothetical end-members: paucilithionite, polylithionite (in part), protolithionite.

Varieties. a. <u>Six-layer monoclinic lepidolite</u> (6M lepidolite). $Li_2O = $ (ca.) 4.0 - 5.1%.

b. <u>One-layer monoclinic lepidolite</u> (1M lepidolite). $Li_2O = $ 5.1 - 7.26%.

c. <u>Three-layer hexagonal lepidolite</u> (3H lepidolite). Composition approaches 4b. Due to twinning (?) (Levinson, 1953). 2V = 0° - small.

d. <u>Three-layer monoclinic</u> (3M lepidolite). $Li_2O = $ 4.1%. One example from Skuleboda, Sweden (see p. 147).

e. <u>Manganian-lepidolite</u>. Maximum reported MnO = 7.55% (Shibata, 1952B). Structure: probably variable, depending on Li content.

f. <u>Magnesian-lepidolite</u> (cited in Berman, 1937). We are unable to determine if any natural material of this composition has been discovered.

g. <u>Polylithionite</u> (in part). $K_2Li_4Al_2Si_8O_{20}(F,OH)_4$. A silicon- and lithium-rich, thus aluminum-poor, lepidolite. Structure: 1M.

Species 5. <u>Taeniolite</u>. $K_2Mg_4Li_2Si_8O_{20}F_4$; Structure: 1M.

Species 6. Zinnwaldite. $K_2(Fe^2_{1-2}Li_{2-3}Al_2)(Si_{6-7},Al_{2-1})O_{20}(F_{3-2},OH_{1-2})$.

The (Al,Fe,Li) group usually contains 6 atoms but may be considerably deficient. Zinnwaldites are chemically much more closely related to the biotites than to the lepidolites. In fact, there is no sharp natural compositional boundary between zinnwaldite and lithian biotite. If a demarcation is to made, we believe the line can best be drawn on the amount of Li and suggest that lithium-iron micas with Li > 1 atoms per unit cell formula be regarded as zinnwaldites, those with Li < 1 be placed with the lithian biotites.

Structurally, however, the problem is more complex. No zinnwaldites have been found with the 6M lepidolite structure so common in the lepidolites. On the other hand, the discovery of a zinnwaldite with a structure very similar to that of lithian muscovite (p. 201) does not permit one to overlook the possibility of at least some zinnwaldites resembling lepidolite structurally.

Synonyms: cryophyllite (in part), lithioneisenglimmer, poly-lithionite (in part), protolithionite, rabenglimmer.

Varieties.
a. One-layer monoclinic zinnwaldite (1M zinnwaldite).

b. Two-layer monoclinic zinnwaldite (2M zinnwaldite).

c. Three-layer hexagonal zinnwaldite (3H zinnwaldite).

d. Ferrian zinnwaldite maximum report Fe_2O_3 = 10.06% (Shibata, 1952B).

Species 7. Phlogopite. $K_2(MgFe^2)_6(Si_6Al_2)O_{20}(OH)_4$. Na can substitute for K up to nearly K:Na = 1:1 (Harada, 1936); minor Rb,Cs, Ba, and very minor Ca also may proxy for K. Fe^2 is almost always present, but Mg predominates greatly over Fe^2. Small amounts of Mn,Fe^3 and Ti may be present. The Si:tetrahedral Al ratio may be larger than 6:2.

There is no well-defined, natural, compositional boundary between ferroan phlogopite and magnesian biotite. Because Fe^2 is a strong chromophae, micas of this type even with only a small per cent of Fe are dark colored and are thus commonly classed as biotites. If a division is required, we suggest that where the ratio of Mg:Fe^2 > 4:2, the mineral should be classed as phlogopite.

Synonyms: aspidolite, barium-phlogopite, barytbiotite, hydro-phlogopite, magnesia mica, natronophlogopite, octophyllite, pholidolite, rhombic mica.

Hypothetical end-members: fluor-phlogopite, hydroxyl-phlogopite.

Varieties.   a. One-layer monoclinic phlogopite (1M phlogopite). The most common type.

b. Two-layer monoclinic phlogopite (2M phlogopite).

c. Three-layer hexagonal phlogopite (3H phlogopite).

d. Manganophyllite. $K_2(Mg_{5-4},Mn_{2-1},Fe^2_{0-0.5}Fe^3_{0-1}Mn^3_{0-1})(Si_6Al_2)O_{20}(OH)_4$. Structure: Generally 1M; some approach a three-layer hexagonal structure. 2M reported by Hendricks and Jefferson (1939).

Although some investigators (e.g., Hey, 1950) class manganophyllites as varieties of biotites, most manganophyllites have little or no Fe^2 and only small amounts of Fe^3. An exception is a Langban, Sweden, mica analyzed by Jakob (No. 8, p. 157, 1925A) which contains 16.94% Fe_2O_3. Apparently Mn is present commonly as Mn^3, rarely as Mn^2.

Synonyms: Manganese mica, manganophyll.

e. Titanian phlogopite. $K_2Mg_5Ti(Si_6Al_2)O_{20}(OH,F)_2$ (Prider, 1940) and Fe^2 and Fe^3 are minor; (OH,F) is deficient; 0.15 atoms of Ti are allotted to tetrahedral positions and 0.84 atoms to octahedral positions on basis of original analysis. Structure: 1M.

Species 8.   Biotite. $K_2(Fe^2,Mg)_{6-4}(Fe^3,Al,Ti)_{0-2}(Si_{6-5},Al_{2-3})O_{20-22}(OH,F)_{4-2}$. Some Na,Ca,Ba,Rb, and Cs for K; Mn for Fe^2, F for OH; Mg may be almost absent. Total F + OH may be very low (Walker and Parsons, 1926).

Synonyms: annite, anomite, caesium-biotite, chromglimmer (in part), euchlorite, eukamptite, ferromuscovite, haughtonite, heterophyllite, hexagonal mica, iron mica, lepidomelane, meroxene, natronbiotite, octophyllite, odenite, odinite, odite, oderite, pterolite, rhombenglimmer, rubellan, siderophyllite, titanglimmer, titanmica, uniaxial mica, waddoite.

Hypothetical end-members: cryophyllite (in part), eastonite, fluor-annite, fluor-biotite, fluor-lepidomelane, fluor-meroxene, fluor-siderophyllite, hydroxyl-annite, hydroxyl-biotite, hydroxyl-lepidomelane, hydroxyl-meroxene, hydroxyl-siderophyllite, manganophyllite (in part),

Varieties.

a. One-layer monoclinic biotite (1M biotite). The most common type.

b. Two-layer monoclinic biotite (2M biotite).

c. Three-layer hexagonal biotite (3H biotite).

d. Six-layer triclinic biotite (6T biotite).

Our studies of the supposed type 6T biotite described by Hendricks and Jefferson (1939) from Sterling, N. Y. (U.S.N.M.C3675) indicates the 1M structure. Dr. H. S. Yoder has studied other material from the same specimen and has found the 2M structure.

e. Twenty-four-layer triclinic biotite (24T biotite).

f. Eighteen-layer triclinic biotite (18T biotite). Amelinckx (1952B).

g. Calcian biotite. A biotite from Kaiserstuhl, Germany, has 14.33% CaO; Zambonini (1919). Structure unknown.

The validity of calcian biotite as a major variety is doubtful. The existence of Ca in the biotite structure has been challenged by Jacob (1929A). Several specimens labelled calciobiotite, from Italian localities, have been found to have the 1M structure.

h. Ferroan biotite. Mg is very minor or absent, Fe is present mainly as Fe^2.

Synonyms: siderophyllite, lepidomelane (in part).

i. Manganian-biotite. Mn as much as 1 atom per unit cell formula. Fe present as Fe^2 or Fe^3.

j. Ferrian biotite. $K_2(Fe^2,Mg)_{3-4}Fe^3_{2-3}(Si,Al,Ti)8O_{20-21}$
$(OH,F)_{4-3}$.

Synonyms: ferribiotite, lepidomelane (in part).

Lepidomelane is commonly employed for iron-rich biotite, but the term has been used to embrace biotites rich in Fe^3, those rich in Fe^2 and those with relatively large amounts of both Fe^2 and Fe^3 (Heinrich 1946),

k. <u>Lithian</u> <u>biotite</u>. $K_2(Fe^2,Mg)_5(Li,Al,Fe^3)_1(Si_7,Al)_8O_{20}$
$(OH,F)_4$.

l. <u>Titanian</u> <u>biotite</u>. $K_2(Fe_2,Mg_3)_5Ti(Si_6,Al_2)_8O_{22}(OH,F)_2$.

Synonyms: ferrititanbiotite, ferriwotanite, titanbiotite,
titanobiotite, wodanite, wotanite. Freudenberg (1919) reports
a biotite with 12.5% TiO_2.

Both titanian biotite and titanium pholgopites are rela-
tively poor in Fe^3 and also are very low in OH and F.

<u>Hydrous</u> <u>Micas</u>. Because most investigators class these minerals with
the clay group, their crystal chemistry is not considered here. However, it is
interesting to note that a new interpretation (Brown and Norrish, 1952) of
the chemistry of one of the species in this group, hydromuscovite, postulates
the replacement of K by oxonium (hydronium) ions H_3O^+. Species and varietal
names included in this group are: bastonite, brammallite, bravaisite, bul-
dymite, damourite (in part), goeschwitzite, grundite, gumbelite, hydrobiotite,
hydro-mica, illite, metasericite, Mg-illidromica, rastolyte, sarospatakite,
sarospatite, sericite (in part), sodium-illite, voigtite.

<u>Micas</u> <u>of</u> <u>Indeterminate</u> <u>Status</u>.

1. Euphyllite - near $(Na,K)Al_3Si_3O_{10}(OH)_2$; may be a mica
intermediate between muscovite and paragonite; or brittle mica or perhaps
a mixture.

2. Mahadevite - near $(K,Na)_{0.97}(Al,Fe,Mg)_{2.66}(Si,Al)_4(O,OH)_{12}$;
supposedly between muscovite and phlogopite in composition. A specimen of
mahadevite has been received from Dr. H. S. Yoder. X-ray studies of this
material indicate the 1M phlogopite structure.

3. Manandonite - a borosilicate of Li and Al closest to
lepidolite in composition; $Li_4Al_{14}B_4Si_6O_{29}(OH)_{24}(?)$; possibly not a mica.

4. Leucophyllite - a high silica phengite; perhaps a mixture.

5. Anthrophyllite - a "mica (?)" (Hey 1950, p. 283).

PART II. MUSCOVITE

I. MINERALOGY OF NORMAL MUSCOVITE

A. Chemistry

The ideal formula for muscovite is $K_2Al_4(Si_6Al_2)O_{20}(OH)_4$. However, all micas exhibit a large number of isomorphous replacements. The following isomorphous substitutions are of major proportions:

for K--Na,Rb,Cs,Ba, and (Ca)

for octahedral Al--Mg,Fe^2,Fe^3,Mn,Li,Cr,V and Ti

for OH--F

for (Si_6Al_2)--the proportion may approach (Si_7Al_1) as in phengite

For normal pegmatitic muscovite the percentages of isomorphously substituted elements may reach relatively large proportions. Many such micas with over 4 percent total iron, for example, have been reported or are now known on the basis on new spectrochemical data. It appears that rose muscovites, which are the last micas to crystallize in the hydrothermal sequence of complex pegmatites, approach most closely to the ideal muscovite composition (Heinrich and Levinson, 1953). They contain very little Fe^2,Fe^3,Mg, or Mn, etc.

Schaller (1950) has pointed out that the high-silica sericites (phengites) contain appreciable amounts of a divalent element, usually Mg. Pagliani (1937) reports a phengite with 7.96 percent MgO.

It has been reported by several authors (Gruner, 1948 and Sudo, 1949) that the fine-grained micaceous minerals which go under such names as illite, hydromica, sericite (in part) and hydromuscovite commonly show a deficiency of K and an excess of H_2O. Sudo (1949) showed that the (OH) content is roughly inversely proportional to the combined content of K,Na, and Ca. Gruner (1948) postulated that the hydronium ion (H_3O+) may occupy the vacant K portions, and Brown and Norrish (1952) supported this idea further by means of calculations. Several occurrences in which large books of normal pegmatitic muscovite are replaced by fine-grained sericite aggregates may be explained by a hydronium sericitic mica replacing a potassium muscovite (Liashchenko, 1940).

For this study 168 muscovites were analyzed spectrochemically. The results are shown in Table I.

TABLE I

NEW SPECTROCHEMICAL DATA ON 168 MUSCOVITES

	Present in % of samples	Range	Average
Fe_2O_3	100	0.63 - 5.1	2.904
MgO	100	0.008 - 1.9	0.931
MnO	100	0.009 - 0.83	0.106
TiO_2	99	0.003 - 0.52	0.128
CaO	100	0.0005 - 0.43	0.0069
SrO	21	0.001 - 0.005	0.0005
BaO	96	0.0002 - 1.1	0.114
Na_2O	100	0.33 - 1.7	1.02
Li_2O	26	0.01 - 1.6	0.089
Rb_2O	0	-	-
Cs_2O	0	-	-
Ga_2O_3	100	0.003 - 0.062	0.0219
Sc_2O_3	31	0.001 - 0.011	0.0011
Co_2O_3	37	0.0002 - 0.0008	0.0002
Cr_2O_3	32	0.0001 - 0.004	0.0001
SnO_2	76	0.0007 - 0.10	0.0067
V_2O_5	65	0.0001 - 0.036	0.0004
F	44	0.10 - 1.48	0.2361

B. Muscovite Structure

1. General and Polymorphism. Jackson and West (1930, 1933) made the first detailed study of the structure of mica (muscovite) and, in addition, confirmed in greater detail the structure of mica and other layered silicates proposed by Pauling (1930) on the basis of his coordination theory. The results obtained by Jackson and West (1930, 1933) on the unit cell dimensions of muscovite are as follows:

$$a_o = 5.18 \text{ Å},$$

$$b_o = 9.02 \text{ Å},$$

$$c_o = 20.04 \text{ Å}.$$

They selected 95°30' as the β angle in contrast with about 100° chosen by Mauguin (1927, 1928A, 1928B). The monoclinic space group determined by Mauguin as C2/c was verified.

The most detailed study of the mica group as a whole was conducted by Hendricks and Jefferson (1939) who found numerous polymorphs. In all, seven different polymorphic modifications embracing the hexagonal, monoclinic and triclinic systems were found among 100 investigated specimens of named micas. Each species, with the exception of muscovite, was found to occur in more than one polymorph. Biotite, for example, appears in five modifications, lepidolite in four, etc. Muscovite, however, Hendricks and Jefferson (1939) found only as the 2-layer monoclinic muscovite type (2M muscovite). They also found one "lepidolite" and one alurgite with this structure.

The distinguishing feature of the muscovite structure is its distortion from that of the ideal mica. This is revealed by the presence of the reflections, (06ℓ) with ℓ odd, which should normally be absent in the ideal arrangement on the basis of the structure-factor calculations of Jackson and West (1930, 1933); see Fig. 5. This distortion results from an incomplete filling of the octahedral positions and is considered by Hendricks and Jefferson (1939) to be the factor permitting only the 2-layered structure for muscovite to form.

Herein also lies the reason why Winchell's (1925, etc.) grouping of the micas into heptaphyllite and octophyllite divisions appears correct, for these terms simply mean that the unit cell contains seven and eight atoms respectively, for twelve O,OH, and F. Muscovite, $KAl_2(AlSi_3)O_{10}(OH,F)_2$, is the type heptaphyllite, and phlogopite, $KMg_3(AlSi_3)O_{10}(OH,F)_2$, is the type octophyllite. This relation may also be expressed by saying that the heptaphyllites have only 2/3 of the octahedral positions filled, whereas the octophyllites have all such positions filled. The presence of (06ℓ) reflections with ℓ odd must imply departure from the ideal muscovite structure given by Jackson and West (1930, 1933). Hendricks and Jefferson (1939) reported these reflections absent in the two-layered biotite-like micas (2M). It follows therefore that muscovite (two-layered) has a structure different from that of the two-layered biotites.

Diffuse scattering is reported in all the micas except muscovite. The scattering is observed in Weissenberg photographs along those reciprocal lattice lines in which h and k are constant, but in which k is not divisable by three ($h_a{}^kb1$, $kb{\neq}3n$). Laue photographs taken normal to the cleavage demonstrate scattering in the form of asterism or radial streaks. Mauguin (1927, 1928A,1928B) first noticed this effect and its significant absence in muscovite, and theorized that it was the result of some type of randomness in the positions of the heavy ions. Hendricks and Jefferson (1939) explained it in terms of constant h and k indices with an apparently continuous variation of the ℓ index.

This results from a variable periodicity in the stacking of mica layers in such a manner that those planes in which the k index is a multiple of three are undisturbed. They further state that if one half of a mica layer is translated by $nb/3$ with respect to the other half, it leaves the layer unchanged with respect to itself but results in a change of the successive layers. This is the factor which permits polymorphism in the micas (but in the muscovite structure distortion prohibits translation). Therefore, continuous scattering arises from destruction of the lattice periodicity perpendicular to the cleavage, resulting from a translation of some layers along the b-axis, parallel with the cleavage, by $nb/3$. Scattering has also been reported in other layered silicate minerals such as stipnomelane, vermiculite, cronstedite, and the chlorites. Essentially the same explanation is given for the phenomenon in these minerals.

In our work we have found at least one example of an apparently normal pegmatitic muscovite with small amounts of diffuse scattering (Fig. 2; compare with Fig. 5). The scattering, however, is observed only along the 02ℓ reciprocal lattice line (only 0-level a-axis photographs have been taken of most muscovites). Scattering of the type described in the preceding paragraphs would require scattering along both the 02ℓ and 04ℓ reciprocal lattice lines. The exact significance of this type of scattering has not yet been determined. Noteworthy perhaps is the fact the specimen (No. 6) from which the photographs of Fig. 2 were obtained has a SiO_2 content of 48 percent which is about 3 percent above normal. Phengites and other high-silica muscovites, as will be shown later in this report are known to crystallize as more than polymorph: the two-layer normal muscovite and the 3-layer hexagonal.
It is suggested that specimen No. 6 may be transitional between normal muscovite and the high-silica muscovites that crystallize in several polymorphs, and the diffuse scattering may be an embryonic attempt at a stacking arrangement other than the 2-layer type.

In a recent paper, Axelrod and Grimaldi (1949) have reported a new polymorph of muscovite that contains three layers in the monoclinic unit cell and has a very small, variable 2V of from 3° to 12°. A portion of this analyzed material has been obtained, and a re-study of it substantiates the existence of this new polymorph. However, there is a striking similarity between the "3-layered monoclinic muscovite" (space group C2 determined by Axelrod and Grimaldi, 1949) Weissenberg photographs and those of the 3-layered hexagonal mica polymorph (space group $C3_112$ or $C3_212$ determined by Hendricks and Jefferson 1939) recorded for biotite, alurgite, phlogopite, zinnwaldite and lepidolite. There is apparently room for a difference of opinion in the interpretation of the X-ray photographs of this muscovite. Axelrod and Grimaldi (1949) chose the former space group because of the presence of diffuse scattering in the (h_ak_a1) zone lines of one of the pseudo a- and pseudo b-axes, for they state (p. 569):
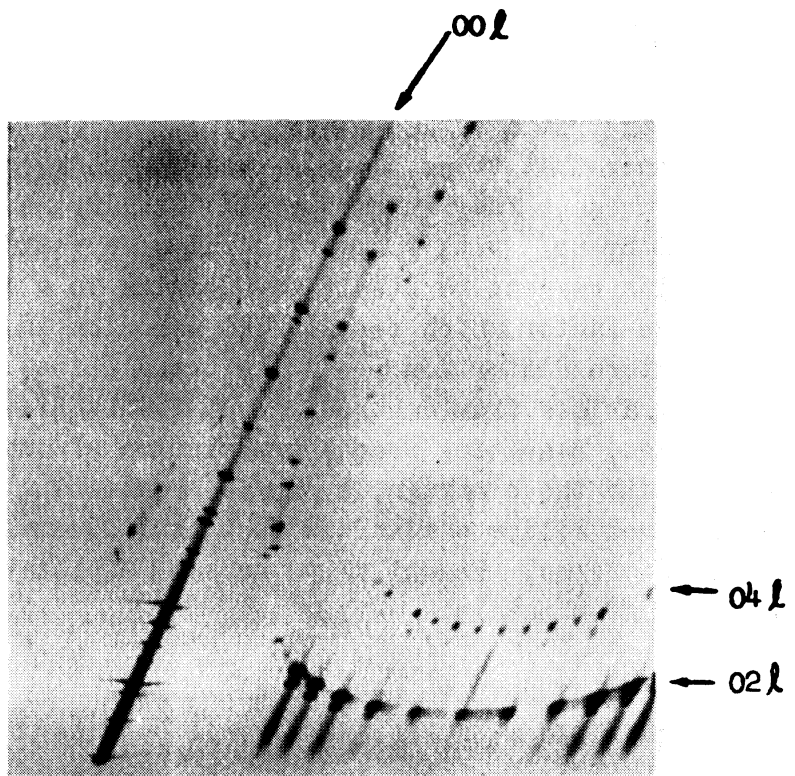
Fig. 2a. Muscovite showing some diffuse scattering along
02 ℓ reciprocal lattice line. 0-level a-axis. Compare
with Fig. 5.
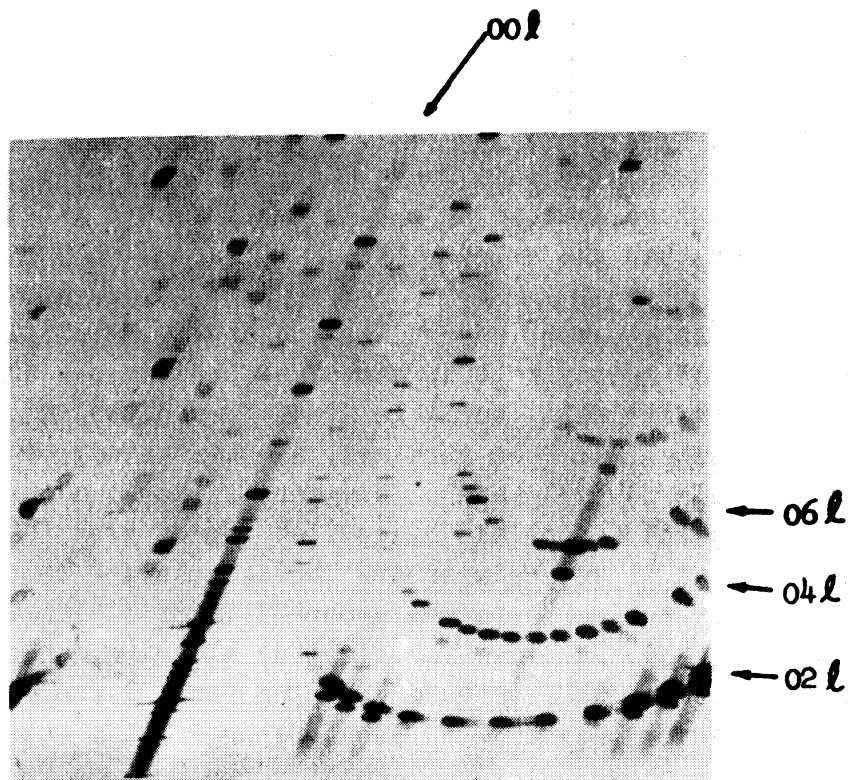
Fig. 2b. Muscovite showing very diffuse scattering along
02 ℓ reciprocal lattice line. 0-level a-axis. Another
photograph of same specimen as Fig. 2a.

"The X-ray symmetry, based on the symmetry of the
individual Weissenberg patterns, was C_{2h} or D_{3d}. If
the differences in diffuseness between Weissenberg
patterns are neglected, a threefold axis is demonstrated
and the Laue symmetry is D_{3d}. If, as we prefer, the optic
axial angle of 12° is not ascribed to strain and the
differences in diffuseness are not neglected, the Laue
symmetry must be taken as C_{2h} with the structure very
close to trigonal."

Many Weissenberg photographs of this polymorph have been taken and
studied by the writers in an attempt to assign definitely a space group to
this muscovite. No distinct differences in diffuse scattering along the recip-
rocal lattice lines described by Axelrod and Grimaldi (1949) were found. Phys-
ical distortion or corrugation of the crystals X-rayed may produce effects
that could outweigh the diffuse scattering inherent in the crystal structure.
Although Axelrod and Grimaldi (1949) consider strain unlikely, we are inclined
to favor this as the cause of the 2V of 12°.

However, it is our opinion that the significant fact concerning this
mica is that it contains 3 layers in the unit cell and not the fact that it is
monoclinic because of its 2V of 12°, or because of diffuse scattering. We are
convinced that if measurements are made with sufficient accuracy, many micas
will be shown to be triclinic by a few minutes or seconds. On the basis of
personal communication with several investigators we have every reason to be-
lieve that the β angle of the 3-layer lepidolite described by Hendricks and
Jefferson (1939) varies slightly from 90° and thus must be considered as having
a symmetry no higher than monoclinic. (We have been informed of β angles of
90°05' by Dr. J. Smith of the Geophysical Laboratory and 90°30' by Mr. S. A.
Forman as measured on photographs from Hendricks and Jefferson's original
material). Hendricks and Jefferson (1939) note that some of the micas they
describe as hexagonal show a small 2V and that all give some diffuse scatter-
ing. They state, however, (p. 762), "There is no doubt but that the hexagonal
description is accurate as a limiting case." We agree with this statement and
believe that the monoclinic nature of these 3-layer micas is probably due to
physical defects and is not inherent in the mica structure. It may be practi-
cal, in the future, if precision of measurement continues to increase, to
class and discuss micas merely as 1-layer, 2-layer, 3-layer, etc., without
regard to crystal system, inasmuch as all multi-layer forms might be derived
from the single-layer form by the application of various combinations of
symmetry operations.

Postel and Adelhelm (1944) have described a white muscovite of late
hydrothermal origin from the Wissahickon complex in which 2V varies from 22°
to 50°. The explanation advanced is that random shift in the structural planes
of mica may have some bearing on the low and variable 2V. In their paper, they

include a Laue photograph showing asterism which, if not due to some physical distortion, may indicate the existence of another muscovite whose structure is aberrant.

A study of the crystal structure in muscovite, as well as in many other micas, was one of the principal objects of this study. The Weissenberg method was used primarily, and usually one photograph about the a-axis was sufficient to identify the polymorph. The structures of about 150 muscovites, chiefly of pegmatitic origin, have been identified and include "normal" muscovites, rose muscovites, sericites and several uncommon varieties such as pinite, margarodite, adamsite and alurgite. Almost all have the 2-layer monoclinic muscovite structure described by Jackson and West (1930, 1933) and Hendricks and Jefferson (1939), except:

1. The 3-layer polymorph of Axelrod and Grimaldi (1949)

2. Some alurgites and other high-silica muscovites and phengites

3. Lithian muscovite

2. _Morphology_. After X-ray studies revealed the prevalence of polymorphism in the micas it was evident that the long standing differences of opinion as to the correct axial ratios and β angle were a natural result of studies by investigators unaware of the fact that there are approximately as many correct interpretations as there are polymorphs. For almost a century, mineralogists had tried to obtain one set of constants applicable to all the micas, or in other words, to force unimorphism onto a group now known to be characterized by polymorphism. Muscovite is the only mica for which a set of constants may be obtained, as it alone, for all practical purposes, crystallizes in but one polymorphic form. Hendricks and Jefferson (1939) showed how the β angle, for instance, varies with the number of layers in the unit cell. If structural identity is contained in one layer, then the β angle will be 100° (approximately); if two layers are required, the β angle will be 95°, and will be 90° if there are three or more layers in the unit cell. With these problems in mind, Peacock and Ferguson (1943) reviewed the whole field of morphology of the micas and muscovite in particular. In the following discussion constant reference is made to their work. Goniometric measurements of lepidolite and other micas are few compared with those for muscovite, and for that reason they are practically omitted in the following discussion. However, a complete understanding of the morphology of mica will be forthcoming, as the problems and methods are essentially the same as those for muscovite.

Before attempting to explain the various crystallographic settings used in the past it would be best to present again the approximate axial ratios for the more common mica polymorphs based on unit cell dimensions determined by X-ray methods.

One-layer polymorph $\quad a_0 = 5.3\text{Å} \quad b_0 = 9.2\text{Å} \quad c_0 = 10.2\text{Å} \quad \beta = 100°$

axial ratio $\qquad\qquad$ a:b:c $\quad = 0.58:1:1.11$

Two-layer muscovite $\quad a_0 = 5.3\text{Å} \quad b_0 = 9.0\text{Å} \quad c_0 = 20.0\text{Å} \quad \beta = 95°30'$

axial ratio $\qquad\qquad$ a:b:c $\quad = 0.58:1:2.22$

Three-layer $\qquad\qquad a_0 = 5.3\text{Å} \quad b_0 = 9.0\text{Å} \quad c_0 = 30.0\text{Å} \quad \beta = 90°$

axial ratio $\qquad\qquad$ a:b:c $\quad = 0.58:1:3.26$

The first crystallographic settings in general use were those of Miller. Peacock and Ferguson (1943) note:

"In Brooke and Miller (1852, p. 389) biaxial mica, as distinct from uniaxial or nearly uniaxial biotite is described as monoclinic, with calculated angles which give elements that compare with the ratios of the single-layered monoclinic lattice cell:

$\qquad$ a:b:c = 0.5774:1:1.1021 $\qquad$ = 99°58' $\quad$ Miller

$\qquad$ a:b:c = 0.58 $\quad$:1:1.11 $\qquad\quad$ = 100° $\qquad$ Structure

Thus Miller chose elements which are appropriate to single-layer micas, and may perhaps correspond to the structural type of the Vesuvian crystal which yielded the measurements, by Phillips, that were adopted by Miller. However, Miller's setting is unsuitable for the two-layer muscovite structure."

Des Cloizeaux (1862, p. 485) developed the most widely used settings for the micas. They were adopted by Tschermak, Dana, Groth, Goldschmidt and others. Dana's (1892) values are:

$\qquad$ a:b:c = 0.57735:1:3.3128 $\qquad\qquad\qquad \beta = 89° 54\ 1/6'$

These values are in very close agreement with the values of the 3-layer polymorph which again are not suitable for muscovite and most micas. However, Tschermak (1877, p. 43) gave a special setting for muscovite from which the following ratios were computed by Dana (1892, p. 621):

$\qquad$ a:b:c = 0.5775:1:2.2175 $\qquad\qquad\qquad \beta = 84° 55'$

These values are in general agreement with the established constants based on X-ray studies. Even though this setting had certain advantages in simplicity of indices, Tschermak (1877) finally discarded it and returned to the incorrect settings of Des Cloizeaux (1862) in order to be consistent with other measurements. In an attempt at justification, Dana (1892, p. 620) remarks:

"The position here taken is that finally adopted by
Tschermak (priv. Contr., May 1891) as the most satis-
factorily showing the relation to other micas, the
chlorites, etc."

Kokscharov (1875), after studying many measurements, came to the conclusion
that all the micas were monoclinic with a β angle of exactly 90°. He remarks
(p. 235):

"Alle Glimmer überhaupt, ohne Ausnahme, wie die Optiker
es behaupteten, zu dem rhombischen System mit einem
monoklinischen Typus, oder richtiger--zu dem monokli-
noëdrischen System mit dem Winkel γ (Zwischen der Ver-
tical- und der Klinodiagonalaxe) = 90°0'0" gehören."
(Kokscharov's angle γ is now referred to as β).

Kokscharov's axial ratios are (in modern form):

$$a:b:c = 0.5774:1:1.6452 \qquad\qquad \beta = 90°0'$$

They do not correspond with any known type, but, if c is doubled, the result,
a:b:c = 0.5774:1:3.2904, compares favorably with the values for the 3-layer
polymorph. These settings are of importance because they were adopted by
Hintze (1897) for all the micas and by Baumhauer (1900) for muscovite.

Goniometric measurements on muscovite have been made by Marginac
(1847), Kokscharov (1875) Tschermak (1877), Baumhauer (1900), and Peacock and
Ferguson (1943). Early measurements were often inaccurate and resulted in the
proposal of several doubtful forms. Peacock and Ferguson (1943) regard the
measurements of Baumhauer (1900) as superior and, after the transformation of
his elements from the unsuitable settings of Kokscharov (1875), calculate

$$a:b:c = 0.5764:1:2.2372 \qquad\qquad \beta = 95°39'$$

which is considered representative of the geometrical elements of muscovite.

In measurements of their own, Peacock and Ferguson (1943) have
recorded the forms c,b,m,y,p,h,ϵ, and o. The following table lists the goni-
ometric measurements for the forms they found.

TABLE II

PEACOCK AND FERGUSON (1943)
MEASUREMENTS ON MUSCOVITE

Form	Measured Mean	Calculated
cb = (001):(010)	89°57 1/2'	90°00'
cy = (001):(021)	77 13	77 20 1/2
cm = (001):(110)	85 02	85 06
cϵ = (001):(112)	61 45	61 53
co = (001):(111)	72 46	72 46 1/2
ch = (001):($\bar{1}$12)	69 54 1/2	70 03
cp = (001):($\bar{1}$11)	82 13	82 06 1/2

Other forms have been established by other investigators but were not found on the crystals measured by Peacock and Ferguson (1943). They consist of (after transformation):

n(130) reported by Marginac (1847) and Tschermak (1877)

e(011) reported by Tschermak (1877)

x($\bar{1}$31) reported by Tschermak (1877)

Calculated angles (by Peacock and Ferguson, 1943) for these forms are

cn(001):(130)   87° 10'

ce(001):(011)   65 48 1/2

cx(001):($\bar{1}$31)   85 25 1/2

Doubtful forms are (032) ($\bar{3}$02) and (221)

Peacock and Ferguson (1943, p. 75) illustrate a stereographic projection of the established forms for muscovite and also show a drawing of a crystal which consists only of the forms observed by them (Figs. 3, 4).

Twinning in muscovite is common and is generally described according to the mica law. In this law (Ford, 1932) the twin plane is in the zone cM
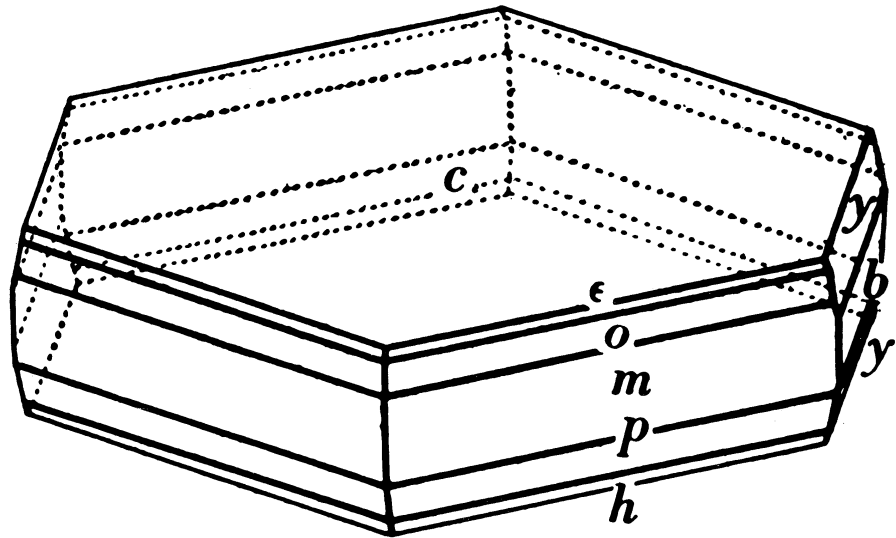
Fig. 3. Stereographic projection of established forms
for muscovite after Peacock and Ferguson (1943).
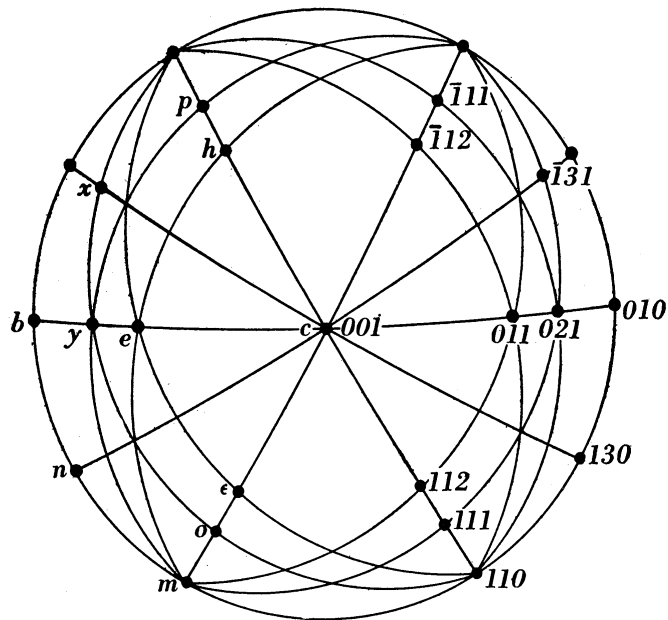
Fig. 4. Crystal drawing of muscovite forms found by
Peacock and Ferguson (1943).

$001_\wedge 221$ and normal to c(001), with (001) as the composition plane. Peacock and Ferguson (1943) state this law in a more general manner (p. 78):

> "The two individuals are in contact on a common plane c(001) and symmetrical by reflection in a plane (hh$l$) which is perpendicular to c(001). Reflection in (hh$l$) gives a "right" twin which is distinct from a "left" twin resulting from reflection in (h̄h̄l)."

In the setting adopted by Hintze (1897), for example, the reflection plane is (110) which in the correct muscovite setting is (33$\bar{1}$). On the basis of the new geometrical elements for muscovite, Peacock and Ferguson (1943) have shown that the twinned crystal is not symmetrical by reflection in the basal plane (001) for the calculated angle (001):(33$\bar{1}$) = 89° 22 1/2'. Reflection in (33$\bar{1}$) would thus require an angle of 1° 15' between the basal cleavages of the twin elements. This discrepancy may be avoided by defining the twin law of muscovite as twin axis $[\overline{3}10]$, composition plane (00$\underline{1}$). The basal planes of the two individuals are thus brought into exact parallelism.

Another type of twinning of more importance to this study may explain the occurrence of uniaxial (and small 2V) muscovites, such as that described by Axelrod and Grimaldi (1949), and lepidolites. It may be defined as twinning axis $[001]$ and composition plane (001) and is discussed in detail with reference to uniaxial lepidolite (page 135).

3. <u>Interferometric Data</u>. The work of Tolansky (1948) has demonstrated that it is possible to obtain some information on various characteristics of mica by means of the application of multiple-beam interferometric techniques using Fizeau fringes and fringes of equal chromatic order. Data may be obtained regarding: (1) exceedingly minute topographic details of cleavage surfaces, including the direction of the cleavage step and the height of the step, (2) the thickness of the mica sheet studied, (3) the birefringence of the mica over a rather extended wave-length range, provided the refractive index is known for one wave length, (4) identification of various types of inclusions, among them solid inclusions of other minerals and vacuoles, and (5) variations in chemical composition.

There has also been an attempt to correlate the structure patterns as revealed by multiple-beam interferograms of doubly silvered mica sheets with electrical power factor and dielectric strength. Tolansky has demonstrated that muscovite from different localities, for example, Bihr, India muscovite as compared with Brazilian muscovite, gives different and typical interferometric patterns that are characteristic within a moderate range of variation. He has also attempted to show that these patterns differ among the various mica species: that muscovite patterns differ from those of phlogopite and biotite. The differences in pattern are due to variations in three features: (1) the

variation in average size of the area of uniform color tint, (2) the number
of inclusions per cubic centimeter, and (3) the average size of the inclusions.

The correlation between interferometric pattern and electrical prop-
erties is as yet apparently not complete or else the published results have
not come to the writers' attention. However, it is suggested that a correla-
tion may be expected between the dielectric power loss of the mica and the
number and size of the inclusions, which, although they are often transparent
because they consist either of liquid or gas or a combination of the two, are
nevertheless strikingly revealed by the interferograms. Although this tech-
nique gives promise for a very exact grading of muscovite, it remains to be
demonstrated, in our opinion, that the distribution, number, and size of these
inclusions is relatively invariable in single sheets, in single books, or in
books from single deposits, or else is variable within rather narrow limits,
so that a technique that employs mica films of the order of 0.01 mm in thick-
ness can be said to sample effectively either a single book or a deposit with-
out requiring an inordinate number of samples.

This technique has also apparently demonstrated that a few varieties
of mica consistently can be cleaved to yield relatively large areas of very
uniform tint, which according to Tolansky, implies that the mica over this area
has been cleaved true to a single molecular plane, not only on one side of
the film, but on both sides of it. Such perfect mica sheets are of value as
windows or absorbers in radioactive experiments. One of the micas tested by
Tolansky, which was from Tanganyika, showed a uniform area of 26 cm^2.

It would be very interesting to explore further the results obtained
by multiple-beam interferometric techniques using a much larger number of sam-
ples from a great many different localities closely correlated with chemical
analyses and optical studies.

Within the past few years a rather considerable amount of study has
been undertaken with respect to the screw-dislocation theory partly by means
of interferometric methods. The theory and mechanism of growth by this method
have been described in detail by Frank (1949). Dislocations have been reported
by various investigators in single crystals of beryl, silicon carbide, paraffin,
and very recently in micas (Amelinckx, 1952A). Apparently dislocations may
explain not only certain external features such as spirals, but also, according
to its advocates, polymorphism in SiC and the micas. Briefly, the theory ex-
plains growth in natural crystals whereby the process of nucleation proceeds
in such a manner that a spiral effect is produced; important factors in this
process include height of the unit cell, supersaturation, and so forth. It
appears quite probable that future work along these lines may yield important
information concerning the cause of polymorphism. The results reported so far
are quite convincing.

C. Optical Properties

1. <u>Indices and 2V</u>. For normal muscovite the general ranges of indices of refraction are as follows: α = 1.552-1.574, β = 1.582-1.610, γ = 1.587-1.616. $\gamma-\alpha$ = 0.036-0.049. However, values both below and above these ranges have been measured and reported. For example, values for α as low as 1.542 and 1.544 are recorded. Values for β as low as 1.579 and for γ as low as 1.578 have been reported. 2V = 30° to 47°, but some phengites have 2V that range from about 20° to essentially 0°. All muscovite is (-); $\alpha \wedge c$ = 0° to -2°, $\beta \wedge a$ = 1° to 3°, γ = b. The optic plane is normal to (010) r > v weak to distinct.

a. <u>Variation of optical properties with composition</u>: There have been several attempts to relate the variation in refractive indices and 2V to variations in chemical composition in the muscovites. Some of the more notable attempts are those of Gädeke (1938) who drew an essentially straight line showing a direct increase between values for β and weight percent Fe_2O_3, embracing only six points. A more elaborate attempt has been made by Winchell, and the most recent results of his efforts are embodied in Fig. 254 of his textbook (Winchell and Winchell, 1951, p. 368). Winchell correlates variation in optical and physical properties with percentages of three "end-member" molecules, muscovite, picrophengite and ferrimuscovite. His diagram shows an essentially straight-line relationship between increasing β index of refraction and content of ferrimuscovite molecule. Another noteworthy attempt at correlation has been by Volk (1939).

It is certain that the refractive indices increase markedly with relatively small increments of ferric iron, but it also seems likely from our studies that a lesser rate of increase is brought about by increasing ferrous iron. Thus the refractive indices of muscovite increase considerably with an increase in the Fe^3:Al ratio and also increase to a lesser degree with an increase in the Fe^2:Mg ratio. Very likely the substitution of Mn for Mg also brings about a slight increase in the refractive indices. Many muscovites contain both ferrous and ferric iron, and the difference in the amounts of increase of the refractive indices caused by equal numbers of ferrous-iron atoms and ferric-iron atoms has not yet been established. We have made numerous plots of β values against weight percent FeO, against weight percent Fe_2O_3, against weight percent FeO + equivalent Fe_2O_3, and against weight percent Al_2O_3, and although all of these display a general trend, none shows a satisfactory direct relationship between increasing refractive indices and either increasing weight of the oxide or decreasing weight percent of the oxides, the latter in the case of Al_2O_3. For these plots we have employed as many as about 60 analyses, including those of Kunitz (1924), Jakob (1925B, 1929A, 1929C) and Volk (1939), as well as some of our own and some by other investigators. One thing seems evident from these plots, and that is that there is a tremendous divergence in the quality in chemical analyses of muscovite. Possibly

also the results indicate that there is a considerable variation in the degree of accuracy of the measurement of refractive indices of muscovite.

Probably the above relationships hold for the birefringence which increases markedly with an increase in ferric iron and to a much lesser extent with increasing ferrous iron and manganese.

The size of the optic angle decreases in a general way with an increase in magnesium, but also decreases in a general way with an increase in ferrous iron and possibly also ferric iron. Conversely, this relationship may be stated by saying that those micas which most closely approach the ideal composition of having little or no magnesium, ferrous iron, or ferric iron, are those with the largest optic angles.

We have also plotted 2V and 2E values measured on micas analyzed by Volk (1939), Jacob (1925B, 1929A, 1929C), Kunitz (1924) and others, as well as micas analyzed specifically for this investigation, against weight percent Fe_2O_3, weight percent Al_2O_3 and weight percent Fe_2O_3 + FeO, and again, although general trends are apparent, the results are far from expressible as a straight-line function. It is interesting to note, however, that if one plots the 2E values obtained by one investigator, in this case those of Kunitz (1924), against the results of his analyses, a much closer approach to a straight-line function is obtained. This may again indicate a considerable difference in quality in both the chemical and optical results by different investigators.

These general optical-chemical relationships are born out by the recent study of Heinrich and Levinson (1953) on rose muscovites. These muscovites approach very closely the ideal muscovite composition, containing generally less than 0.5 percent of combined Fe_2O_3, FeO, MnO, and MgO. They consistently have slightly higher 2V values than other muscovites and have consistently lower refractive indices than other muscovites.

b. Changes of optical properties with temperature: Rinne (1925) has reported that muscovite becomes almost uniaxial when heated in air for about three hours at 1000°C. A second heating at 1000°C restores the original 2E. This transformation may be accompanied by structural changes, the character and extent of which are as yet unknown.

We attempted to repeat this phenomenon but were unsuccessful. About two dozen runs were made under various conditions (i.e., in air and vacuum) in a high-temperature furnace capable of maintaining a temperature as high as 1200°C within 2 or 3 degrees indefinitely. The results indicated that at about 700°C the specimens (muscovite) change considerably in external physical appearance through the loss of color and through expansion. A decrease in 2V also accompanies the physical changes. It appears logical to assume that when water is driven out of the structure, or the iron is oxidized, the above changes take place. On cooling or reheating, the micas did not resume their original properties.

Others have studied various other physical and structural effects on micas heated to various temperatures under varying conditions. Powell and Griffiths (1937) noted that the thermal conductivity of muscovite varies little up to temperatures of 600° (the maximum tested), whereas that of phlogopite decreased as much as 1/3 the initial value at 200°C. On cooling, the reversibility was incomplete. Wood (1937) showed, by means of X-rays, the interdependence of thermal and structural characteristics. Those phlogopites which exhibited a decrease in thermal conductivity at elevated temperatures, also showed displacement of the elemental units (layers) from the normal position at approximately the same temperature. Apparently the atomic arrangement was not affected. With the advent of differential thermal analysis many investigators have studied the micaceous and clay minerals (particularly the fine-grained varieties) by means of this technique. Very little new information has been reported, however.

2. <u>Variation of Color with Other Optical Properties and Composition.</u> Muscovites show considerable variation in color, ranging from essentially colorless through shades of brown and buff (the so-called ruby- and rum-colored mica) to shades of green. The problem of color nomenclature has been studied in a systematic way by Jahns (1945) and by Jahns and Lancaster (1950). The chromophores in muscovite are Fe^2, Fe^3, Mn^3, and Cr. It is possible that in a very few cases Ti may also play a very minor role. It has been demonstrated by Heinrich and Levinson (1953) that the rose muscovites owe their color to the essential absence of Fe^2 and that Mn is dominant over Fe^3. Under such conditions it is possible for Mn to be oxidized to the higher valence, in which state it becomes a powerful pigmenting agent, even though present in very small amounts. The dark-brown micas and those of reddish-brown tints apparently owe their color mainly to Fe^3. This is particularly true for the very dark-brown micas which often have magnetite and hematite inclusions, probably due to exsolution. The green color of most muscovites is believed to be due to the predominating presence of Fe^2. When either Fe^2 or Fe^3 is present in amounts of about 1 percent or more of the oxide, the pigmenting power of any Mn present is essentially masked. The colorless or nearly colorless muscovites apparently owe their lack of color to a close balance between the amounts of Fe^2 + Fe^3 and the amount of Mn. The coloring effect of chromium is well illustrated in fuchsites and mariposites, which owe their bright-green color to even relatively minor amounts of chromium.

Within a single pegmatite that contains both green and brown or ruby muscovites, the green variety consistently shows the larger 2E and will generally have lower indices of refraction than the brown variety. This is in accord with the relationship between increase in Fe^3 with increasing indices and decreasing optical angle.

In Table III the 168 analyzed muscovites have been classified into 2 main color groups: (1) green, olive green and greenish brown, and (2) light brown and ruby. Although the assignment for some specimens is somewhat arbitrary and the two color categories are undoubtedly somewhat

oversimplified, the color differences of the two are reflected in remarkably consistent chemical differences. Green muscovites are considerably richer in total Fe (expressed as Fe_2O_3) and in Mn, whereas ruby muscovites contain more Ti and usually more MgO. Mg is not considered to be a chromophore element. Cr, normally a green chromophore, actually is more common and abundant in the ruby muscovites.

It is, of course, not possible, by spectrochemical analytical techniques, to analyze separately for Fe^3 and Fe^2. Green muscovites, although they contain more total iron, have generally lower refractive indices than ruby muscovites. The ruby muscovites owe their higher indices to their greater content of Fe^3 and Ti, both of which cause much more marked optical changes than equivalent amounts of Fe^2.

TABLE III

VARIATION OF COLOR WITH COMPOSITION IN MUSCOVITE (NEW DATA)

For each district and composition, two numbers
are given: the top number is the average per-
centage for green, olive-green, and greenish-
brown muscovite; the bottom number is the average
percentage for light-brown and ruby muscovite.

Districts	Avg. * % Fe_2O_3	Avg. % MgO	Avg. %TiO_2	Avg. % MnO	Avg. % Cr_2O_3
1. Franklin-Sylva district, North Carolina and north Georgia	3.59 2.55	0.97 1.18	0.126 0.376	0.082 0.053	
2. Spruce Pine district, N.C.; Shelby-Hickory district, N.C.; Thomaston-Barnesville district, Ga.; Alabama district	4.35 2.09	1.21 0.98	0.111 0.214	0.056 0.045	
3. Average of micas in districts 1 and 2	3.88 2.39	1.07 1.12	0.120 0.323	0.072 0.049	
4. New England; Black Hills, S.D.; Colorado; Petaca district, N. Mex.; Montana; Latah Co., Idaho	4.07 2.38	0.23 0.99	0.097 0.104	0.388 0.072	
5. Average of all muscovites	3.92 2.46	0.68 1.09	0.109 0.281	0.215 0.056	0.0007 0.0255

*Total Fe, expressed as Fe_2O_3

D. Zoning

Color zoning is very common in muscovite, particularly core-margin types, with usually brown or ruby cores and greenish-brown or lighter-brown rims. Oscillatory zoning also occurs, usually with thin darker-brown zones alternating with broad light-brown bands, although less commonly green and brown zones alternate. Such zones are parallel with crystal outlines and thus are hexagonal or rhombohedral in outline. Some such zoned crystals also show concomitant color variations in pairs of sectors which join at the center and probably indicate differential absorption during growth. Some ruby micas are characterized by oriented sets of closely spaced darker-colored streaks arranged at right angles in a cross hatch or grating pattern which resembles the gridiron twinning pattern of microcline. Cleavage sheets of such crystals also may show a division into three pairs of sectors, each pair being characterized by different orientation of the grating streaks. The streaks in adjacent sectors are oriented at 120° to 60° to one another. The sectors may result from twinning, and the grating pattern may represent embryonic exsolution.

Color zoning may involve the c-axis direction with sheets at one end of the crystal differing in color from those at the other.

Some muscovites that are marked by A structure show color streaks parallel with the reeves, somewhat different in shade from the rest of the sheet.

Muscovite also commonly displays inclusion zoning of hematite, usually with stained cores and clear margins, although alternations of stained and clear zones also are recorded.

Some examples of different types of zoning in muscovite are noted below.

Minas Gerais, Brazil. "Symmetrically zoned color patterns, hexagonal in outline, are sharply defined and most commonly are composed of recurrent zones of two different tints of ruby. Rarely, a rhomb-shaped ruby core is surrounded by hexagonally shaped zones of lighter-colored ruby. In some specimens, cores of ruby mica are surrounded by zones of green and yellow-green mica. Three well-defined linear patterns are known. In one, linear bichromatic streaks are oriented along two sets of reeves; in another, called checkerboard or plaid, the bichromatic lines are at right angles to each other; the third, called 'hen track' ... has two sets of streaks parallel to the adjacent reeve sets, and the third is parallel to the limiting crystal face (010 or 110)." Two irregular color patterns have also been found in this district (Pecora, Klepper, and Larrabee, 1950).

Ejiba, Nigeria. Jacobson and Webb (1946) report three-dimensional color zoning in muscovite from an albitized pegmatite. At one end of a crystal the muscovite is apple-green and at the other it is lilac. The colors grade into each other along the length of the c-axis. The only chemical difference in the two types is a slightly higher iron-manganese ratio in the former.

San Diego County, California. Some zoned crystals have been reported from the Pala District, (Jahns and Wright, 1951) with green centers and intermediate zones that are yellow to white.

Madison County, Montana. Stoll (1950) reports sheet mica that is colorless and shows a criss-cross pattern or banding of rum-colored areas.

Taos County, New Mexico. Optical zoning was reported by Heinrich and Levinson (1953) in pink muscovite flakes from the Harding pegmatite. Many of the muscovite flakes have a very thin rim that displays lower birefringence than the main central part of the crystal.

Rio Arriba County, New Mexico. Heinrich and Levinson (1953) also report small flakes of muscovite from the Petaca District that show pink and green colors grading into each other.

Clear Creek County, Colorado. Muscovite from the Ajax pegmatite was reported by Hanley et al. (1950) to show alternating color strips of clear and ruby color.

Southeastern Piedmont Area. Jahns (1945) summarizes the color characteristics of the muscovites of this area as follows:

> a) Many books are color zoned with gridiron or chessboard patterns, concentric color bands, strips, or faint irregular mottling.
>
> b) Nearly all books contain pale rims that are more greenish than the interior. This appears to be secondary zoning.
>
> c) Color variations in individual books are more pronounced where the muscovite is light- to olive-green.

New England. Sterrett (1923) summarizes the types of zoning in New England muscovite as follows:

> "Some muscovite shows variations in color that accord with crystal structure. The variations generally appear in bands

that follow the outline of the crystal. Thus, ... one may
see a dark rum-colored center surrounded by a fringe of light
rum or yellow; ... or the center may be light and the border
zone dark. In some sheets there are alternating bands of
varying color."

Woodward (1951) reports an unusual type of structural zoning in the
green muscovite of the Lord Hill pegmatite near Stoneham, Maine. One
book of green muscovite grades from perfectly normal flat folia on one
side of the specimen into a gray botryoidal folia on the other. The
botryoidal structure may be due to late volume increase because of the
addition of materials to the pre-existing muscovite.

A group of selected specimens showing especially distinct color zon-
ing was selected for optical study. Indices of refraction were measured on an
Abbé refractometer and 2E was determined by the Mallard equation. The optical
data are in Table IV:

TABLE IV

OPTICAL DATA ON SELECTED
ZONED MUSCOVITES

Specimen	Core	Intermediate Zone	Margin
No. 5. Gregory pegmatite, Jackson Co., N. C.			
Color	green		clear
α	1.562		1.565
β	1.598		1.602
γ	1.603		1.612
2E	72°		67°20'
No. 7. Jasper pegmatite, Jackson Co., N. C.			
Color	dark-brown	medium-brown	light-brown
α	1.565	1.564	1.566
β	1.602	1.601	1.601
γ	1.606	1.605	1.605
2E	67°25'	65°15'	62°40'

TABLE IV CONT.

Specimen	Core	Intermediate Zone	Margin
No. 9. New Wolff pegmatite, Franklin-Sylva District, N. C.			
Color	clear		light brown
α	1.569		1.572
β	1.607		1.612
γ	1.613		1.615
2E	54°10'		59°15'
No. 15. Sheep Mountain pegmatite, Franklin-Sylva District, N. C.			
Color	stained		clear
α	1.566		1.567
β	1.602		1.604
γ	1.608		1.608
2E	61°40'		60°10'
No. 46. Ledford Cove pegmatite, Franklin-Sylva District, N. C.			
Color	stained		clear
α	1.562		1.565
β	1.598		1.601
γ	1.607		1.607
2E	62°30'		63°30'
No. 57. Poll Miller pegmatite, Franklin-Sylva District, N. C.			
Color	medium-brown		clear
α	1.560		1.560
β	1.595		1.593
γ	1.604		1.607
2E	70°00'		69°45'
No. 64. Slagle-Drake pegmatite, Franklin-Sylva District, N. C.			
Color	clear		spotted
α	1.564		1.559
β	1.601		1.596
γ	1.607		1.606
2E	65°30'		63°

TABLE IV CONT.

Specimen	Core	Intermediate Zone	Margin
No. 75. Big Ridge pegmatite, Franklin-Sylva District, N. C.			
Color	light-brown		medium-brown
α	1.562		1.566
β	1.599		1.601
γ	1.606		1.606
2E	65°30'		65°40'
No. 82. Big Ridge pegmatite, Franklin-Sylva District, N. C.			
Color	light-brown		medium-brown
α	1.565		1.566
β	1.599		1.603
γ	1.605		1.606
2E	64°50'		64°50'
No. 124. Plumtree pegmatite, Spruce Pine District, N. C.			
Color	medium-green		light-green
α	1.570		1.569
β	1.610		1.603
γ	1.615		1.609
2E	64°15'		67°10'
No. 128. Wildcat pegmatite, Spruce Pine District, N. C.			
Color	dark-green		light-green
α	1.570		1.569
β	1.610		1.606
γ	1.616		1.616
2E	70°00'		67°30'

TABLE IV CONCL.

Specimen	Core	Intermediate Zone	Margin

No. 190. Muscovite pegmatite,
Latah County, Idaho

Color	light-brown		clear
α	1.562		1.562
β	1.598		1.599
ɣ	1.602		1.604
2E	63°15'		63°

No. 374.
Hebron, Maine

Color	medium-tan	light-tan	clear
α	— —	1.563	1.560
β	1.594	1.598	1.591
ɣ	1.603	1.601	1.598
2E	67°10'	65°15'	61°50'

The above data indicate that slight differences in indices of refraction and in size of the optic angle accompany variations in color. The darker colored zones and those that are stained or contain inclusions tend to have higher indices and larger optic angles.

II. MINERALOGY OF VARIETAL MUSCOVITES

A. Oellacherite and Other Reported Barium-containing Micas.

The term oellacherite was first used by Dana (1867) for the barium-bearing mica (4.65-5.91% BaO) from Pfitschthal, Tyrol, originally called margarite by Oellacher (Kenngott, 1860). Chemical analyses of oellacherite are reported by Rammelsberg (1862, 1879) and Sandberger (1875). The occurrence has been described by Weinschenk (1896, p. 471). A specimen of oellacherite (No. 1092) originally from the Karabacek collection, from the type locality, was borrowed from the Harvard Mineralogical Musuem. The oellacherite is in flakes 1 to 2 mm across, and is pseudomorphous after a curved, bladed mineral, probably kyanite, in white quartz. X-ray studies show that it has crystallized with the normal muscovite structure.

A flame test for Ba gave completely negative results. Since the analyses are old, some question arises regarding their validity. Spectrochemical analyses of about 200 muscovites, mainly from pegmatites, indicate

that the BaO content of such micas ranges from 0.0002 to 1.1 percent. Simpson (1932, p. 67) reports a muscovite from Lower Chittering, S. W., Western Australia, pseudomorphous after corundum; this mica is reported to contain 1.16 percent BaO.

The mineral from the type locality has the following optical properties:

$$\beta = 1.576$$
$$\gamma = 1.578$$
$$2V = 43°$$

These values are well within the lower part of the range for normal muscovite. It is to be expected that if significant amounts of Ba are present, the indices should be somewhat higher than those normal for muscovite.

Kultiassov and Dubinkina (1946) have described a vanadium-bearing variety of oellacherite from the Karatau, U.S.S.R., vanadium deposits. Three varieties of the mineral are distinguished:

1. Light-green, silky, fibrous aggregates, intimately intergrown with columnar quartz.

2. Dark-green, pearly tablets imbedded in coarse quartz veins.

3. Dark grayish-green flakes and scales on fault surfaces.

Chemical analyses of the three varieties show similarities to oellacherite with respect to the BaO content and to roscoelite with respect to the V_2O_3 content:

	1.	2.	3.
V_2O_3	6.18	17.92	16.44
BaO	7.15	5.27	7.20

X-ray powder data are presented for the fibrous and flaky varieties (1 and 3). Inspection of the d-spacings reported for the fibrous type indicates the presence of considerable quartz. Practically all the spacings (and comparable intensities) listed on the ASTM card for quartz are recorded. When these lines are subtracted from the list of d-spacings, only weak reflections remain. No mineral with these remaining spacings could be found on the ASTM cards. In many of our samples we have noted that a relatively small amount of quartz contamination will produce marked effects on mica powder patterns. These observations cast considerable doubt on the homogeneity of the material on which the analysis was made.

The reported spacings for type 3 are equally confusing. Although the first line recorded has d = 9.955, which is close to that of an important muscovite line, quartz lines also predominate, and the remaining spacings are unassignable. They do not correspond, however, to the muscovite or even the general mica spacings. If the analyzed material represents the same sort of mixture of which the X-ray powder photograph was taken, then its value is nil. The authors (Kultiassov and Dubinkina, 1946, p. 190-191) admit that (in translation) "The content of SiO_2 in the analyses appears to be somewhat high.

This is because the mineral forms fine intergrowths with quartz, and as a result it was impossible to get completely pure material for analysis." They also suggest that the CaO and SO_3 reported may be due to admixed gypsum. We have compared the lines remaining after quartz lines were subtracted with the powder diffraction pattern of gypsum and find no correlation. There is no record that the analyzed minerals were first examined microscopically for purity.

In view of this information and in the light of the reinterpretation of the X-ray powder data, the identity of this material can hardly be considered well established.

Through the courtesy of Dr. S. O. Agrell, we have been permitted to examine a hithertofore undescribed specimen (No. 1349) of a "titan-oellacherite" from a slag. X-ray studies of this material by means of the Weissenberg method indicate that the mica has crystallized as the 1-layer monoclinic polymorph. Because of the high MgO content (20.19 percent) it seems undesirable to call this mica oellacherite; barian phlogopite appears more acceptable. Furthermore, many phlogopites have 1-layer form, whereas muscovite does not. The formula of this material is:

$$(K_{0.8}, \ Ba_{0.8}Ca_{0.4}, Na_{0.3}) \ (Mg_{4.5}, Fe^2_{0.1}, Mn)(Ti_{0.3}, Fe^3)$$

$$(Si_{4.1}, \ Al_{3.3}, \ Ti_{0.6}) \ O_{21.0} \ (F_{2.4}, \ OH_{0.6}).$$
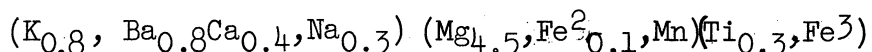
A barian biotite (barytbiotite) with 5.11 percent BaO has been reported by Daub (1913) from Kaiserstuhl, Germany, but the presence of 8.17 percent CaO in the analysis makes its validity suspect. Many analyses of phlogopites with from 0.4 to 1.3 percent BaO have been reported, and, in general, Ba seems to occur more commonly and in larger amounts in phlogopites than in any of the other micas.

A synthetic Ba mica (13.84 percent BaO) has been described by Daimon (1952); it is also a barian phlogopite. The name "oellacherite" should be discarded and "barian muscovite" should be substituted, if it can be shown that the original material actually contains the amount of BaO reported in the old analysis.

Barium-muscovite from Franklin, N. J.: A mineral with 9.89 percent BaO, from Franklin, New Jersey, described by Bauer and Berman (1933), has been called barium-muscovite. Specimens of this material (Nos. 540, 1095, 1127) have been obtained from the Harvard Mineralogical Museum and from the U. S. National Museum. All specimens are massive and pink in color, not unlike some so-called oncosines. The formula, as calculated from the Bauer analysis, is:

$$(K_{1.2}, \ Ba_{0.6}, \ Na_{0.4}, \ Ca_{0.1}) \ (Mg_{0.3}, Zn_{0.2}, Mn_{0.1})Al_{3.4} \ (Si_{5.9}, Al_{2.1})$$

$$O_{20.1} \ OH_{3.9}$$
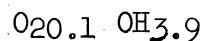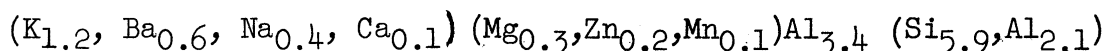
which is in close agreement with the requirements of the formula for a muscovite-type mica. Microscope examination demonstrates the essential homogeneity

of the material. It consists of exceedingly minute flakes of a colorless mineral, finely intergrown. The mean index of refraction is about 1.59; the birefringence is that of fine-grained muscovite. Examination by means of the electron microscope also substantiates the micaceous nature of the mineral. X-ray powder photographs taken of three specimens yielded nearly identical patterns with small differences in intensities of a few lines between the material from the Harvard Collection and that from the National Museum. Although it is of the general mica type, the pattern is unique and is unlike that of any known mica polymorph. Table 54 , column 3, reports the d-spacings for this mica and compares them with those of normal muscovite (Column 1, Table 54 .

The above observations indicate that the material is homogeneous and that it is a mica, but that it can hardly be referred to as a barium-muscovite. Until the exact structure has been determined, the position of the mineral in the mica group remains uncertain. Since its structure is apparently unique, it should perhaps be accorded species rather than varietal status and should receive a name of its own.

In conclusion, it may be said that the status of the barium-containing micas is confused. It appears that a barian muscovite probably does not contain more than 5.91 percent BaO, if the old analyses are reliable. The effect of Ba on the structure remains to be clarified.

B. Ferrian and Manganian Muscovite

A survey of the literature reveals that Fe^{+3} has been reported up to a maximum of 5.70 percent Fe_2O_3 in muscovite (Tschermak 1878). Ferric, as well as ferrous iron, substitutes for octahedral aluminum. Many examples of muscovite with 2 to 4 percent Fe_2O_3 are available in the literature. On the basis of new data obtained by spectrochemical studies obtained for this report, the above observations are substantiated. Rose muscovites and other late hydrothermal micas have the least amount of ferric and ferrous iron, usually less than 0.5 percent. Several high-ferrous muscovites have been reported such as that by Wülfing (1886) (FeO = 6.55 percent). However, high-ferrous muscovites are usually associated with phengites; the rise in Fe^{+2} or Mg, or Mg + Fe^2, as a generalization, accompanies an increase in Si.

The structure of ferrian muscovite is most probably 2M muscovite. This statement is based on X-ray studies of the "alurgite" from Cajon Pass described by Webb (1939) which has been shown by Hendricks and Jefferson (1939), and verified by us, to have the normal muscovite structure; it is actually a ferrian (5.32 percent Fe_2O_3) muscovite.

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."

This or similar material was studied spectrochemically by Erämetsä, Sahama and Kanula (1943), who determined the presence of 1 percent Rb_2O and 0.03 percent Cs_2O. Pehrman (1945) also describes a blue "radial" mica from Matt-kärr, which probably represents similar material. He found it to have 2V = 39°20' by means of the universal stage. A specimen of the Eskola mineral was borrowed from Harvard Mineralogical Museum (No. 87441) and studied. The material is deep purplish-blue in color, very fine-grained, and shows no megascopic structural features.

Microscopic examination reveals that is is a mixture of four different species, in order of abundance:

1. Shreds of colorless, fine-grained mica, n = 1.56

2. Shreds of pale lilac to purplish-gray mica, n = 1.59

3. Quartz

4. Scattered fine grains of a mineral with low birefringence, numerous minute inclusions, and n = ca. 1.63; probably apatite.

The purplish mica, which has the birefringence of normal muscovite, is subordinate to the colorless mica and occurs intergrown with it as scattered clumps of irregular, interwoven flakes, which appear to have been formed at the expense of the colorless mica. Studies of the purple mineral by means of the electron microscope reveal that it is distinctly micaceous.

The powder X-ray photograph of this material also shows the presence of very strong quartz lines. If these are subtracted from the pattern, the remaining lines are probably indicative of a mica, because of the presence of a strong reflection near 10Å; but the structural type is unidentifiable. In view of the optical demonstration that two intergrown micas are present, the pattern may also represent a composite of two polymorphs. From the fine-grained and intimate nature of the two-mica intergrowth and the similarity in their physical properties, it is unlikely that separation of the two can be effected by ordinary mechanical means. A spectrochemical analysis (spec. 1093) of the Harvard material labeled mangan-muscovite is given in Table V. This analysis corresponds with one to be expected from a mica or a mixture of micas contaminated by quartz (high SiO_2) and by apatite (1.05 percent CaO).

It can be stated the purple mineral is a mica, probably of the muscovite type, but it is to be noted that a number of other muscovites with larger amounts of MnO have been reported, which have not been designated mangan-muscovite. The varietal name "manganian muscovite" is preferable, in any event. Although only a maximum of 2.32 percent MnO has been reported in muscovite (Ellsworth, 1932) the writers believe that the manganian-muscovite variety is justified. Most muscovites contain small amounts (< 1 percent)

MnO which undoubtedly occupy octahedral positions. These micas are normal with respect to optical, chemical and structural properties. The maximum amount of manganese that can enter the muscovite structure, or its effects, is as yet unknown.

TABLE V

SPECTROCHEMICAL ANALYSIS
OF MANGAN-MUSCOVITE
(ESKOLA, 1914)

SiO_2	-	49.
Al_2O_3	-	32.
K_2O	-	10.
Fe_2O_3	-	0.15
MgO	-	0.21
Na_2O	-	0.08
MnO	-	1.6
BaO	-	0.0008
Ga_2O_3	-	0.012
SrO	-	0.004
CaO	-	1.05

Analyst: Charles E. Harvey

C. Chromian muscovite

        Until 1946, the chrome micas were in a state of considerable confusion. At that time, Whitmore, Berry, and Hawley reviewed the whole problem and clarified many outstanding inconsistencies.

        Fuchsite was first described by Schafhäutl (1842) for the green chrome mica from Schwarzenstein in the Zillerthal. An analysis of this material is reported in Dana (1882) as containing over 5 percent Cr_2O_3. Six additional analyses are presented in Dana (1892) on chrome micas from Tyrol; Sysevsk; Urals; Belgium; Maryland; Aird Is., Lake Huron; and Ouro Preto; in these analyses Cr_2O_3 is reported from 0.84 to 3.95 percent. Additional analyses on fuchsite from Switzerland have been reported by Prior (1908) with 0.87 percent Cr_2O_3; from South Africa by Partridge (1937) with 0.85 percent Cr_2O_3; from New Zealand by Hutton (1940) with 0.27 percent Cr_2O_3 and (1942) with 4.60 percent Cr_2O_3; from Colorado by Wherry (1915) with 6.08 percent Cr_2O_3 (the highest recorded chromium content of fuchsite); and from Pointe du Bois, Manitoba, by Whitmore, et al. (1946) with 4.81 percent Cr_2O_3. Additional occurrences of material called fuchsite have been reported in the

literature. Typical of recent interest is the occurrence in the Colorado Plateau as discussed by Kerr, et al. (1951).

Partridge (1937) preferred the term "chrome-muscovite" to "fuchsite." Ödman (1938) favored "mariposite" for a low chromium mica even though his material is optically similar to low chromium fuchsites. Hutton (1942) suggested that: (1) the term "mariposite" be abandoned, (2) potash micas with less than 1 percent Cr_2O_3 be described as chromiferous muscovite, and (3) micas with more than 1 percent Cr_2O_3 be described as fuchsite.

Mariposite was named originally for the green mica occurring in the Mother Lode district in California. Analyses of this mariposite and one from Ontario are tabulated by Whitmore, et al. (1946). The feature that distinguishes mariposite from fuchsite is the high $SiO_2:Al_2O_3$ ratio and consistently low (0 to 0.78)percent Cr_2O_3 content. This has been pointed out by Schaller (1916, 1950), Winchell (1925), and Whitmore, et al. (1946). In agreement with Whitmore, et al. (1946), mariposite is considered as chromian phengite.

Very little had been done in the way of X-ray studies of the chrome micas before Whitmore, et al. (1946). Only Pauling (1930) made observations on fuchsite as an outgrowth of his study of muscovite. Whitmore, et al. (1946), on the basis of X-ray studies on material from Pointe du Bois, Tyrol, and Ontario concluded that fuchsite and muscovite are structurally identical. A new chemical analysis on fuchsite from Pointe de Bois yielded the following formula:

$$(K,Na,Ca)_{3.64} \; (Al,Mg,Cr,Fe^3,Fe^2)_{7.92} \; (Al_4,Si_{12})O_{38.61}(OH)_{9.39}$$
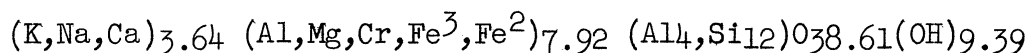
which compares favorably with the formula for muscovite. They further concluded that in fuchsite Cr may replace Al up to 4.81 percent Cr_2O_3 which is close to one atom per unit cell. On the basis of these new data, the term "chromian muscovite" is recommended.

X-ray studies in this laboratory by means of the Weissenberg method were carried out on about one dozen fuchsites including the material from Pointe du Bois described by Whitmore, et al. (1946)and the chromium-containing muscovite described by Hutton (1940). The results obtained are in complete agreement with those reached by Whitmore, et al. (1946); the term "chromian muscovite" is thus adopted. Mariposite is considered by us (in close agreement with others) to be a variety of phengite and as such is discussed under that heading.

The optical properties of fuchsite are, likewise, discussed by Whitmore, et al. (1946). They show the following variations: α = 1.559 to 1.569, β = 1.593 to 1.604, γ = 1.597 to 1.611, γ -α = 0.035-0.042, and 2V=30° to 46°. The average β = 1.598 and 2V = 36°. The optic plane is

normal to (010). X<Y<Z; pleochroism has been noted by some authors as very distinctive, with

$$\alpha = \text{robbin's-egg blue}$$

$$\beta = \text{yellow, yellowish-green}$$

$$\gamma = \text{blue green}$$

In thick flakes, dispersion is noted as distinct with r > v. Whitmore, et al. (1946) conclude that the indices of chromian muscovite (fuchsite) increase with increasing chromium. Some of the indices overlap those of mariposite but are generally lower. 2V on mariposite has been reported as small (0° to 14°).

The chrome micas variously classed as chromiferous muscovite, fuchsite, and mariposite occur chiefly in two types of geological environments: (1) hydrothermal veins and replacement deposits and (2) metamorphic schists, gneisses and quartzites. The first type of geological environment has been subdivided by Whitmore, et al. (1946) into three variations: (1) with ankerite, quartz sulfides, and gold, (2) with biotite and actinolite, and (3) with cordundum, biotite, and kyanite. These investigators conclude(p. 15), that the chrome micas "have invariably been formed either by metasomatic processes or deposited directly with other minerals and veins. Minerals associated with them suggest they were formed under conditions ranging from moderate to fairly high temperatures and pressures...." It may very well be that many of the chrome muscovites, if not most of them, have been formed by deposition from hydrothermal solutions or by metasomatic replacement as these investigators suggest. However, there are numerous examples of the occurrences of chrome micas in regional metamorphic rocks, particularly schists and quartzites in which there is no evidence of metasomatism. Chromiferous muscovite quartzites have been encountered by the writer in several districts in southwestern Montana. The rocks are fine-grained, well foliated quartzites whose color varies from very pale green to deep blue green. The depth of the color is directly proportional to the amount of chromiferous muscovite present. No peridotites lie close to the bands and lenses of green quartz, nor is there any evidence of hydrothermal alteration of these rocks.

Fuchsite also occurs in some marbles as is evidenced by the occurrence at Marble, Colorado, (Wherry, 1915) and the occurrences at Binenthel, Switzerland, in dolomites (Prior, 1908). Several aberrant occurrences of fuchsite are recorded by Taddei (1940). He notes fuchsite in a pegmatite in Tessin, Switzerland. Fuchsite also is reported to occur in the emerald-bearing veins along with calcite, apatite, quartz, pyrite and albite at Somondoco, Colombia.

Thus, although it appears that these additional recorded appearances of fuchsite substantiate the general conclusions of Whitmore, et al. (1946) that most of these micas are of hydrothermal or metasomatic origin, there are apparently also a few cases in which the chromiferous muscovite formed under regional metamorphism without assistance of hydrothermal solutions.

<u>D</u>. <u>Phengite</u>

<u>1</u>. <u>Introduction</u>. The term "phengite" includes several kinds of high-silica muscovites and it is therefore necessary to discuss the varieties individually.

(a) Phengite $K_2(Mg,Fe,Al)_4(Si_7Al)O_{20}(OH)_4$ -- related to normal muscovite but with high silica content and increased Mg and Fe^2 in place of Al. The effect of this substitution on the optical constants is discussed in detail on page 57. Two optic phases: biaxial and uniaxial (hexagonal phengite).

(b) Mariposite (preferably, chromian phengite and hexagonal chromian phengite). Chemically, mariposite has decidedly lower Cr_2O_3 (0.10 to 0.78 percent) and Al_2O_3 contents than fuchsite but decidedly higher SiO_2; the latter feature necessitates it being considered as a variety of phengite. The Al:Si ratio for atoms in four-fold coordination varies between 2:14 and 1:15 as compared with 4:12 in fuchsite. The structural formula of mariposite as calculated by Whitmore, et al. (1946) compares favorably, nevertheless, with fuchsite and muscovite.

The indices of mariposite, on the basis of the available information given by Whitmore, et al. (1946) are: α = 1.56 to 1.58; β = 1.601 to 1.624; γ = 1.60 to 1.63. 2V is usually small but one is reported as much as 40°. The indices of mariposite are generally higher than those for fuchsite of similar Cr_2O_3, and in some cases even higher than for Cr_2O_3-rich fuchsites.

Some mariposite shows pleochroism X = deep blue green and Y = Z paler green. Overlapping of value of the optical constants for mariposite and fuchsite make accurate determination difficult by these methods.

(c) Alurgite--for a discussion of alurgite see pp. 76

(d) Leucophyllite described by Starkl (1883) from Austria, is an exceedingly high-silica muscovite (57.11 percent SiO_2). The

analysis, however, shows only 3.39 percent K_2O (plus 1.42 percent Na_2O) which may indicate that the analyzed material was impure, leached, etc. Schaller (1950) has discussed the series musco-vite-leucophyllite, in which he includes phengite, alurgite and mariposite. He uses the formula $KAlMg(Si)_4O_{10}(OH)_2$, for the leucophyllite end-member; this varies considerably from that reported by Starkl (1883) with regard to alkalies and aluminum.

Schaller (1950), in interpreting the chemical composition of the high-silica sericites, places them into a muscovite-leucophyllite series and demonstrates that they also contain important amounts of a divalent element, usually magnesium. He also notes that their optical properties and specific gravities cannot be correlated with variation in chemical composition. Varieties placed by Schaller (1950) in this series include phengite, mariposite and alurgite (in part).

Other investigators including Whitmore, Berry, and Hawley (1946), Hutton (1940), and Webb (1939) also have discussed the relationships of these micas to each other and to other members of the mica family. However, very little structural work has been done on them. Hendricks and Jefferson (1939) note that uniaxial alurgite from St. Marcel, Italy, has crystallized as the 3-layer hexagonal polymorph, whereas the so-called alurgite from Cajon Pass, California, (Webb, 1939) has the normal muscovite structure. Whitmore, et al. (1946), in their work on the chrome micas, note that mariposite conforms chemically with phengite, which agrees closely with Schaller (1950), and they regard it as a chromiferous variety of phengite. However they had no specimens of mariposite suitable for single-crystal X-ray studies and state that none are recorded by other investigators.

High-silica micas commonly have been reported to exhibit two optical phases, a biaxial and a uniaxial or nearly uniaxial phase, which may be inter-grown in the same sheet. These two optical phases are structurally distinct. The uniaxial type has a 3-layer hexagonal structure whereas the biaxial type has the normal 2-layer monoclinic muscovite structure. Structural determinations can be made either by means of Weissenberg photographs, as demonstrated by Hendricks and Jefferson (1939), or by means of powder photographs, as shown by Levinson (1953).

We have obtained a number of high-silica muscovites suitable for X-ray investigation. This section correlates structural types with optical variations in these micas and presents our conclusions on their classification.

Dr. H. S. Yoder (personal communication) has informed us that the high-silica sericites from Amelia, Va. described by Glass (1935) have the 1-layer monoclinic structure. We have not yet had the opportunity to study these micas. It should be noted that these micas are of pegmatitic origin, whereas the high-silica micas discussed in this section are generally of different paragenesis.

2. Alurgite. Alurgite, St. Marcel, Italy: Both uniaxial and biaxial alurgites from St. Marcel, Italy, the type locality, were X-rayed. The uniaxial sections have crystallized as the 3-layer hexagonal polymorph and the biaxial parts as the normal 2-layer muscovite polymorph. Penfield (1893) noted that 2E was as large as 57°; Larsen and Berman (1934, p. 165) report alurgite, presumably from the same locality, with 2E as much as 98°. The Penfield (1893) analysis, the only analysis of the type alurgite, apparently was made on material of both uniaxial and biaxial character. Penfield (1893, p. 289) states:

"The largest cleavage plates that have been observed are about 15 mm in diameter. When examined in convergent polarized light some show a uniaxial, others a biaxial interference figure, the double refraction being rather strong and negative .... The uniaxial character is undoubtedly the result of twinning, for plates can be found, which in some parts are uniaxial, in others biaxial, and where the plane of the optical axes is in two different positions. Some, moreover, give a confused interference figure, as would be expected from two or more biaxial plates, placed one above the other in twin positions. There is no difference in specific gravity between the uniaxial and biaxial plates, they are identical in behavior before the blowpipe and there is every reason for believing that they are the same chemical substance."

Although it appears possible to simulate a uniaxial interference figure by systematically stacking thin muscovite sheets (Reusch, 1869 and Sugi, 1940), it is structurally impossible to arrange sheets with the 2-layer muscovite structure in any way so that the 3-layer hexagonal structure results.

Although Penfield (1893) states that alurgite is monoclinic, he notes (p. 289):

"When treated with a mixture of strong hydrofluoric and sulphuric acids the biaxial plates show monosymmetric etching figures while those of the uniaxial plates are triangular or hexagonal, similar to those of muscovite and biotite, described by H. Baumhauer."

The 3-layer hexagonal structure on St. Marcel alurgite with 2V = 0° was first established by Hendricks and Jefferson (1939).

Alurgite also is reported to occur at Ceres, Valli di Lanzo, Italy (Zambonini, 1922; Gennaro, 1925).

Alurgite, East Tyrol, Austria: Meixner (1939) reports alurgites from several localities in East Tyrol, Austria. He confirms the presence of uniaxial and biaxial phases in the St. Marcel mica and states that the red mica from "Matrei", East Tyrol, has (p. 697), "Genau die gleichen Eigenschaften ... "

Three specimens of alurgite from this locality were obtained from the Museum of Natural History of Vienna. Our examinations show:

Vienna Number	General Appearance	Optical Characteristics	Structure
J2407	Very minute rose flakes	$2V$ = ca. $35°$	2-layer monoclinic muscovite (Weissenberg).
G5930	Cryptocrystalline, deep red. Assoc. Mn-oxides	Birefringent, 2V not determinable	2-layer monoclinic muscovite (powder)
G5931	Cryptocrystalline, deep red	Birefringent, 2V not determinable	2-layer monoclinic muscovite (powder)

Since chemical analyses of material from this locality are not available, the mineral cannot be classed as alurgite with certainty. The existence of two optical phases whose presence is intimated by Meixner (see above) was not substantiated.

Alurgite, India: Fermor (1909), in a systematic study of the manganese deposits of India, reported four occurrences of red and pink mica which he provisionally regarded as alurgite. A small specimen labeled "Alurgite, Ikabis State, Central India", was received from the Harvard Mineralogical Museum. It bears a general color resemblance to alurgite from St. Marcel; the fragment is biaxial with a large 2V. Weissenberg photographs indicate that it has the normal 2-layer muscovite structure.

Alurgite, Cajon Pass, California: Type material of the alurgite described by Webb (1939) was rechecked by us and the results are identical with those of Hendricks and Jefferson (1939). This mica is not an alurgite, chemically, as has been pointed out by Schaller (1950) (also by Webb, 1939),

for its silica content is that of a normal muscovite. Since its structure
is that of normal muscovite too, it must be classed merely as a ferrian
muscovite. Professor Webb (personal communication) states that he is no
longer convinced that his material is true alurgite.

Alurgite, Ultevis, Sweden: Alurgite described by Ödman (1950)
contains only 43.67 percent SiO_2 and 4.36 percent Fe_2O_3 and is chemically,
therefore, a normal ferrian muscovite. X-ray photographs of flakes of this
mica show that it has the normal 2-layer muscovite structure.

We believe that the term "alurgite" should be dropped entirely and
material of such character be referred to as manganian phengite (or hexagonal
manganian phengite). For the historical treatment, we shall temporarily
continue to use the term alurgite.

3. Mariposite. No mariposite suitable for single-crystal work
could be obtained. However, two specimens of fine-grained mariposite, one
from Carson Hill, Calaveras County, California and the other from San Francis-
quito Canyon, Los Angeles County, California, described by Murdoch and Webb
(1938), have been studied by the powder method. These specimens possess the
normal 2-layer muscovite structure. Unfortunately, the material is so fine
grained and has such poor crystal development that it is impossible to deter-
mine the optical properties with certainty. Although Murdoch and Webb (1938)
state that the mariposite is found in flakes 10 to 12 mm in diameter, these
flakes actually are composites of innumerable, much finer-grained flakes.
However, they show considerable birefringence on basal cleavages, indicating
that they are biaxial. Specimens of mariposite with a small 2V, described
by Knopf (1929), from the Mother Lode area of California could not be obtain-
ed. Sandréa (1950) described a mariposite with 2V<10° from Hoggar, Algeria,
but again samples were unobtainable.

Whitmore, et al. (1946) mention that specimens from the Rawhide
Mine, California, (Harvard No. 86911) have small 2V's. These specimens were
obtained from Harvard for restudy but we are unable to substantiate the pres-
ence of mariposite flakes with small optic angles. The mariposite is ex-
tremely fine-grained, and the few shreds that were isolated show considerable
birefringence. All efforts to isolate enough material for an X-ray powder
photograph were likewise unsuccessful; the photographs show the presence of
quartz and a few extraneous lines.

The table of optical properties of mariposite given by Whitmore,
et al. (1946, p. 14) leaves the impression that a small optic angle is very
common in mariposite. Whitmore, et al. (1946) did not study any of the mari-
posites by means of X-rays. We predict that any uniaxial or nearly uniaxial
mariposite that may be found will be shown to have crystallized as the 3-layer
hexagonal polymorph, for mariposites are merely chromian high-silica musco-
vites.

4. Phengite. Phengites from Italy: The phengite analyzed and described by Pagliani (1937) from Fontane, Germanasca Valley, is reported to have a 2V of about 11°. The 2V of the type material we received is larger than 11°, ranging generally from 18° to 35° (estimated). Weissenberg photographs of sections with 2V near 35° show the normal 2-layered muscovite structure, but photographs of the sections with the smaller 2V have combined patterns of what appears to be the 2-layered muscovite form and the 3-layered hexagonal polymorph. It is probable that patterns of the phengite with 2V = 11° might be those of the 3-layered hexagonal polymorph alone.

Specimens of a mica called muscovite by Jakob (1929B, No. 43) have been received and studied. This mica, from Passo di Ganano, Soazza, Val Mesocco is uniaxial and has crystallized with the 3-layer hexagonal structure. It is a high-silica muscovite (SiO_2 = 49.01 percent), and has the required, relatively large content of a divalent element (MgO = 3.91 percent). This mica was called muscovite by Jakob (1929B), rather than phengite, probably because of its occurrence and paragenesis, for the term phengite apparently has been used by some investigators for high-silica muscovites of secondary origin. Although this mica is predominantly uniaxial, several small areas in a few flakes show birefringence and have a moderate 2V. One such isolated biaxial part proved to have the normal 2-layer muscovite structure.

Phengites with the two-layer muscovite structure: Jakob has analysed five other high-silica muscovites (Table VI) all of which we have checked and found to have the normal 2-layer monoclinic muscovite structure.

TABLE VI: PHENGITES WITH THE TWO-LAYER STRUCTURE
ANALYZED BY JAKOB (1925B,1929B, 1929C)

Jakob's No.	Jakob's Designation	SiO_2	MgO	FeO	Fe_2O_3
18	muscovite	50.79	2.80	1.34	2.94
44	phengite	50.64	4.15	1.01	2.97
45	sericite	48.80	4.74	0.00	5.86
46	sericite	50.20	3.82	0.00	9.62
54	muscovite	50.04	3.64	0.85	2.44

Jakob's (1929B) Nos. 45 and 46, although they have a mica composition and the muscovite pattern as shown by powder photographs, are actually fragments of sericite schist. It was thought that the high silica content of these specimens might be due to admixed quartz. Microscopic examination revealed about 3 to 5 percent admixed epidote in No. 45, but No. 46 is essentially pure mica.

There exist, of course, many additional examples of muscovites described by other investigators, which are silica-rich and probably have only a biaxial, two-layer phase. The examples cited above suffice to demonstrate that not nearly all high-silica muscovites, of various varieties, have small optic angles and the 3-layer hexagonal structure. Many phengites, probably even a majority of them, are biaxial and possess the normal 2-layer monoclinic muscovite structure.

5. Muscovite, Sultan Basin, Washington. The only reported muscovite with a small 2V and without unusually high Si + Mg and/or Fe^2 is the 3-layer monoclinic muscovite described by Axelrod and Grimaldi (1949). A portion of their analyzed material was made available to us for restudy by means of Weissenberg photographs. As explained on page 47, a study of our Weissenberg photographs substantiates the existence of the 3-layer polymorph but leaves the system in doubt, for we do not consider the differences in diffuse scattering to be of sufficient magnitude to warrant classing the structure as monoclinic. If this mica should be considered as being 3-layer hexagonal and if the SiO_2 determination is correct, it will be the only muscovite with small 2V studied by us in which the three-layer structure is not associated with high silica content.

6. Other reported uniaxial or near-uniaxial muscovites. The mica from Mt. Mucrone, Switzerland, described by Kunitz (1924) is uniaxial and is called phengite (SiO_2 = 50.45 percent) by him. Specimens of this material could not be secured for X-ray studies.

Axelrod and Grimaldi (1949, p. 560) note, "Carl Schmidt found, in a gneiss from the Adula Mountains, Switzerland, a green pleochroic muscovite that was uniaxial. It became biaxial on heating and on cooling became uniaxial again." Analyses of what appears to be similar uniaxial material, and also of an associated biaxial mica (2E = 52°) from the Adula Mountains, also collected by Schmidt, have been reported by Wülfing (1886).

	Mica I	Mica II
SiO_2	47.69	47.72
TiO_2	0.11	0.18
Al_2O_3	28.30	25.96
Fe_2O_3	1.02	1.76
FeO	3.88	6.55
MgO	2.72	2.30
K_2O	9.06	10.18
Na_2O	1.87	1.70
H_2O	4.07	3.42
	98.72	99.77

Mica I is reported to be uniaxial; Mica II, biaxial.

A sample of one of these phengites was obtained from the Museum of Natural History of Vienna (No. G3399). From the appearance of the material, it seems to be similar to Mica I. However, optical examination of the sample reveals the presence of two phases: a uniaxial type and a normal biaxial muscovite, which are otherwise markedly similar. Weissenberg photographs of the uniaxial variety again show it to possess the 3-layer hexagonal structure, whereas the mica with the normal 2V has crystallized as the common 2-layer monoclinic muscovite type. In view of this admixture of the two varieties in material supposed to be homogeneous, it is doubtful that the chemical differences reported by Wülfing (1886) are significant. Further proof of the composite nature of the sample is given by a powder X-ray photograph which shows lines characteristic of both polymorphs in approximately equal intensities.

Scheerer (1862, p. 63) also described two uniaxial micas from a gneiss near Freiberg, Germany, which are high in silica, SiO_2 = 50.77 percent and 51.80 percent.

Buryanova (1940) reports a yellowish-green mica from a granitic pegmatite of the Korosten pluton in Volhynia associated with albitized orthoclase and lamellar albite that has 2V = 8° (γ = 1.629 $\pm$ 0.0045). It is pleochroic in shades of light green and occurs with fluorite along cracks in quartz. In the spaces between the sheets of the nearly uniaxial mica is found a lighter green, markedly biaxial mica with strong dispersion of the optic axes (r<v). Buryanova (1940) believes these micas are probably closely related to the potash micas, "gilbertite and cookeite," formed by the hydrolysis of orthoclase. He further notes that light-green micas found with fluorite have marked dispersion of the optic axes, either r>v or r<v, whereas in similar micas unaccompanied by fluorite the dispersion of the optic axes is not to be observed. From this he concludes that fluorine plays an active role in changing the optic properties of green micas. This hypothesis certainly requires further substantiation before it is acceptable.

Clark and Hunt (1915) concluded that the mica in a dolomitic marble from Cockeysville, Maryland, is a muscovite, even though it has the optical properties of a phlogopite. Their conclusion was based on a partial rock analysis which showed a discrepancy in the Al_2O_3:SiO_2 ratio for phlogopite. A specimen of this mica has been X-rayed and the structure was determined to be that of the 1-layer monoclinic polymorph, with intensities identical with those of other 1-layer phlogopites. Thus it is a phlogopite and not a muscovite with a small 2V.

Shibata (1952A, p. 135) notes, with respect to a mica from the Bunsen pegmatite, Korea: "White mica associated with pink lepidolite is

uniaxial negative under the microscope and is considered as phengite." An analysis of this material by Shibata (1925B, p. 150) has SiO_2 = 43.24%, MgO = 0.90%, and FeO + Fe_2O_3 = 1.93%. It appears therefore that he calls this mica a phengite on the basis of its uniaxial character and not because of its chemistry. Shibata (1952B) also describes a "lithium phengite" from the Uruchin Mine, Korea and states that it (p. 153): " ... is optically uniaxial and has high contents of iron and magnesium. These characteristics coincide with those of phengite and it is to be called for (sic) 'Liphengite' by its high content of lithium." Actually this mica also has an essentially normal lithium-bearing muscovite composition, with SiO_2 = 47.44%, Mg = 1.41% and FeO' + Fe_2O_3 = 1.87% and Li_2O = 3.02%. These micas need X-ray study.

Postel and Adelhelm (1944) have described a white muscovite of late hydrothermal origin from the Wissahickon complex in Pennsylvania in which the 2V varies from 22° to 50°. The explanation they advanced is that random shifts in the structural planes of the mica may have some bearing on the low and variable 2V. We have been unable to obtain any of this material for X-ray study but suggest that the small 2V may be due to a rotation of the individual normal muscovite layers similar to that described by Reusch (1869) and Sugi (1940).

7. Discussion. Clearly there is a definite relationship between the formation of the 3-layer hexagonal polymorph and the high silica content of these muscovites. All 3-layer hexagonal muscovites examined by us are of the high-silica type (phengites), but not nearly all phengites have this structure. Apparently a larger number has crystallized as the 2-layer monoclinic, normal muscovite polymorph. The only known possible exception to this generalization is the 3-layer, low-silica muscovite of Axelrod and Grimaldi (1949) which has a normal muscovite composition. Its structure, as has been discussed above, may be interpreted as monoclinic or hexagonal (p.47)

There appear to be three possible causes of the polymorphic variation:

(1) twinning,

(2) isomorphous compositional changes, and

(3) environmental factors.

As has been noted previously, no possible twin combinations of the 2-layer monoclinic structure can be arranged to yield the 3-layer hexagonal structure.

No systematic differences in composition are known to distinguish the hexagonal from the monoclinic phengites. The statement in the Textbook of Mineralogy (Ford, 1932, p. 660) regarding muscovite: "2V variable, usually

about 40°, but diminishing in kinds (phengite) relatively high in silica" is incorrect. Kunitz (1924, p. 383) has stated: "Bei einem weiteren Vergleich dieser Analyse ... lassen sich im allgemeinen keine grosseren Abweichungen in der chemischen Zusammensetzung festsletten, die eine befriedigende Erklä-rung für die sprunghafte Änderung im Achsenwinkel geben könnten." He suggests that the optically anomalous nature of the phengites might be related to details in the arrangement and bonding of certain atoms. There is, of course, no certainty as to whether the few available analyses were carried out on completely uniaxial or entirely biaxial material. This also apparently holds for those by Wülfing (1886). However, from the small-scale nature of the optical variation, probably material of both types was involved in each anal-ysis. The phengite of Pagliani (1937) is a good example, for, although the literature indicates that the analysis was performed on material with a 2V of about 11°, type specimens sent by Professor Pagliani also have a pronounced biaxial phase. At first it was believed that the uniaxial portions of the high-silica muscovites might represent a mica approaching the octo-phyllite group in composition. This was ruled out however, for the calculated atomic ratios indicate that all of these micas fall definitely within the hep-taphyllite (muscovite) group.

It does not seem likely, moreover, that the control of the poly-morphism can be described entirely to compositional variations, as is the case in the muscovite-lepidolite series (Levinson, 1953). Phengites of appar-ently the same composition may or may not crystallize as the 3-layer hexa-gonal polymorph. If the high silica content exercises any control on the structure, it is a permissive rather than a requisite control.

The environmental factors are even more difficult to evaluate. Table VII lists the recorded uniaxial or nearly uniaxial phengites together with their modes of occurrence and available information regarding their origin. Although the data are incomplete, it is striking that none of these micas has formed by crystallization from a granitic or pegmatitic magma. They are either of hydrothermal origin or occur in metamorphic rocks, in which their exact origin is unspecified. Although muscovites of secondary origin are characterized, in general, by a somewhat higher silica content (the so-called high-silica sericites) than magmatic muscovites, there are some peg-matitic phengites. These have axial angles of normal size, and those checked by us possess the normal 2-layer monoclinic muscovite structure (e.g., No. 18, Jakob 1925B and No. 54, Jakob 1929C). These relations suggest that if the polymorphism is controlled in any way by, or related in any manner to, the high silica content, an unknown physical factor, furnished by a hydrothermal or metamorphic environment, also must be active before the high-silica musco-vite can crystallize as the 3-layer hexagonal polymorph. Any such postulated environmental control must be exceedingly delicate in order to account for the small-scale intergrowths of the two polymorphs in the same sheet.

Foster (1952) has observed a correlation in montmorillonites be-tween degrees of swelling and octahedral substitution. In particular the

TABLE VII: OCCURRENCES OF UNIAXIAL OR NEARLY UNIAXIAL PHENGITES AND MUSCOVITES

Name	Locality	Optic Angle	Reference	Occurrence and origin
Alurgite	St. Marcel, Italy	2E=0-57°	Penfield, 1893	Metamorphic manganese deposit
Alurgite	East Tyrol, Austria	Uniaxial and Biaxial	Meixner, 1939	Metamorphic manganese deposit
Mariposite	Mother Lode, Calif.	2E=0-36°	Knopf, 1929	With ankerite in hydrothermally altered serpentinite lenses, near gold-quartz veins
Mariposite	Hoggar, Algeria	2V=10°	Sandrea, 1950	Micaceous schist
Phengite	Fontane, Italy	2V=18-35°	Pagliani, 1937	Mica schist overlying talc lens
Muscovite	Val Mesocco, Switz.	2V=0°	Jakob, 1929B	Alpine cleft vein in eclogite
Phengite	Mt. Mucrone, Switz.	2V=0°	Kunitz, 1924	Gneiss
Phengite	Adula Mtns., Switz.	2V=0°	Wülfing, 1886	Gneiss
Phengite	Freiberg, Germany	2V=0°	Scheerer, 1862	Gneiss
Yellowish-green mica	Volhynia, Russia	2V=8°	Buryanova, 1940	Secondary, in fluorite veinlets in pegmatitic quartz
Muscovite	Wissahickon Complex, Pennsylvania	2V=22-50°	Postel and Adelhelm, 1944	Late stage hydrothermal reaction in granite
Muscovite	Sultan Basin, Wash.	2V=3-15°	Axelrod and Grimaldi, 1949	Veinlets and replacements in granodiorite

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substitution of Fe^{+3}, Mg^{+2} and Fe^{+2} ions for Al^{+3} tends to alter the surface energy of the montmorillonite structure. In the case of the high-silica muscovites it seems possible that the distortion, which Hendricks and Jefferson (1939) indicate as the cause for the invariant muscovite structure, may be correspondingly reduced as a consequence of the above-mentioned substitutions, thus permitting other polymorphs to form. It must be emphasized, however, that a decrease in distortion as shown by diminished intensitites of (06ℓ) reflections with ℓ odd in the lithian muscovite structure (Levinson, 1953) is not to be observed on any of the Weissenberg photographs of the biaxial alurgites and phengites. Nevertheless, it is possible that the polymorphism of the high-silica muscovites may be explained by the fact that these micas, although they approach the ideal muscovite (heptaphyllite) formula, actually are composed of layers with less distortion than those of normal muscovite; the parts that crystallize as the 3-layer polymorph may be relatively undistorted and are thus permitted more choice in stacking arrangement.

The cause or causes of polymorphism in muscovite cannot, at this time, be specified. In order to solve this problem, more uniaxial muscovites must be studied and new analyses of material of uniaxial character must be compared with analyses of material that is entirely biaxial.

E. Lithian Muscovite

In the course of X-ray studies of so-called lepidolites a new variation of the muscovite polymorph has been discovered. A 0-level a-axis Weissenberg photograph of this form is shown in Fig. 6 (compare with Fig. 5). It is apparently confined to "lepidolites" with a low Li_2O content and to muscovites with a relatively high Li_2O content. For this variation, the term "lithian muscovite" is proposed, to distinguish it from normal muscovite. Lithian muscovite is not to be confused with lithium muscovite, the hypothetical end-member used by Stevens (1938) and Berggren (1941). This variety is not common, having been found in only 10 micas of the approximately 500 muscovites and lepidolites studied. The following characteristics of lithian muscovite illustrate its close structural similarity to normal muscovite. Both have:

1. Space group $C2/c$.

2. Cell dimensions (approximate; measured on Weissenberg photographs) $a_0 = 5.2$ Å; $b_0 = 9.0$ Å; $c_0 = 20.0$ Å; $\beta = 95°30'$.

3. (06ℓ) reflections with ℓ odd present.

4. Optic plane perpendicular to (010).
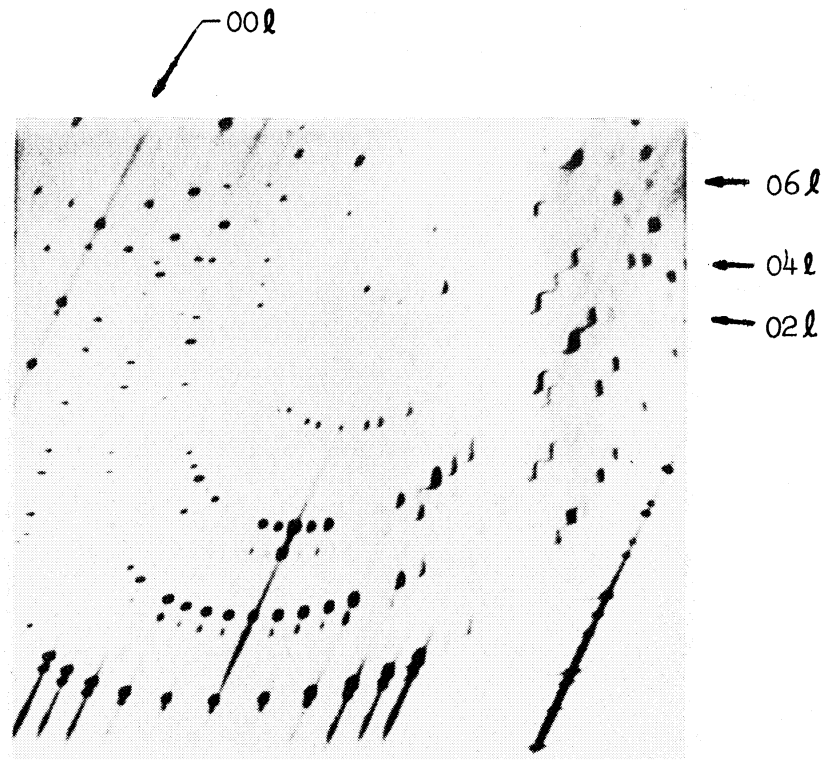
Fig. 5.   0-level a-axis Weissenberg photograph of
   normal muscovite.
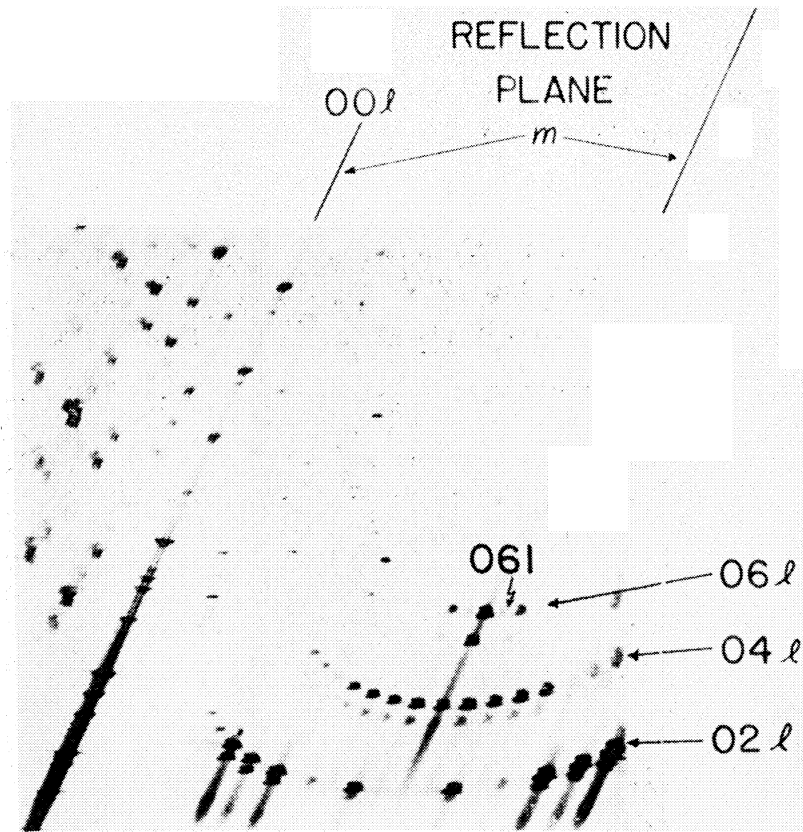
Fig. 6. 0-level a-axis Weissenberg photograph of lithian muscovite.

The following points are different:

1. Indices are in the normal lepidolite range
   α = 1.532; β = 1.552; γ = 1.556.

2. Several differences occur in intensity of reflections.

   The more important intensity differences for (0kℓ) reflections in normal muscovite and lithian muscovite are given in Table VIII. The observed intensities of normal muscovite are those of Hendricks and Jefferson (1939). The lithian muscovite structure has been found in pegmatitic micas from: Tördal, Norway; Newry, South Portland and Topsham, Maine, Eight Mile Park, Colorado; Gunnison County, Colorado; Pala, California; Usakos, South West Africa; Kimito, Finland; and Londonderry, Western Australia.

TABLE VIII: APPROXIMATE OBSERVED INTENSITIES
OF SOME (0kℓ) REFLECTIONS OF NORMAL
MUSCOVITE AND LITHIAN MUSCOVITE

Plane	Normal Muscovite	Lithian Muscovite
020	w	a
022	mw	vw
026	a	vw
045	a	vw
061	w	vw
065	vw	vvw
066	w	vvw
067	vw	vvw
069	w	vvw

   The data in Table VIII demonstrate that on the basis of the presence of (06ℓ) reflections with ℓ odd, lithian muscovite must be considered as having crystallized with the muscovite type structure, but with lesser distortion than in normal muscovite, for most of the (06ℓ) reflections with ℓ odd recorded are extremely weak. This indicates that lithian muscovite approaches more closely the octophyllite micas in structure and composition for, as Hendricks and Jefferson (1939, p. 738) note: these reflections " ... are absent for the two layer biotite-like micas and none is observed for any of the micas that give (h0ℓ) intensities of the single layer structure (except muscovite)."

   A "lepidolite" No. 1 analyzed by Stevens (1938), which contains only 2.70 percent Li_2O, has crystallized with the 2-layer muscovite structure, according to X-ray analysis by Hendricks and Jefferson (1939). Inasmuch as it

contains such a relatively high Li_2O content, Winchell (1942, p. 116) referred to this specimen as a lithium-bearing muscovite. It seemed possible, therefore, that the lithian muscovite structure shown in Fig. 6 was identical with the "lepidolite" analyzed by Stevens (1938) and that Hendricks and Jefferson (1939) neglected to report the intensity differences. Weissenberg photographs of a sample of Stevens No. 1, confirm that this "lepidolite" has the normal muscovite structure as stated by Hendricks and Jefferson (1939). Therefore, Fig. 6 of lithian muscovite illustrates a new variation of muscovite which must contain at least more than 2.70 percent Li_2O.

Rowledge (1945) has briefly described a dozen micas and made partial analyses, including Li_2O. X-ray studies of portions of these analyzed muscovites and "lepidolites" show that all have crystallized as the normal 2-layer muscovite polymorph. Five of the "lepidolites" have Li_2O contents between 2.17 and 2.60 percent. Similarly, three partially analyzed "lepidolites" from Western Australia described by Murray and Chapman (1931) were X-rayed and found to have crystallized with the normal muscovite structure. These micas contain 3.18, 3.24 and 3.32 percent Li_2O. This information, coupled with the results obtained from Stevens No. 1 (2.70 percent Li_2O), and the micas described by Rowledge (1945), substantiates the conclusion that much more lithium may enter the muscovite structure without sensible distortion than has generally been realized.

In a specimen of mica from Newry, Maine, labeled "lepidolite," the lithian muscovite structure grades into that of normal muscovite. The dozen Weissenberg photographs of flakes from this specimen show the critical reflections to vary in intensity between those of normal and those of lithian muscovite. Spectrographic analysis of this material indicated 4.1 percent Li_2O. A spectrographic analysis of a lithian muscovite from Tordal, Norway shows 4.3 percent Li_2O. (These quantitative spectrographic analyses are of a preliminary nature. Future wet chemical analyses may indicate they are in need of revision.) Therefore lithian muscovite probably must contain at least 3.3 percent Li_2O and can contain possibly as much as 4.3 percent Li_2O.

In attempting to explain a form of muscovite with such a high lithium content, one must inquire if (1) it is possible for such a large amount of lithium to replace aluminum isomorphously in the muscovite structure, and if (2) the increased number of lithium atoms are sufficient to cause a reflection, such as (020), which is present in the muscovite structure, to disappear in the lithian muscovite structure. Since the ionic radii of these elements are similar, $Al = 0.57Å$, $Li = 0.69Å$, the substitution is possible and the valence difference may be countered by other substitutions, such as Si^{+4} for Al^{+3}. However, it is not necessary to have isomorphous replacement of aluminum by lithium. Since the muscovite (heptaphyllite) structure has only 2/3 of its octahedral positions filled, with increasing Li content, the vacant positions gradually can be occupied by additional lithium atoms. This implies

the existence of a series between heptaphyllite muscovite with a distorted structure, toward octophyllite lepidolite with an undistorted structure. The presence of extremely weak (06ℓ) reflections with ℓ odd in lithian muscovite supports this concept. If these reflections are entirely absent, the mica has the undistorted 2-layer octophyllite type structure.

It is concluded, therefore, that Li ions, in terms of as much as 3.3 percent Li_2O, can enter the muscovite structure without causing any determinable structural variation. More lithium, probably occupying normally vacant octahedral positions, shifts the structure toward that of the octophyllite micas. In lithian muscovite these changes are already probably of sufficient magnitude to cause a reflection such as (020) to disappear. Actually this hypothesis can only be proved by means of structure factor calculations which have not, however, been undertaken in this study.

The formula of lithian muscovite approximates $K_2(Al,Li)_{ca\ 5.0}$ $(Si_{6-7}, Al_{2-1})\ O_{20}\ (OH,F)_4$; optical properties of representative specimens are presented on page 153. The chemical and structural relationship of lithian muscovite to normal muscovite and the various lepidolites is discussed under the muscovite-lepidolite series (p. 139).

III. OCCURRENCE

A. Granites and Related Rocks

Muscovite is a relatively rare constituent of granites, aplites, and a number of other closely allied acid igneous rocks. Its most characteristic occurrence in granites is with biotite in muscovite-biotite granites (also known as two-mica granites and formerly called binary granites). Muscovite granites that contain essentially no biotite also have been reported, but such rocks appear to be unusual and very rare. In addition muscovite occurs in a group of quartz-rich rocks that have been described under various names, including such types as esmaraldite, beresite, alaskite, greisen, and silexite. Most of these rock varieties, however, are either metasomatically altered granitic rocks or represent quartz-muscovite rocks of hydrothermal origin, either replacement or vein material.

In aplites, muscovite is reportedly the most widespread mica, being more common than biotite, according to Johannsen (1932). However, it is rarely abundant in aplites, and there exist numerous aplites in which it is entirely absent. Likewise there are others that contain both muscovite and biotite. In muscovite-biotite granites the muscovite usually occurs in close association with biotite, not uncommonly in parallel intergrowth or overgrowth with the dark mica. The contacts between the two micas may be those of the crystallographic outline of the biotite, or, more commonly, irregular. These overgrowths or combined overgrowths and replacements have large-scale counterparts in pegmatites. Muscovite also may occur as subhedral to anhedral flakes that cut across biotite flakes and are apparently formed by replacement of the latter. In fact, some investigators believe that most muscovite in two-mica granites has been formed through the deuteric replacement of biotite. In some cases this is attested to by the presence of by-product magnetite and relict biotite. Granites that contain muscovite are usually microcline granites and do not contain a pyroxene, very rarely an amphibole.

B. Pegmatites

1. General. The main rock type and the chief type of mineral deposits in which muscovite occurs is pegmatite of granitic or granodoritic composition. In such deposits mica usually occurs in large crystals, which, if relatively thick and well formed, are known as books. However, many pegmatites of these types may also contain considerable fine-grained muscovite in flakes and scales and also much dense or massive cryptocrystalline muscovite, usually of secondary origin and commonly referred to as sericite.

On the basis of their internal structure, pegmatites may be divided into three main types: (1) unzoned pegmatites in which there is no differentiation into units of contrasting petrology, (2) zoned pegmatites which are characterized by units of contrasting mineralogy or texture or both which are arranged concordantly, generally parallel with the overall shape and structure of the deposit. They commonly show an approach toward bilateral symmetry with respect to a central unit or core. For a complete discussion of the characteristics of zones and zoned pegmatites, reference should be made to Heinrich (1948) and to Cameron, et al. (1949). (3) Complex pegmatites which contain both primary zones and superimposed younger and not uncommonly transgressive or discordant secondary units, such as veins and replacement bodies. Muscovite occurs in all three structural types of pegmatites and occurs in all of the various zone types and secondary units.

2. **Deposits.** However, deposits of muscovite in pegmatites are restricted and may be classified as follows:

a. **Disseminated books in unzoned pegmatites:** These are uncommonly of major commercial significance and normally, the size and number of books are small. The mica books are indiscriminately scattered throughout the entire thickness of pegmatite. The pegmatites are usually coarse grained masses of quartz, sodic plagioclase and/or microcline and of varying, but usually low, percentages of book mica. In a few cases, thin border zones may be present. The mica in some may be somewhat concentrated in a very irregular fashion, closer to the walls or along ill-defined quartzose parts of the pegmatite.

Pegmatites with disseminated muscovite are usually relatively thin tabular bodies or pods of short extent.

b. **Wall-zone deposits:** This type probably is the most common, widespread, and valuable from the standpoint of the total quantity of high-quality mica produced from it. In this type, large books of muscovite occur concentrated in the wall zone of pegmatites together with sodic plagioclase, usually oligoclase, and quartz. Minor amounts of microcline and other accessory constituents, such as biotite, also may be present. Wall-zone muscovite deposits may occur in both the hanging-wall and footwall parts of this zone or may be confined to only one side.

In some pegmatites, particularly those that dip flatly, there are marked differences in color, structural defects, and concentration between the hanging-wall and footwall muscovites. If color differences are present, the hanging-wall muscovite commonly is ruby, whereas the footwall mica is green. The hanging-wall concentration may be richer or leaner than that of the footwall, but often is distinctly richer.

Not uncommonly within the zone there may be second-order concentrations or shoots, which have been localized along a minor structural feature

of the pegmatite such as a pipe-like roll in the contact. In some pegmatites this shoot has been formed along the trough-like keel at the bottom of the pegmatite where the hanging-wall and footwall units merge; in others, the wall-zone concentration lies along a sharp upper ridge or crest.

Where large wall-rock xenoliths have been enclosed, wall zones of mica also may have been formed in somewhat more interior parts of pegmatites.

c. Intermediate-zone deposits: One type of intermediate-zone deposit is the core-margin type; in it, the mica books lie directly adjacent to the central unit, particularly if the core consists of quartz. It is not always certain merely from the position or the shape of this type of deposit whether it is a primary magmatic intermediate zone or whether it represents a replacement unit localized along the core-intermediate zone contact. In flat-lying deposits this designation may become particularly troublesome, for in such, the core-margin unit typically is confined to the footwall side of cores, whereas in steeply dipping or vertical pegmatites it is equally well developed on both sides. Not uncommonly the mica books project into the core quartz and, after removal, leave impressions (mica "footprints").

Where mica zones are developed on both sides of a core, there also may be marked variations in quality and concentration. If the core is developed as isolated pods the core-margin zones are also markedly discontinuous. With this type there may also perhaps be grouped the so-called pod deposits, which are characterized by very poorly developed or embryonic zoning and in which the muscovite is more or less restricted to margins of irregularly defined cores, of coarse-textured quartz, or quartz-microcline bodies scattered irregularly throughout finer-grained pegmatite. Another variant is found in the Alabama mica pegmatite district in which disc-like masses of quartz, usually a foot or less long, are scattered rather uniformly in subparallel position throughout finer-grained pegmatite and are fringed by clusters of small mica books. Both the pod and quartz-plate types of deposits represent types transitional between deposits in unzoned pegmatites and those in some zoned pegmatites.

d. Central-unit deposits: These are relatively uncommon and show several variants. In some pegmatites of the southeastern states the core consists of "burr-rock," a quartz-rich material with scattered, small, partly aligned muscovite books. In a few pegmatites, for example, several in the Alabama district, a narrow central portion is occupied by a tabular muscovite-rich mass which, in some cases at least, may represent the coalescence of the two arms of a core-margin type of muscovite deposit beyond the end of the quartz core.

In certain pegmatites, usually the narrower ones in a district, muscovite books are disseminated throughout a central zone of pegmatites that

are bizonal. In such cases, however, the central unit is not a typical core, but consists of material of wall-zone composition, and thus it is problematic whether deposits of this type should best be referred to as the wall-zone type or the central-zone type of deposit. Where such pegmatites develop quartz cores at lower horizons, the mica not uncommonly remains in the wall zone.

e. Fracture-controlled veins and replacement deposits: Thin tabular lenses or vein-like masses as well as larger, more irregular bodies rich in muscovite can be found cutting across the zonal structure of pegmatites in several districts. Some of these secondary units contain considerable quartz, microcline, and plagioclase, as well as book muscovite, whereas others consist essentially of muscovite. In the narrower veins the mica flakes are commonly oriented parallel to the plane of the fracture, but in some of the larger, more irregular secondary units the muscovite occurs typically in comb structure, with the long axes of the blades more or less at right angles to the plane of the unit. Also in these deposits large irregular masses of small, randomly oriented books ("bull" mica) are common, as well as coarse rosettes and irregular clusters of radially or semiradially arranged blades and books. Wedge and A structure are particularly abundant.

Fracture-guided or contact-guided replacement masses form large and conspicuous units in pegmatites of some areas, transecting and corroding one or more zones and replacing other zones entirely or in part. Such units commonly consist of albite (including cleavelandite) and muscovite, as well as a large number of various accessory minerals. The secondary mineralization is rarely haphazard, however, and usually is related, in position and intensity to the overall shape and structure of the pegmatite or to position and shape of some of the zones. Closely spaced parallel or subparallel fracture sets, commonly curved, may form the basis for the beginning of the replacement. In other cases the contact between the core and the adjacent intermediate zone has served to localize the initial replacement. If the pegmatite is horizontal or flat-lying, core-margin replacement may be unilateral, confined to the footwall side of the core, exclusively.

The intensity of the replacement is commonly strongest along keels or basal troughs of markedly plunging pegmatite bodies, although rolls in hanging-wall contacts also serve, less commonly, as loci of concentration.

f. Combined deposits: Some pegmatites contain mica concentrations in two or more types of units; for example, it may occur in the wall zone and in a core-margin unit, or in a pod-like deposit together with fracture-controlled muscovite veins. Usually in such deposits there are significant structural and chemical differences between the micas from the two different types of units.

3. Exomorphic Mica. In a few districts, relatively large mica books formed by replacement and/or recrystallization occur in the wall rock close to the margin of the pegmatite. In some cases only scattered books are present, whereas in other deposits small local concentrations have been mined.

Other occurrences: Graphic to dendritic intergrowths of fine-grained flaky muscovite with various minerals are common in the outer parts of pegmatites in various regions. The most commonly intergrown species is quartz in which the muscovite forms plumose to dendritic aggregates. Heinrich (1948) has described the occurrence of these widespread and characteristic intergrowths in some Colorado pegmatites. Other less common intergrowths occur with microcline, garnet, and tourmaline.

Fine-grained secondary muscovite (sericite) is also abundant, commonly as a replacement of microcline or plagioclase, but also forming at the expense of tourmaline, beryl, topaz, spodumene, amblygonite, garnet, and many other less common species. In some cases this pseudomorphic muscovite may be in the form of relatively coarse scales and may be the variety rose muscovite (Heinrich and Levinson, 1953).

C. Hydrothermal Deposits

Muscovite is a not uncommon constituent of quartz veins in many districts, particularly in those in which pegmatites are common. Some of these mica-bearing veins are difficult to classify, for they may be somewhat feldspathic and resemble quartz-rich pegmatites. In some examples the muscovite occurs as parallel, coarse flakes near the vein margins and may be at least partly of endomorphic origin. Muscovite occurs in some alpine-type veins believed by Swiss mineralogists to have been formed by precipitation of material leached from wall rocks.

Lithium-bearing muscovites (lithionite) are reported as common constituents of tin veins and greisen. Coarse muscovite is generally believed to be indicative of high-temperature environment for the veins or lodes. On the other hand sericitization of wall rock is most strongly developed accompanying mesothermal deposits. In this connection the work of Roy (1949) may have significance.

D. Metamorphic Rocks

Muscovite is a widespread product of the low-grade regional metamorphism of argillaceous and arenaceous sediments in such rocks as slates, sericite phyllites, chlorite-sericite schists, muscovite quartzites, schistose grits, muscovite schists, and muscovite-biotite schists. Muscovite also

persists not uncommonly into higher-grade rocks such as sillimanite gneisses and garnet-biotite gneisses. Low-grade regional metamorphism of intermediate igneous rocks produces calc schists with albite, sericite, chlorite and epidote. Similar metamorphism of more felsic types results in albite-sericite schists or muscovite schists. Muscovite-glaucophane schists also are known.

Muscovite also forms during the early stages of the contact metamorphism of argillaceous sediments, but with advancing metamorphism, gives way to orthoclase.

During retrograde metamorphism, aluminous silicates are commonly transformed into sericite; these include andalusite, kyanite, cordierite, sillimanite, potash feldspar, plagioclase and staurolite ("shimmer aggregate"). Some investigators insist that these changes require the introduction of potassium, whereas others believe that sufficient potassium is available through the decomposition of the potash feldspar.

Muscovite is also formed by means of low- to medium-grade metasomatism, notably in contact aureoles close to the granite, where it may replace earlier-formed higher-grade aluminum silicates. Exomorphic muscovite also occurs commonly in wall rocks along the margins of pegmatites, for example, in the Petaca district, New Mexico, in the Black Hills, South Dakota, in the Latah County district, Idaho, and in central Colorado. At the Clein deposit in the Alabama district a sheath of fine-grained muscovite flakes has been developed around a body of aplite intruded into biotite gneiss (Heinrich and Olson, 1953).

Muscovite has been synthesized by Noll (1932) at about 200°C. and Roy (1949) has resynthesized decomposed muscovite at 650°C. under high water-vapor pressure, showing that the mineral is stable in such an environment to at least this temperature.

E. Sedimentary Rocks

The micas that occur in sedimentary rocks as authigenic constituents belong chiefly to the illite group and are characterized by a deficiency in K ions, whose positions are partly occupied by hydronium ions (Brown and Norrish, 1952). This investigation has not been concerned with the illite group of minerals. Of course, detrital muscovite and biotite also occur in various clastic sedimentary rocks.

F. Variation of Composition with Occurrence

Several attempts have been made to plot any systematic relationship that may exist between variation in composition and variation in occurrence.

The first unsuccessful plot, made by the senior author several years ago, involved a recasting of several hundred muscovite analyses into three components: $A = Al_2O_3$, $B = FeO + MgO + MnO + Li_2O$, $C = Fe_2O_3 + TiO_2 + Mn_2O_3$. These were recalculated to the basis of 100 percent and plotted on triangular graphs for the following groups: granitic muscovites, pegmatitic muscovites, hydrothermal muscovites, metamorphic muscovites and sedimentary muscovites.

Another similar attempt used as the three components: $A = FeO + MgO + MnO$, $B = Al_2O_3 + Li_2O$, $C = SiO_2 + Fe_2O_3 + TiO_2$. This resulted in a better distribution of the points, but, in general, the spread within each paragenetic group approached the variational range for the entire collection of analyses. Some trends, however, were apparent from the plot: (1) Pegmatitic muscovites show the greatest range of compositional variation. (2) Most of the high-silica muscovites (phengites) are formed under conditions of regional metamorphism. (3) Fine-grained muscovites ("sericites") of low-temperature origin tend to have potassium slightly deficient and water slightly in excess. (4) F and Li appear almost entirely in pegmatitic and in high-temperature hydrothermal muscovites.

IV. PEGMATITIC MUSCOVITE

A. Structural Defects

Many muscovite books are imperfect crystals, containing a variety of macroscopic imperfections.

1. Reeves: Shallow, closely spaced, parallel grooves or narrow corrugations that interrupt the cleavage continuity to various depths. The reeves are oriented parallel with (110) and (010). Several combinations of lines are common: A structure--2 sets of reeves in a V-pattern, intersecting at 60°; double A structure--3 sets of reeves, joining at a common center at angles of 60°; herringbone structure ("fishback" or "fishbone")--2 wide sets of reeves at 120°, bisected by a third, prominent, narrow set; radial reeves--at 60°, six sets radiating from a common center.

2. Ruling: Sets of parallel parting planes, cutting the cleavage at 67° and parallel with (110) and (010). One, two, or three sets may be present, resulting in strips, rhombs, or diamond-shaped pieces.

3. Fractures: Usually curved, sharp breaks that are not crystal-lographically oriented. May be semi-radial from large inclusions, particularly quartz. Extremely fine cracks that appear only as the book is split are called hair cracks.

4. Wedge structure: The books are wedge shaped in cross section, that is, the sheets or laminae are tapering. Wedge and A structure not uncommonly occur together.

5. Warping: Ridges or waves of varying dimensions and extent, from minute, abundant crenulations to broad warps or bucklings. Cleavage steps are sharp minute flexures that result in a stairstep surface on cleavage faces.

6. Block structure: Some mica books consist of slightly disoriented, discrete units whose orientations deviate a few degrees from those of their neighboring blocks. The surfaces of the blocks extend inward to the crystal center. This is one of the causes of imperfect cleavage across the entire book ("locked" or "tangled" mica). Concentric structure is a

variation of this in which small mica plates are so misoriented with
respect to their neighbors that the aggregate forms a curving, hemi-
spherical mass. One way in which such a structure forms is by depo-
sition over the curved surface of a sheave of cleavelandite blades.

1. Muscovite-Biotite Overgrowths and Intergrowths. Muscovite and
biotite commonly occur together in composite books or strips, in which the
two micas have their cleavage planes parallel or nearly so. In some types one
species surrounds or nearly encloses the other, and the boundary between the
two may be a euhedral crystal outline or it may be highly irregular. Commonly,
biotite is enclosed in muscovite. In a few of these the enclosed crystal is
markedly pyramidal in habit and tapers parallel to c, so that at one end of
the crystal the central mica occupies a much smaller part of the sheet than
at the other. Normally the included plate or crystal is bounded by prism and
pinacoid faces. A few rare examples consist of a euhedral muscovite core, an
intermediate biotite strip, and a muscovite margin.

In other composite books there is no regular pattern formed by the
two micas. The cleavages of the two are still generally parallel, but the dis-
tribution of the two types either across the cleavages or parallel to c is not
systematic.

Some examples of these overgrowths and intergrowths are described
below. The only nonpegmatitic occurrence of muscovite and biotite intergrowths
was reported by Johannsen (1938). A nepheline-bearing diorite called dun-
gannonite contains small amounts of yellow to greenish-brown biotite intergrown
with muscovite. However, subparallel intergrowths of the two micas are not
uncommon in two-mica granites.

Composite books of pegmatitic biotite and muscovite, as much as 12
inches across, have been reported from the following representative localities:

Southeastern Piedmont District: The Mitchell Creek Mine, Upson
County, Georgia, has produced muscovite-biotite intergrowths of several va-
rieties. Furcron and Teague (1943) report sheet muscovite books wrapped by
sheet biotite, biotite books included in muscovite, and muscovite included
in biotite. Both species have a common cleavage plane. Lester (1946) found
two types of intergrowths at this locality. The first is a tight intergrowth
in which the two micas possess a common cleavage plane. Thin sheets of this
type can be cleaved as easily across the line of contact as within the in-
dividual micas. The second type is a loose intergrowth with straight boundar-
ies; the two micas possess common planes of cleavage, but tend to separate
readily when cleaved across the contact.

Rum-colored muscovite showing heavy A structure and containing
six-sided inclusions of biotite has been reported by Furcron and Teague (1943)

from the Peters Mine in Monroe County, Georgia. They also found a minor
occurrence of biotite crystals included in sheets of muscovite from the follow-
ing localities in Georgia:

> Dick Fletcher Mine, Monroe County
> W. M. Gooch Mine, Lumpkin County
> W. A. Sullivan Mine, Union County
> Chapman Mine, Elbert County.

In Mitchell County, North Carolina, Sterrett (1923) found crystals
and sheets of biotite with included crystals of muscovite and vice versa.
The two micas generally occur in parallel intergrowth and have a common
cleavage. Studies of a specimen of biotite enclosing a rhombic crystal of
muscovite showed the two micas to have approximately parallel percussion fig-
ures and optic planes.

Some pegmatites of the Spruce Pine District, North Carolina, con-
tain biotite intimately mixed with oligoclase and muscovite. Maurice (1940)
reports that, in the outer zone of the pegmatites, muscovite frequently forms
an outer zone around biotite crystals. Thin, flaky sheets or laths of biotite
occur as veinlets in parallel intergrowth with muscovite. The laths are
usually less than 4 inches long and 1/16 inch thick.

Sterrett (1923) also reported parallel intergrowths of biotite and
muscovite from the Hamilton Mine, Ashe County, North Carolina, the Big Ridge
Mine, Haywood County, North Carolina, and the Chalk Hill Mine, Macon County,
North Carolina. Pogue (1911) found six-sided plates of biotite enclosed in
muscovite at the Buck Creek deposit, Macon County, North Carolina. Examples
also are cited by Jahns and Lancaster (1950).

Northeastern United States: Several occurrences of biotite-muscovite
intergrowths have been reported from New England pegmatites. However, none
of the descriptions are detailed; apparently the occurrences are not common.
Generally, the micas intergrow in parallel position with a common basal cleav-
age. The reported localities are:

> Island Mine, Creshire County, New Hampshire
> Sterrett (1923) and Olson (1942)
>
> Holden Mine, Granton County, New Hampshire
> Sterrett (1923)
>
> Patten Mine, Granton County, New Hampshire
> Sterrett (1923)
>
> Old Lithia Mine, Chatham, Connecticut
> Shannon (1920)

Middletown, Connecticut
Pogue (1911)

Philadelphia, Pennsylvania
Hall (1885)

Lenni, Delaware County, Pennsylvania
Hall (1885)

Western United States: Biotite-muscovite intergrowths from the Western states have been described from a few pegmatites. Hanley, et al. (1950) describe green-gray, heavily stained muscovite from the Rosemont Mine, Micanite District, Park and Fremont Counties, Colorado. The muscovite is ruled, ribboned, has pronounced A structure, and is commonly intergrown with biotite. The same authors also report blades of intergrown muscovite and biotite as long as 6 feet from the School Section Mine, Eight Mile Park, Fremont County, Colorado. Other scattered occurrences were at the following localities:

Levi Anderson Mine, Latah County, Idaho
Sterrett (1923)

Custer County, South Dakota
Pogue (1911)

Canada: Spence (1930) reports parallel intergrowths of muscovite and biotite from dikes of the lower St. Lawrence area. The following three varieties of intergrowths have been found at the Pied des Monts mica mine near Murray Bay, Quebec:

(1) core of muscovite surrounded by a rim of biotite,
(2) books that are part muscovite and part biotite, the boundary running diagonally across the cleavage sheet, and
(3) alternate layers of muscovite and biotite.

Minas Gerais, Brazil: Occurrences of intergrown biotite and muscovite were noted by Pecora, et al. (1950). Generally, the intergrowths are in parallel position and occur in the border zones of the pegmatites.

Germany: Linck (1901) reported regular and irregular intergrowths of biotite and muscovite from Veltlin. He found that the rays of a percussion figure near the boundary would cross the boundary without interference. The two species of micas have a common cleavage plane, but the planes of the optic axes are at an angle of 60°.

Scharizer (1886) described regular overgrowths of muscovite and lepidomelane from the pegmatites of Schüttenhofen, Germany. A six-sided

crystal of lepidomelane is completely surrounded by a rhombic crystal of muscovite. Both species have a common basal cleavage. The axial plane of the lepidomelane is parallel with the long ray of a percussion figure, whereas the axial plane of the muscovite is normal to the long ray of the percussion figure; the two optic planes are therefore at an angle of 60°.

Kimito, Finland: Pehrman (1945) reports weathered books of inter-layered biotite and muscovite from the granitic pegmatite of Kimito.

Orientation Observations: A group of specimens of intergrown musco-vite and biotite was selected from the Michigan mica collection for optical study. An attempt was made to find the relationship between percussion figures and optic planes in the two species. In all cases, the percussion figures are essentially parallel. The greatest deviation from parallelism was found to be 9°. The most common angle between the optic planes of the two species is approximately 60°. However, angles of 0° and 90° were also noted. Generally, the line of contact between species is sharp. In a few specimens, the contact is ragged or feathered. In no specimens was a gradation found between muscovite and biotite. The data obtained are listed below.

Specimen Number	Locality	Angle Between Main Ray of Percussion Figures
Optic Planes Perpendicular		
85	Putnam, Haywood County, North Carolina	0°
Optic Planes Parallel		
149	Mauldin Road prospect, Upson County, Georgia	5°
1393	Location unknown	0°
Optic Planes at an Angle of 60°		
47	Ledford Cove, Macon County, North Carolina	9°
48	Ledford Cove, Macon County, North Carolina	0°
68	Big Ridge, Haywood County, North Carolina	6°

Specimen Number	Locality	Angle Between Main Ray of Percussion Figures

<u>Optic Planes at an Angle of 60° (cont.)</u>

84	Lower East Fork, Haywood County, North Carolina	4°
153	Mitchell Creek, Georgia	3°
177	M and G, Alabama	6°
282	Eight Mile Park, Fremont County, Colorado	0°
1391	Hebron, Maine	0°
1392	Delaware County, Pennsylvania	0°
1394	North Carolina	0°

Jahns and Lancaster (1950) state (p. 15) that the pressure and percussion figures of the inclusions commonly are oriented normal to those of the host.

Conclusion. Grigoriev (1936), who studied the interrelations of muscovite and biotite in some detail, concluded that the similarity in crystal structure, cleavage, and crystal form allows for the possibility of (1) regular overgrowths of one mica by another, (2) oriented intergrowths, and (3) possible mutual replacement.

The relations of biotite to muscovite in the composite crystals can be grouped as follows:

1. Euhedral biotite plates enclosed in muscovite (common).

2. Euhedral to subhedral pyramidal biotite enclosed in muscovite (uncommon).

3. Euhedral muscovite plates enclosed in biotite (rare).

4. Euhedral muscovite in euhedral to subhedral biotite enclosed in muscovite (very rare).

5. Anhedral strips and plates of biotite enclosed by muscovite or irregularly intergrown with muscovite (common).

6. Alternating sheets of muscovite and biotite (rare).

B. Inclusions

1. Magnetite and Hematite. Magnetite and hematite are by far the most common and abundant included minerals. They occur as very thin (usually <0.01 to 0.1 mm) plates, needles, skeletal crystals, and lattice-work aggregates (Frondel and Ashby, 1937) oriented parallel to either percussion or pressure arms. Concentrations of the iron oxides occur usually in the central parts of books, whereas margins are usually clear. Most of the hematite inclusions appear to have formed as the result of exsolution. At the initial temperatures of crystallization the muscovite structure is capable of retaining considerable iron isomorphously. With a decrease in temperature the tolerance is decreased and the iron is ejected and segregated as hematite units. The outer parts of crystals, which formed at lower temperatures than the interior, could hold less iron originally and thus were able to retain all of this lesser amount of iron upon cooling. In books with such inclusion zoning, the muscovite of the clear margins not uncommonly has higher refractive indices (more Fe^2 and/or Fe^3) than that of the core from which the maximum amount of iron has been removed through exsolution.

In pegmatites where muscovite and biotite occur together in the same zone, iron-oxide inclusions are rare in the muscovite, probably because the normally earlier crystallization of the biotite abstracted most of the available iron from the pegmatitic solution.

2. Other Minerals. Numerous other mineral species occur as thin inclusions in muscovite (Frondel, 1936, 1940; Jahns and Lancaster, 1950). The shape and orientation of many such mineral plates or needles have been controlled by the principal crystallographic directions of their host, and the inclusions are primary. Other inclusions display little or no orientation and are randomly intergrown with the mica either as films along cleavages or even transecting the books and thus breaking the cleavage continuity. Such mineral inclusions may be either primary or secondary. Books marked by abundant inclusions of grains or rods of quartz, albite, apatite and a few other species are termed sand-pitted or gritty.

3. Other Materials. Gas bubbles of primary origin, or minute secondary air pockets form inclusions in some types of muscovite. Green amorphous patches and films of undetermined organic material are included in weathered muscovite.

C. Variations in Individual Pegmatites

1. Concentrations. Despite the fact that muscovite can be a constituent of any pegmatite zone and of replacement units, most concentrations

occur in: (1) wall zones, (2) intermediate (core-margin) zones, and
(3) replacement units. Unzoned pegmatites that contain disseminated books
also are of economic significance in some districts. Individual pegmatites
may contain concentrations in more than one unit, although if muscovite occurs
in more than one unit, normally it will be much more abundant in one than in
the other.

The concentration of mica in a single-zone is not always uniform.
In some pegmatites, the hanging-wall part of the wall zone will alone contain
muscovite concentrations. Exceptionally the footwall counterpart alone may
have them. In other pegmatites concentrations within the wall zone are local-
ized along rolls or bulges in the contact, so that mica shoots have been de-
veloped. The margins of xenoliths may serve as a false wall along which a
concentration of wall-zone mica is repeated.

2. Structural Defects and Color. Within an individual zone the
color, degree of staining (iron-oxide inclusions), and structural defects are
usually uniform, although numerous exceptions are known. The Gregory Mine
in the Franklin-Sylva District of North Carolina, for example, contains ruby
muscovite in the hanging-wall part of the wall zone, green muscovite in the
footwall part of the same zone, and where the two merge around the keel of
the pegmatite, the books are color-zoned, some with green cores and ruby
margins.

Commonly, if two different colored micas occur in the same pegmatite,
that of the wall zone is brown or ruby, whereas that of the core-margin zone
is green. If the wall-zone mica is green, then if any core-margin muscovite
is present, it also is green or silvery. Brown or ruby core-margin muscovite
does not occur with green wall-zone muscovite. Muscovite of fracture fillings
and replacement units commonly is green, silvery, or almost colorless. Rose
muscovites are of late replacement origin.

Muscovite-biotite intergrowths are essentially confined to outer
zones of pegmatites, and, generally, wall zones containing green mica contain
little or no biotite. Moreover, in wall zones that contain both biotite and
ruby or brown muscovite, the latter normally is relatively free of iron-oxide
inclusions. Inclusions of iron oxide usually are more common and abundant in
green than in ruby muscovite.

The muscovite of wall zones more commonly is in relatively small,
flat books, free of A or herringbone structure but may contain a relatively
large number of small mineral inclusions (except iron oxides) and unoriented
fractures. In contrast, core-margin mica is characterized more commonly by
relatively large size, widespread A or herringbone structure, and iron-oxide
staining. Mica from replacement units or fracture fillings shows much wedge-
A or wedge herringbone structure and extreme variation in book size. Inter-
mediate zones or pegmatites in which microcline is the dominant feldspar also
contain strongly reeved muscovite.

3. _Composition_. Information is now available on the compositional homogeneity of single muscovite books, on the variation of books within the same zone of an individual pegmatite, or on the variation between muscovites from different zones within individual pegmatites. Table IX shows two analyses on sheets from opposite ends of a single thick book of muscovite from the Tilley pegmatite in the Franklin-Sylva district, North Carolina. They show remarkable uniformity.

In Tables X through XVII are recorded chemical analyses of different books from the same zone of various pegmatites.

TABLE IX

SPECTROCHEMICAL ANALYSES* OF MUSCOVITE SHEETS FROM
OPPOSITE ENDS OF A SINGLE THICK BOOK
(Tilley pegmatite, Franklin-Sylva district, N. C.)

	436 a	436 b
Fe_2O_3	3.5	3.7
MgO	1.5	1.5
MnO	.02	.02
TiO_2	.30	.27
CaO	.002	.004
BaO	.11	.075
Na_2O	1.2	1.25
Ga_2O_3	.015	.015
Cr_2O_3	.0005	.0003
V_2O_5	.008	.005

Absent: Li_2O, Rb_2O, Cs_2O, SrO, Sc_2O_3, SnO_2, Co_2O_3 and F.

* Analyses by C. E. Harvey

TABLE X

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES FROM THE SAME ZONE WITHIN THE BIG RIDGE PEGMATITE
(Franklin-Sylva district, N. C.)

	67	70	72	73	75	76	77	81	82	Volk**
Fe_2O_3	2.2	2.1	2.2	2.2	2.4	2.0	1.8	2.1	2.1	2.09♣
MgO	1.5	1.2	1.2	1.3	1.3	1.3	1.15	1.2	1.3	1.57
MnO	.04	.04	.06	.04	.04	.03	.05	.04	.04	
TiO_2	.16	.26	.27	.13	.15	.21	.21	.17	.16	0.89
CaO	.002	.009	.015	.003	.02	.003	.004	.01	.03	0.80
SrO	--	.003	.003	.002	.002	.002	.003	.002	.002	
BaO	.39	.52	.44	.47	.44	.47	.46	.43	.41	
Na_2O	.64	.72	.60	.60	.55	.97	.67	.75	.81	1.59
Li_2O	--	--	--	--	--	--	--	--	--	
Rb_2O	--	--	--	--	--	--	--	--	--	
Cs_2O	--	--	--	--	--	--	--	--	--	
Ga_2O_3	.017	.017	.02	.02	.018	.017	.02	.022	.015	
Sc_2O_3	.004	.005	.006	.003	.004	.003	.006	.004	.003	
Co_2O_3	.0003	.0004	.0004	.0004	.0004	.0004	.0005	.0005	.0004	
Cr_2O_3	.0001	.0002	.0002	.0002	.0003	.0001	.0002	.0002	.0002	
SnO_2	.0007	.0007	.001	.001	.0008	.0007	.001	.002	.0008	
V_2O_5	.003	.006	.006	.004	.005	.005	.006	.005	.003	
F	--	--	--	--	--	--	--	--	--	

* Analyses by C. E. Harvey

** Am. Min., 24, 255-266, 1939

♣ Total iron

TABLE XI

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES
FROM THE SAME ZONE WITHIN THE POLL MILLER PEGMATITE
(Franklin - Sylva district, N.C.)

	54	55	56	57	59	362
Fe_2O_3	2.3	2.8	2.0	2.3	2.5	2.7
MgO	.95	1.2	1.2	.90	1.2	.85
MnO	.13	.12	.03	.10	.04	.03
TiO_2	.07	.08	.21	.05	.08	.07
CaO	.002	.001	.002	.002	.002	.0007
SrO	--	--	--	--	--	--
BaO	.06	.002	.17	.005	.05	.003
Na_2O	.90	.85	.78	.90	.80	1.2
Li_2O	--	--	--	--	--	--
Rb_2O	--	--	--	--	--	--
Cs_2O	--	--	--	--	--	--
Ga_2O_3	.026	.022	.018	.024	.015	.017
Sc_2O_3	--	--	.003	--	.001	--
Co_2O_3	.0002	--	.0003	--	.0004	--
Cr_2O_3	--	--	--	--	.0001	--
SnO_2	.006	.003	.0007	.005	.001	--
V_2O_5	.007	.0001	.002	.002	.0004	--
F	--	--	--	--	--	--

*Analyses by C. E. Harvey

TABLE XII

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES
FROM THE SAME ZONE WITHIN THE TILLEY PEGMATITE
(Franklin-Sylva district, N. C.)

	18	20	21	22	23
Fe_2O_3	2.3	3.7	4.2	3.4	3.3
MgO	.78	1.1	1.3	1.4	1.15
MnO	.08	.08	.024	.06	.11
TiO_2	.09	.20	.13	.16	.13
CaO	.002	.002	.002	.004	.43
SrO	.003	--	--	.002	--
BaO	.23	.10	.03	.08	.09
Na_2O	.66	.68	1.5	.33	.65
Li_2O	--	--	--	--	--
Rb_2O	--	--	--	--	--
Cs_2O	--	--	--	--	--
Ga_2O_3	.017	.018	.016	.02	.022
Sc_2O_3	.003	.002	--	.003	.002
Co_2O_3	.0003	.0004	--	.0005	.0004
Cr_2O_3	.00	.0008	--	.0003	.0004
SnO_2	.0007	.001	.004	.002	.002
V_2O_5	--	.01	--	.006	.01
F	--	--	--	--	--

* Analyses by C. E. Harvey

TABLE XIII

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES FROM THE SAME ZONE
WITHIN THE RUBY, NORTON, SHEEP MOUNTAIN, AND DOC NICOLS PEGMATITES
(Franklin-Sylva district, N. C.)

| | Ruby | | Norton | | Sheep Mtn. | | Doc Nicols | |
	60	61	132	133	15	16	2a	2b
Fe_2O_3	2.6	2.1	4.4	4.2	2.3	2.2	3.0	3.3
MgO	.92	.97	1.4	1.25	.85	.75	.95	1.0
MnO	.017	.012	.09	.06	.018	.04	.04	.04
TiO_2	.15	.21	.13	.20	.42	.38	.07	.05
CaO	.0007	.0005	.002	.001	.001	.001	.004	.002
SrO	--	--	--	--	--	--	--	--
BaO	.04	.04	.01	.055	.27	.28	.008	.006
Na_2O	1.7	1.2	1.0	1.05	.91	.80	.83	.62
Li_2O	--	--	--	--	--	--	--	--
Rb_2O	--	--	--	--	--	--	--	--
Cs_2O	--	--	--	--	--	--	--	--
Ga_2O_3	.017	.016	.014	.014	.013	.015	.021	.022
Sc_2O_3	--	--	--	--	--	.003	--	--
Co_2O_3	--	--	.0006	.0006	--	.0002	.0003	--
Cr_2O_3	--	.0002	--	--	.0004	.0004	--	--
SnO_2	.004	.002	--	--	--	.0007	.003	.001
V_2O_5	.003	.004	--	.001	.024	.036	.009	.002
F	--	--	.20	.10	--	--	--	--

* Analyses by C. E. Harvey

TABLE XIV

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES FROM THE SAME ZONE WITHIN THE MITCHELL CREEK AND JOHNSON PEGMATITES
(Thomaston-Barnesville district, Georgia)

	Mitchell Creek			Johnson	
	150	151	154	146	147
Fe_2O_3	1.7	1.5	1.6	2.8	2.8
MgO	1.35	1.25	1.5	1.05	1.05
MnO	.03	.029	.07	.029	.033
TiO_2	.30	.29	.22	.14	.15
CaO	.002	.003	.003	.0006	.001
SrO	--	--	.002	--	--
BaO	.14	.15	.13	.004	.004
Na_2O	1.25	1.75	.80	1.2	1.25
Li_2O	--	--	--	--	--
Rb_2O	--	--	--	--	--
Cs_2O	--	--	--	--	--
Ga_2O_3	.014	.015	.02	.02	.02
Sc_2O_3	--	--	.005	--	--
Co_2O_3	--	--	.0004	--	--
Cr_2O_3	.001	.002	.004	--	--
SnO_2	--	--	.014	.002	.002
V_2O_5	.011	.018	.03	--	--
F	--	--	--	.25	.25

* Analyses by C. E. Harvey

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TABLE XV

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES FROM THE SAME ZONE WITHIN THE SCHOOL SECTION PEGMATITE
(Eight Mile Park, Colorado)

	279	281	290	295	300	305
Fe_2O_3	3.1	3.3	3.4	3.1	2.9	3.2
MgO	.91	.90	.92	.85	.75	.96
MnO	.07	.055	.05	.10	.049	.14
TiO_2	.048	.12	.23	.05	.11	.11
CaO	.002	.003	.002	.001	.002	.003
SrO	--	--	--	--	--	--
BaO	--	.0006	.001	--	.001	.002
Na_2O	.94	1.6	1.4	1.15	.74	.97
Li_2O	--	--	--	.19	--	.22
Rb_2O	--	--	--	--	--	--
Cs_2O	--	--	--	--	--	--
Ga_2O_3	.016	.018	.019	.018	.014	.017
Sc_2O_3	--	.004	.005	--	.004	--
Co_2O_3	--	--	--	--	--	--
Cr_2O_3	--	--	--	--	--	--
SnO_2	--	.004	.005	.002	--	--
V_2O_5	--	.001	.003	--	--	--
F	.62	.39	.25	.73	.27	.87

* Analyses by C. E. Harvey

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TABLE XVI

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES FROM THE SAME ZONE WITHIN THE MEYERS QUARRY PEGMATITE
(Eight Mile Park, Colorado)

	312	316	319	320	324
Fe_2O_3	2.5	3.4	2.3	2.5	3.6
MgO	.16	.90	.13	1.1	1.1
MnO	.21	.06	.12	.07	.07
TiO_2	.009	.07	.008	.014	.08
CaO	.003	.002	.002	.003	.002
SrO	--	--	--	--	--
BaO	.0007	.0008	--	.0002	.0003
Na_2O	.80	.62	.75	.88	.70
Li_2O	.52	.06	.23	.11	.13
Rb_2O	--	--	--	--	--
Cs_2O	--	--	--	--	--
Ga_2O_3	.024	.021	.025	.025	.022
Sc_2O_3	--	.002	--	--	.003
Co_2O_3	--	.0003	--	.0002	.0003
Cr_2O_3	--	--	--	--	.001
SnO_2	--	.001	--	--	.002
V_2O_5	--	--	--	--	.006
F	.53	.18	.35	.27	.17

* Analyses by C. E. Harvey

TABLE XVII

SPECTROCHEMICAL ANALYSES* OF DIFFERENT MUSCOVITES FROM THE SAME ZONE WITHIN THE GLOBE PEGMATITE (Petaca district, New Mexico)

	209	210
Fe_2O_3	4.2	3.2
MgO	.03	.01
MnO	.49	.81
TiO_2	.15	.045
CaO	.001	.002
SrO	--	--
BaO	.003	.0008
Na_2O	1.45	1.1
Li_2O	.59	.34
Rb_2O	--	--
Cs_2O	--	--
Ga_2O_3	.04	.035
Sc_2O_3	--	--
Co_2O_3	--	--
Cr_2O_3	--	--
SnO_2	.004	.005
V_2O_5	--	--
F	1.48	.98

* Analyses by C. E. Harvey

These comparisons show that, in the main, the variation in composi-tion among different books from the same zone in a single pegmatite is small. This is particularly true of those pegmatites of the eastern districts in which the mica-bearing units are continuous throughout a large part of the pegmatite. In those deposits of some of the western districts, in which the mica-bearing units are markedly discontinuous, the variation of the mica from one isolated part of a unit to that of another is considerably more pronounced.

In Tables XVIII and XIX are presented some comparisons of micas from different units or micas of different colors from the same pegmatite. In all cases the differences in color are a reflection of marked differences in chemical composition.

TABLE XVIII

SPECTROCHEMICAL ANALYSES* OF MUSCOVITES FROM THE GREGORY PEGMATITE
(Franklin-Sylva district, N. C.)

| | green footwall zone 4 | Zoned book from keel | |
		green core 5a	ruby margin 5b
Fe_2O_3	1.8	2.7	5.0
MgO	.71	.91	.33
MnO	.03	.03	.05
TiO_2	.03	.01	.014
CaO	.002	.001	.01
SrO	--	.001	.001
BaO	.30	.22	.002
Na_2O	.85	.66	.58
Li_2O	--	--	--
Rb_2O	--	--	--
Cs_2O	--	--	--
Ga_2O_3	.011	.01	.016
Sc_2O_3	--	--	--
Co_2O_3	--	--	--
Cr_2O_3	--	--	--
SnO_2	--	.003	.011
V_2O_5	--	.002	.0008
F	--	--	--

* Analyses by C. E. Harvey

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TABLE XIX

SPECTROCHEMICAL ANALYSES* OF MUSCOVITES FROM KISER PEGMATITE
(Franklin-Sylva district, N. C.)

| | wall zone ruby | | core margin green |
	44a	44b	45
Fe_2O_3	2.6	2.6	3.5
MgO	1.1	1.4	1.3
MnO	.05	.07	.12
TiO_2	.20	.06	.05
CaO	.002	.0006	.001
SrO	.003	--	--
BaO	.52	.07	.37
Na_2O	.77	.70	.75
Li_2O	--	--	--
Rb_2O	--	--	--
Cs_2O	--	--	--
Ga_2O_3	.017	.02	.018
Sc_2O_3	.004	.002	--
Co_2O_3	.0005	.0003	.0003
Cr_2O_3	--	--	--
SnO_2	.001	.001	.001
V_2O_5	.002	.0004	.0004
F	--	--	--

* Analyses by C. E. Harvey

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D. Variations in Pegmatite Districts

1. Occurrence and Color. Within a single district the pegmatites commonly contain mica concentrations in several types of units, but usually one type of deposit predominates. Throughout the southeastern states the bulk of the larger mica books occurs in wall-zone concentrations. In the Alabama district, for example, disseminated and quartz-plate types of deposits occur in over 50 percent of the pegmatites, core-margin in nearly 20 percent. The wall-zone type is found in only about 10 percent, yet over 50 percent of the total production has come from the last. In the Franklin-Sylva district about 30 percent of the pegmatites contain wall-zone deposits, a little more than 20 percent are of the disseminated type, and about 35 percent are of the core-margin type.

Commonly in a single district one color type predominates in the mica, though most districts show considerable variation. In most of the Piedmont districts of North Carolina, South Carolina, Georgia, and Alabama the dominant color is ruby. In the Ridgeway-Sandy Ridge district of Virginia and North Carolina green mica is common. In other southeastern districts there appear systematic color variations which can be correlated with the position of the pegmatite with respect to its parent batholith. These occur in the Spruce Pine, Franklin-Sylva, Ridgeway-Sandy Ridge and Hartwell districts (see Heinrich, 1953).

In the Latah County, Idaho, district ruby micas predominate. Light brown types are common in Montana. In the Eight Mile Park area of Colorado the core-margin mica is very pale ruby, whereas that of fracture-controlled units is silvery green. Most of the Petaca, New Mexico, mica is green.

2. Composition. Tables XX through XXVIII show the variation in minor and trace elements in muscovites from nine districts. Fe_2O_3 averages show considerable variation: high in Spruce Pine, Petaca and Eight Mile Park districts; intermediate in Franklin-Sylva; low in Shelby-Hickory, Thomaston-Barnesville, Alabama, and Montana, and very low in Idaho. MgO averages show much less variation, but are conspicuously low for Petaca micas and somewhat low for Idaho micas. Conversely, the Petaca district micas have high MnO contents. TiO_2 averages show less variability and are lowest in micas from Montana. CaO averages are very similar except for that of the Franklin-Sylva micas, which is relatively high. Sr is absent in four districts, and highest in Franklin-Sylva micas. BaO averages vary widely: high in Franklin-Sylva, Spruce Pine, and Alabama; low in Shelby-Hickory, Thomaston-Barnesville, Montana; and very low in Idaho, Petaca, and Colorado muscovites.

Na_2O averages show less variation; that for the Georgia district is somewhat higher than the norm. Li_2O is absent in four districts, uncommon and

low in three (Spruce Pine, Alabama, Idaho), common and moderate in Eight Mile Park and very common and highest in Petaca micas. Rb_2O and Cs_2O were not detected in muscovites from any district. Ga_2O_3 is present in all the analyzed muscovites; the averages are very uniform, except for the Petaca district which is about double those of the others. Sc_2O_3 is absent in the Idaho micas, very low in Petaca micas, relatively uniform for other districts. Co_2O_3 is absent in one district, very low in most and highest in Spruce Pine muscovites. Cr_2O_3, absent in one district, is uncommon and very low in all others. SnO_2 has a remarkable concentration in the Idaho district, is relatively high in the Petaca district, and is fairly uniform for the rest. Idaho, Petaca, and Montana districts are characterized by low V_2O_5. F is low in Franklin-Sylva, Shelby-Hickory, Georgia, and Montana; absent in Alabama; moderate in Colorado, Spruce Pine, and Idaho; and exceptionally high and invariably present in Petaca muscovites.

These variations in averages serve to show that certain elements, namely, Fe,Mn,Ba,Li,Sn, and F, show marked variation from district to district. These variations are ascribable to two causes: (1) fundamental differences in the parent magma from which the pegmatite liquids were withdrawn--variation by provinces; (2) differences in crystallization stage of the muscovites - some are magmatic, others hydrothermal--variation by differentiation.

TABLE XX *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Franklin-Sylva District, North Carolina, and north Georgia)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	1.6 - 5.1	2.77
MgO	100	.33 - 1.7	1.12
MnO	100	.009 - .17	.058
TiO_2	100	.01 - .52	.166
CaO	100	.0005 - .43	.015
SrO	33	.001 - .005	.0009
BaO	100	.0004 - .62	.193
Na_2O	100	.33 - 1.7	.826
Li_2O	0	----	---
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.010 - .026	.017
Sc_2O_3	44	.001 - .006	.0014
Co_2O_3	59	.0002 - .0008	.0002
Cr_2O_3	49	.0001 - .002	.0002
SnO_2	74	.0007 - .011	.0018
V_2O_5	83	.0001 - .036	.0066
F	8	.13 - .42	.029

* Based on spectrochemical data by C. E. Harvey

TABLE XXI *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Spruce Pine District, North Carolina)

Oxide	Present in % of Samples	Range		Average
Fe_2O_3	100	1.6	- 5.1	3.78
MgO	100	.25	- 1.9	1.03
MnO	100	.015	- .09	.066
TiO_2	100	.01	- .30	.11
CaO	100	.0005	- .004	.0013
SrO	9		.003	.0002
BaO	100	.0005	- 1.10	.215
Na_2O	100	.42	- 1.4	.917
Li_2O	9		.27	.02
Rb_2O	0		----	---
Cs_2O	0		----	---
Ga_2O_3	100	.013	- .033	.021
Sc_2O_3	36	.001	- .003	.0003
Co_2O_3	28	.003	- .005	.0007
Cr_2O_3	9		.003	.0002
SnO_2	72	.001	- .007	.002
V_2O_5	82	.001	- .012	.003
F	54	.10	- .73	.17

* Based on spectrochemical data by C. E. Harvey

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TABLE XXII *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Shelby-Hickory District, North Carolina)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	1.6 - 2.7	2.08
MgO	100	.74 - .98	.88
MnO	100	.013 - .08	.035
TiO_2	100	.069 - .19	.119
CaO	100	.0005 - .002	.0012
SrO	0	----	---
BaO	100	.005 - .09	.003
Na_2O	100	.70 - 1.05	.89
Li_2O	0	----	---
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.01 - .032	.019
Sc_2O_3	20	.006	.0012
Co_2O_3	20	.0004	.00008
Cr_2O_3	0	----	---
SnO_2	80	.002 - .006	.003
V_2O_5	40	.005	.002
F	20	.12	.024

* Based on spectrochemical data by C. E. Harvey

TABLE XXIII *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Thomaston-Barnesville District, Georgia)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	1.5 - 2.8	2.08
MgO	100	.68 - 1.35	1.08
MnO	100	.012 - .10	.037
TiO_2	100	.03 - .40	.19
CaO	100	.0006 - .003	.0016
SrO	10	.002	.0002
BaO	100	.003 - .15	.058
Na_2O	100	.80 - 1.75	1.24
Li_2O	0	----	---
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.013 - .025	.018
Sc_2O_3	20	.002 - .005	.0007
Co_2O_3	20	.0004 - .0006	.0001
Cr_2O_3	40	.0008 - .004	.0008
SnO_2	50	.002 - .014	.003
V_2O_5	50	.0007 - .03	.0065
F	40	.15 - .28	.093

* Based on spectrochemical data by C. E. Harvey

TABLE XXIV *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Alabama District)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	1.4 - 2.4	1.88
MgO	100	.88 - 1.5	1.13
MnO	100	.014 - .10	.052
TiO_2	100	.06 - .21	.11
CaO	100	.001 - .004	.002
SrO	15	.002	.0003
BaO	100	.001 - .68	.166
Na_2O	100	.38 - 1.6	.84
Li_2O	29	.07 - .26	.047
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.011 - .022	.017
Sc_2O_3	57	.003 - .005	.002
Co_2O_3	29	.0003 - .0004	.0001
Cr_2O_3	43	.001	.0001
SnO_2	71	.004 - .012	.004
V_2O_5	71	.001 - .017	.005
F	0	----	---

* Based on spectrochemical data by C. E. Harvey

TABLE XXV *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Latah County District, Idaho)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	1.05 - 1.9	1.61
MgO	100	.24 - 1.1	.64
MnO	100	.03 - .12	.07
TiO_2	100	.003 - .12	.068
CaO	100	.001 - .004	.0026
SrO	0	----	---
BaO	100	.001 - .007	.004
Na_2O	100	.65 - 1.0	.86
Li_2O	40	.04 - .05	.018
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.01 - .022	.018
Sc_2O_3	0	----	---
Co_2O_3	0	----	---
Cr_2O_3	60	.002	.0001
SnO_2	100	.024 - .10	.058
V_2O_5	60	.003 - .011	.0004
F	80	.14 - .21	.14

* Based on spectrochemical data by C. E. Harvey

TABLE XXVI *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Petaca District, New Mexico)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	3.2 - 5.6	4.24
MgO	100	.008 - .43	.119
MnO	100	.28 - .83	.47
TiO_2	100	.045 - .27	.105
CaO	100	.001 - .025	.005
SrO	0	----	---
BaO	100	.0004 - .06	.0059
Na_2O	100	.75 - 1.6	1.08
Li_2O	88	.29 - .85	.46
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.024 - .062	.041
Sc_2O_3	6	.004	.0002
Co_2O_3	12	.0004 - .0006	.0001
Cr_2O_3	6	.0002	.00001
SnO_2	100	.003 - .04	.014
V_2O_5	29	.0005 - .002	.0004
F	100	.65 - 1.48	1.00

* Based on spectrochemical data by C. E. Harvey

TABLE XXVII*

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Eight Mile Park District, Colorado)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	2.3 - 4.7	3.25
MgO	100	.13 - 1.15	.79
MnO	100	.049 - .28	.102
TiO_2	100	.008 - .31	.094
CaO	100	.001 - .005	.0023
SrO	0	----	---
BaO	69	.0002 - .007	.0011
Na_2O	100	.62 - 1.8	1.12
Li_2O	69	.06 - .52	.17
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.014 - .048	.023
Sc_2O_3	42	.002 - .005	.0016
Co_2O_3	16	.0002 - .0003	.00004
Cr_2O_3	10	.001	.0001
SnO_2	53	.001 - .056	.006
V_2O_5	37	.001 - .007	.0015
F	100	.10 - 1.15	.48

* Based on spectrochemical data by C. E. Harvey

TABLE XXVIII *

MINOR AND TRACE ELEMENTS IN MUSCOVITES FROM PEGMATITES
(Southwestern Montana)

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	1.7 - 2.2	1.97
MgO	100	.53 - 1.4	.84
MnO	100	.01 - .06	.028
TiO_2	100	.01 - .12	.057
CaO	100	.001 - .004	.003
SrO	17	.003	.0005
BaO	100	.002 - .19	.036
Na_2O	100	.87 - 1.15	1.00
Li_2O	0	----	---
Rb_2O	0	----	---
Cs_2O	0	----	---
Ga_2O_3	100	.011 - .033	.019
Sc_2O_3	17	.003	.0005
Co_2O_3	50	.0003 - .0005	.0002
Cr_2O_3	17	.001	.0002
SnO_2	100	.003 - .012	.006
V_2O_5	83	.0006 - .009	.0004
F	17	.10	.017

* Based on spectrochemical data by C. E. Harvey

E. Commercial Quality Factors

Factors that affect the utility and economic value of sheet muscovite are:

(1) size of books,

(2) structural defects, and

(3) inclusions.

The book size varies with the zone, deposit, and district and even greatly within a single zone. The relative abundance of structural defects such as A, herringbone, other reeve structures and wedge structure is related markedly to the type of occurrence of the muscovite, and governs the maximum size of flat, perfect sheets that may be cut from the books. The distribution of rulings and cracks also is, to a lesser extent, related to the zone type in which the muscovite occurs. These defects also limit the size of sheets obtainable from larger books. Pinholes, most of which result from the liberation of small included grains of crystals of various minerals during splitting, are more abundant in ruby or brown micas of wall zones. Haircracks are likewise concentrated in this type. Both defects result in conductance through the mica sheet or film with attendant rupture or puncture, if the hole or crack does not extend entirely through the sheet.

Inclusions of hematite and magnetite ("staining") are more common in green than in ruby micas. Heavily stained mica, generally classed by visual inspection as electrical stock (No. 3), may contain a high proportion of grade E-1 (Jahns and Lancaster 1950, p. 46). These investigators state (p. 46), "There appears to be a partial correlation between power factor and the amount of hematite-magnetite stain in the mica, in that most of the E-2 and E-3 pieces are heavily stained but evidently there are numerous exceptions. Some pieces that qualified as E-1, for example, were more densely stained than many with a higher power factor."

Likewise there is no general correlation between power factor and color. Despite the antipathy of industry toward green mica, correctly prepared sheets and films of green muscovite are similar in quality to ruby muscovite. Possibly the aversion to green mica lies to some extent in the fact that, owing to a higher proportion of structural defects, large flat sheets are more difficult to prepare from it.

PART III. LEPIDOLITE

I. MINERALOGY

A. Composition

The generalized formula for lepidolite may be written as:

$$K_2(Li,Al)_{5-6}(Si_{6-7}Al_{2-1})O_{20-21}(F,OH)_{3-4}.$$
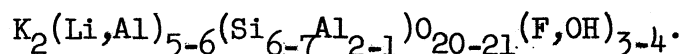
In addition to the substitutions indicated by the formula, the following appreciable isomorphism has been noted:

for K -- Na,Rb,Cs;

in octahedral positions -- Fe^2,Fe^3,Mn,Mg.

The following elements have been reported and when present are usually in extremely small amounts: Ca,Sn,Ga,Cb,Ti,Ba,Sr; Rb and Cs in some types are found in considerable amounts (Rb_2O = 3.2 percent and Cs_2O = 1.90 percent, Lundblad, 1942); MnO in amounts as high as 6.0 percent reported in new spectrochemical analyses prepared for this report. The OH:F ratio varies considerably and OH may become negligible.

Many unique theories have been advanced to explain the chemical variation in this species. The two most recent and widely accepted at present are those of Stevens (1938) and Winchell (1942). Both employ the end-member concept whose fundamental units are:

Stevens (1938)

$KLi_2AlSi_4O_{10}F_2$ polylithionite

$K(Mn,Fe,Mg)_3AlSi_3O_{10}(OH,F)_2$ biotite

$$K_4Li_6Al_6Al_4Si_{12}O_{40}(OH,F)_8 \quad \text{lithium muscovite}$$

$$KAl_2(AlSi_3)O_{10}(OH,F)_2 \quad \text{muscovite}$$

Winchell (1942)

polylithionite $K_2Li_4Al_2Si_8O_{20}F_4$

protolithionite $K_2LiFe_4Al_3Si_6O_{20}F_4$

paucilithionite $K_2Li_3Al_5Si_6O_{20}F_4$
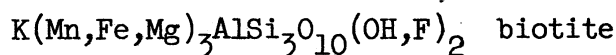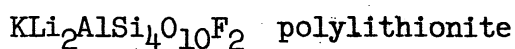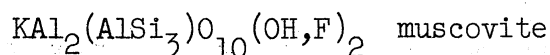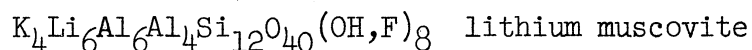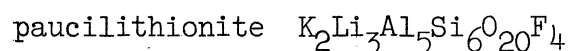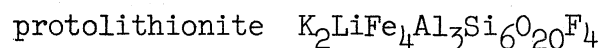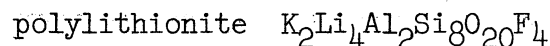
Winchell (1942, p.121) points out that an additional end-member, **muscovite**, is needed to explain the lithia micas. However, he accepts only the above three as end-members of the lepidolite system and considers that the "muscovite molecule enters into crystal solution in this system only very little, if at all." He further notes that almost all lepidolites are deficient in Li_2O by approximately 1 percent and considers this due to interlayered, but not isomorphously combined, muscovite.

The end-members of Stevens (1938) and Winchell (1942) are useful today, from our point of view, inasmuch as they indicate the wide possible chemical variation occurring in lepidolites.

The work of Stevens (1938) and Winchell (1942) is typical of those who favor the end-member concept. It has been pointed out by Heinrich (1946), Rabbitt (1948), and others that the theoretical end-molecules have no significance in the structure of the micas and other minerals and that variation in chemical compositions of various series cannot be adequately expressed by them. Collins (1942) further notes that the suitability of plotting optical properties against these hypothetical compounds is doubtful.

For this study 26 lepidolites were analyzed spectrochemically. The results are shown in the **Table XXIX**.

B. Structure

1. Polymorphism. Hendricks and Jefferson (1939) found three different polymorphic variations in lepidolites:

1-layer monoclinic:

$$a_o = 5.3\text{Å} \quad b_o = 9.2\text{Å} \quad c_o = 10.2\text{Å} \quad \beta = 100°$$

Space Group: Cm

TABLE XXIX *

NEW SPECTROCHEMICAL DATA ON 26 LEPIDOLITES

Oxide	Present in % of Samples	Range	Average
Fe_2O_3	100	.03-3.2	.48
MgO	100	.007-.27	.043
MnO	100	.014-6.0	1.16
TiO_2	35	.005-.12	.014
CaO	100	.001-.05	.007
SrO	69	.002-.02	.006
BaO	65	.0002-.001	..0003
Na_2O	100	.14-.72	.41
Li_2O	100	1.3-4.4	3.77
Rb_2O	100	.12-1.0	.54
Cs_2O	62	.04-.37	.08
Ga_2O_3	100	.003-.046	.017
Sc_2O_3	8	.002-.013	.0006
Co_2O_3	0	-	-
Cr_2O_3	4	.0001	..000004
SnO_2	100	.002-.16	.021
V_2O_5	8	.0005-.002	.0001
F	100	1.9-6.8	5.41

*Based on spectrochemical analyses of C. E. Harvey

6-layer monoclinic:

$$a_o = 9.2\text{Å} \quad b_o = 5.3\text{Å} \quad c_o = 60.0\text{Å} \quad \beta = 90°$$

Space Group: Cc

This polymorph is unique in that the values of a_o and b_o are interchanged as compared with all other mica polymorphs.

3-layer hexagonal:

$$a_o = 5.3\text{Å} \quad c_o = 30.0\text{Å}$$

Space Group: $C3_1 12$ or $C3_2 12$

Approximately 300 samples (50 analyzed chemically) of lepidolite from about 100 different specimens have been X-rayed and their symmetry has been determined. All the lepidolite polymorphs described by Hendricks and Jefferson (1939) have been recorded, and in general the results agree with theirs. One anomalous type discovered is a lepidolite whose structure is that of the normal 1-layer monoclinic polymorph but whose optic plane is normal to the side pinacoid instead of parallel to it. Unfortunately, owing to its small size, the crystal was lost during manipulation of the goniometer head upon which it was mounted. Also a new lepidolite polymorph has been found (3-layer monoclinic) which is described at the end of this section.

Much of the structural data on lepidolites has been published in a paper, "Studies in the Mica Group; Relationship between Polymorphism and Composition in the Muscovite-Lepidolite Series," by Levinson (1953), which resulted from the natural-mica research supported by the Signal Corps contract. The information on the following pages represents an elaboration and extension of this information.

2. Single-Crystal Variations. Most of the specimens analyzed by Stevens (1938), which were structurally investigated by Hendricks and Jefferson (1939, p. 761), were restudied. Table XXX illustrates the high degree of correlation between Hendricks and Jefferson (1939) and the writers' data. The two sets of results are in general agreement. However, some very significant variations may be observed. In Stevens (1938) No. 3, two very small pieces of lepidolite were found from which Weissenberg patterns could be obtained. Both of these gave identical 6-layer monoclinic diffraction patterns. Inasmuch as an optical orientation method is mainly used, the mass of the sample could not be studied by the Weissenberg method because most of the crystals had wavy extinction and gave distorted interference figures. For Stevens No. 6, Hendricks and Jefferson (1939) report the 6-layer monoclinic structure.

Two Weissenberg photographs of this sample indicated respectively the presence both of the 6-layer lepidolite polymorph and the 2-layer lithian muscovite type. discussed previously. Therefore, the chemical analysis presented by Stevens (1938) is a composite analysis of the two forms.

TABLE XXX

STRUCTURE OF MICAS ANALYZED BY STEVENS (1938)

Stevens (1938) No.	Li_2O	Total No. of Atoms with Six-fold Coordination	Hendricks and Jefferson (1939)	This Report
1	2.70	2.48	Muscovite	Normal muscovite
2	3.51	2.54		Too fine grained
3	3.70	2.60	Too fine grained	6-layer monoclinic
4	3.81	2.58	for study	Too fine grained
5	3.96	2.63		Too fine grained
6	5.04	2.73	6-layer monoclinic	6-layer monoclinic and lithian muscovite
7	5.05	2.86	6-layer monoclinic	Not available
8	5.11	2.71	Single layer	Single layer
9	5.33	2.81	Single layer	Single layer
10	5.39	2.94	Single layer	Not available
11	5.51	2.85	None available	Not available
12	5.64	2.85	6-layer monoclinic	Not available
13	5.78	2.91	Single layer	Single layer
14	5.89	2.94	3-layer hexagonal	3-layer hexagonal
15	6.18	2.95	Single layer	Single layer
16	6.84	2.98	Single layer	Single layer
17	7.26	2.97	Single layer	Single layer

A systematic study was made of one book of lepidolite for the purpose of determining the extent of polymorphic variation within a single crystal. Specimen No. 514 from the Opportunity pegmatite, in Gunnison County, Colorado was chosen. The specimen is approximately 1-1/2 inches across and 3/4 inch thick. It has a uniform, typical pink color and is embedded in a mass of cleavelandite with grains of microlite. The sheets are a composite of many crystals. Between many of these crystal units are fine-grained aggregates of lepidolite with irregular form and anomalous extinction. The optic

planes of the various crystal units in a single sheet are always at 30°, or
some multiple thereof, to each other. This appears to be the case regardless
of the shape of the crystals in question, and whether or not they are separated
by the fine-grained aggregate. 2V on all sections of the sheet, regardless of
polymorph, remains almost invariant. In all, 27 Weissenberg photographs of
small crystals from two sheets, one near the top and the other in the center
of the book were made (see Table XXXI). Of the 14 photographs taken from the
sheet near the top, 12 show the 6-layer monoclinic structure and 2 the 1-layer
monoclinic structure. However, on 7 of the photographs of the 6-layer mono-
clinic form, the pattern of lithian muscovite was superimposed, in some cases
only faintly present. Of the 13 photographs obtained from crystals in the cen-
tral sheet, 12 had the 6-layer monoclinic structure and one had the 1-layer
monoclinic pattern; only one lithian muscovite pattern was recorded and this again
was superimposed faintly on a 6-layer monoclinic pattern. This shows the wide
polymorphic variation possible in lepidolite both within sheets and also paral-
lel to the c-axis direction. Yet one polymorph, in this case the 6-layer type,
is dominant throughout the book. An inspection of Table XXX indicates that ex-
cept for numbers 12 and 14, those lepidolites with more than 5.1 percent Li_2O
have crystallized with the 1-layer structure. It is necessary therefore to
explain the apparently erratic results obtained in the case for No. 12, which
has the 6-layer structure and No. 14 with the 3-layer hexagonal structure.
In the case of No. 12, unfortunately, no material was available for restudy.
It seems possible however, that further investigation could reveal 1-layer
forms present in addition to the 6-layer type found by Hendricks. A re-
determination of the Li_2O content also would be desirable.

3. Uniaxial Lepidolite. Uniaxial lepidolite (No. 14) which has cry-
stallized with the 3-layer hexagonal structure has a composition identical with
that of lepidolites that have crystallized as the 1-layer polymorph. Such uni-
axial lepidolites or nearly uniaxial lepidolites are very rare. A review of
the literature revealed that Baumhauer (1903, 1912) first described such
material from Mursinsk. He proposed the term microlepidolite for lepidolites
of this character in contrast to macrolepidolites with large 2V. No chemical
analyses were reported. Unixial lepidolite (5.83-6.15 percent Li_2O) from
Western Australia has been described by Winchell (1925), Simpson (1927),
Murray and Chapman (1931), Stevens (1938) and Hendricks and Jefferson (1939).
This uniaxial lepidolite is associated with a normal biaxial mica of almost
identical composition. Winchell (1925, p. 424), in agreement with Simpson
(1927), suggests that the uniaxial character "may be due to fine twinning
on (001)." Hendricks and Jefferson (1939) reported the structure of uniaxial
material from Londonderry as the 3-layer hexagonal type, whereas the biaxial
material was the 1-layer polymorph.

Jakob (1927) analyzed uniaxial lepidolite (4.93 percent Li_2O) from
Usakos, and Pehrman (1945) analyzed and described similar material from Kimito
(4.99 percent Li_2O). Specimens of both of these analyzed micas were found to
have small areas of a biaxial phase. X-ray studies show that the uniaxial

TABLE XXXI

EXTENT OF POLYMORPHIC VARIATION IN ONE LEPIDOLITE BOOK
FROM OPPORTUNITY PEGMATITE, GUNNISON COUNTY, COLORADO

Sheet near top of book

514 (a)___ 6-layer monoclinic
    (b)___ 6-layer monoclinic + 2-layer lithian muscovite (faint)
    (c)   6-layer monoclinic + 2-layer lithian muscovite
    (d)   6-layer monoclinic + 2-layer lithian muscovite
    (e)___ 6-layer monoclinic + 2-layer lithian muscovite (faint)
    (f)   1-layer monoclinic
    (g)___ 6-layer monoclinic + 2-layer lithian muscovite
    (h)___ 6-layer monoclinic + 2-layer lithian muscovite (very faint)
    (i)___ 6-layer monoclinic
    (j)___ 6-layer monoclinic
    (k)   1-layer monoclinic
    (l)   6-layer monoclinic + 2-layer lithian muscovite
    (m)   6-layer monoclinic

Sheet near center of book

514 (1)___ 6-layer monoclinic
    (2)   6-layer monoclinic
    (3)   6-layer monoclinic
    (4)   6-layer monoclinic
    (5)   6-layer monoclinic
    (6)___ 6-layer monoclinic
    (7)___ 6-layer monoclinic
    (8)___ 6-layer monoclinic + 2-layer lithian muscovite (faint)
    (9)   6-layer monoclinic
 (10)___ 6-layer monoclinic
 (11)   6-layer monoclinic
 (12)   6-layer monoclinic
 (13)   1-layer monoclinic

Diffuse scattering observed in:

514 (a)   514 (1)
    (b)   (6)
    (e)   (7)
    (g)   (8)
    (h)   (10)
    (i)
    (j)

portions have crystallized as the 3-layer hexagonal polymorph but that the bi-axial parts have the lithian muscovite structure. Since it has been demonstrated that the lithian muscovite structure is confined to a lower Li_2O range, the Li_2O contents of the Usakos and Kimito micas represent composite results which are lower than those obtainable from strictly uniaxial specimens.

Lundblad (1942, p. 58) notes that an analyzed specimen of lepidolite from Varuträsk has both uniaxial and biaxial portions. The analysis of this material seems unreliable; 65.62 percent SiO_2 in a lepidolite is too high. Miss Berggren (the analyst), according to Lundblad (1942), seems to suspect quartz contamination. Under these conditions it seems best to eliminate this mica from consideration.

Recently Macgregor (1945) described a lepidolite from Southern Rhodesia with a small 2V. A specimen similar to the described material surrounds a core of muscovite and in turn is enclosed by lepidolite with a large 2V. The muscovite structure is normal, and the lepidolite with the large 2V has crystallized as the 1-layer polymorph, whereas the uniaxial portion has crystallized as the 3-layer hexagonal form. The association of the 1-layer and 3-layer hexagonal polymorphs is identical with that of the Western Australia material. Chemical analyses of these lepidolites are not available.

If one takes into account the presence of lithian muscovite in the Usakos and Kimito lepidolites, it seems reasonable to infer that the Li_2O content of lepidolite of entirely uniaxial character may approach or even exceed the lowest Li_2O content of any of the samples with the 1-layer structure.

Structurally the 3-layer hexagonal polymorph can be geometrically interpreted as three 1-layer forms "twinned" on (001) with succeeding layers rotated 120° in the same direction (either clockwise or counterclockwise). Hendricks and Jefferson (1939, p. 746) with relation to the 3-layer hexagonal structure state that, "a particular layer is repeated about a three-fold screw axis normal to the cleavage." The powder patterns (Fig. 7) of the 3-layer hexagonal polymorph and the 1-layer monoclinic polymorph are almost identical. The cause of this rotation remains to be explained.

Reusch (1869) first obtained an almost perfectly uniaxial figure in muscovite by stacking rotated sheets in this fashion. Although his sheets were of considerable thickness, the over-all optical effect appears to be the same.

Ford (1932) explains the uniaxial effect on the basis of rotation of the plane of polarization. He notes (p. 328):

"A particularly interesting case,.. is that of the special
properties of superposed cleavage-sections of mica. If three or
more of these, say of rectangular form, be superposed and so
placed that the lines of the axial planes make equal angles of
60° (45°, etc.) with each other the effect is that polarized
light which has passed through the center suffers circular polar-
ization, with a rotation to the right or left according to the
way in which the sections are built up. The interference-figure
resembles that of a section of quartz cut normal to the axis...
Further, it is easy from this to understand how it is possible to
have in sections of certain crystals (e.g., of clinochlore) por-
tions which are biaxial and others that are uniaxial, the latter
being due to an intimate twinning after this method of biaxial
portions."

Similar effects have been reported in feldspars (Sugi, 1940), talc
and serpentine.

4. Powder X-ray Studies. Hendricks and Jefferson (1939) note that
Stevens (1938) Nos. 2,3,4, and 5, which appear to be in the interval between
the 2-layer muscovite type and the 6-layer lepidolite type, are too fine-
grained for study and speculate (p. 763):

"It is tantalizing to think that samples 2 to 5 owe their poor
crystal development to their close approach to the limit of the lepi-
dolite solid solution in muscovite."

It seems very likely that these fine-grained micas owe their macrostructural
defects to small-scale variations in their crystal structure which itself
probably is assignable to their chemically transitional position in the
muscovite-lepidolite series. In order to test this possibility, powder
X-ray studies on these micas were attempted.

It was first necessary to obtain powder X-ray data for each of the
known polymorphs. The only powder data on muscovites or lepidolites found
in the literature were for normal muscovite (Nagelschmidt, 1937) and for the
3-layer muscovite polymorph (Axelrod and Grimaldi, 1949). Grim and Bradley
(1951) list partial data for normal muscovite and a 1-layer lepidolite and
give photographs of the two patterns. A complete set of powder pictures was
compiled by X-raying powdered single crystals whose structure had first been
determined by the Weissenberg method. This set of standard photos, which
includes the polymorphs of muscovite and lepidolite under study, is shown
in Fig. 7; the d-spacings of forms not previously published are given in
Table XXXII.

The structures of the fine-grained lepidolites analyzed by Stevens
(1938) have been determined:

Stevens No.	Per Cent Li_2O	Structure by Hendricks and Jefferson	This Report		Total Six-Fold Coord.
			Weissenberg	Powder	
2	3.51	⌐	too fine-grained	6-layer lepidolite + 2-layer muscovite	2.54
3	3.70	{ too fine-grained	6-layer	6-layer lepidolite + 2-layer muscovite	2.60
4	3.81		too fine-grained	6-layer lepidolite + 2-layer muscovite	2.58
5	3.96	⌊	too fine-grained	6-layer lepidolite + (2-layer muscovite?)	2.63

That these fine-grained lepidolites are combinations of forms and not single structures confirms the idea that the poor crystal development is related to their composition. Micas with these combined structures may be termed transitional. It is noteworthy that crystals of Stevens No. 3 large enough for Weissenberg photographs are 6-layer forms, but the poorly developed crystals from the same specimen have a combination 6-layer lepidolite and 2-layer muscovite structure. It is likely that the well developed crystals have a higher lithium content than the intergrown portions. In all cases the 6-layer lepidolite polymorph predominates over the muscovite type in the transitional types.

Owing to the close similarity between the powder patterns of normal muscovite and lithian muscovite, the exact type of muscovite in the combined powder patterns of the transitional micas is difficult to determine with certainty. However, careful study of the intensities indicates the probable presence of the lithian muscovite type.

5. Variation of Structure with Composition. On the basis of the structural data obtained it seemed advantageous to plot Li_2O content against the polymorphic types of the muscovite-lepidolite series: normal muscovite, lithian muscovite, transitional micas, 6-layer lepidolite, 1-layer lepidolite and 3-layer hexagonal lepidolite. The result of plotting the Li_2O contents of 35 analyzed micas against their structures is presented in Fig. 8. Many more micas with less than 2 percent Li_2O were studied, but there is no need to record these, as they all have the normal muscovite structure.
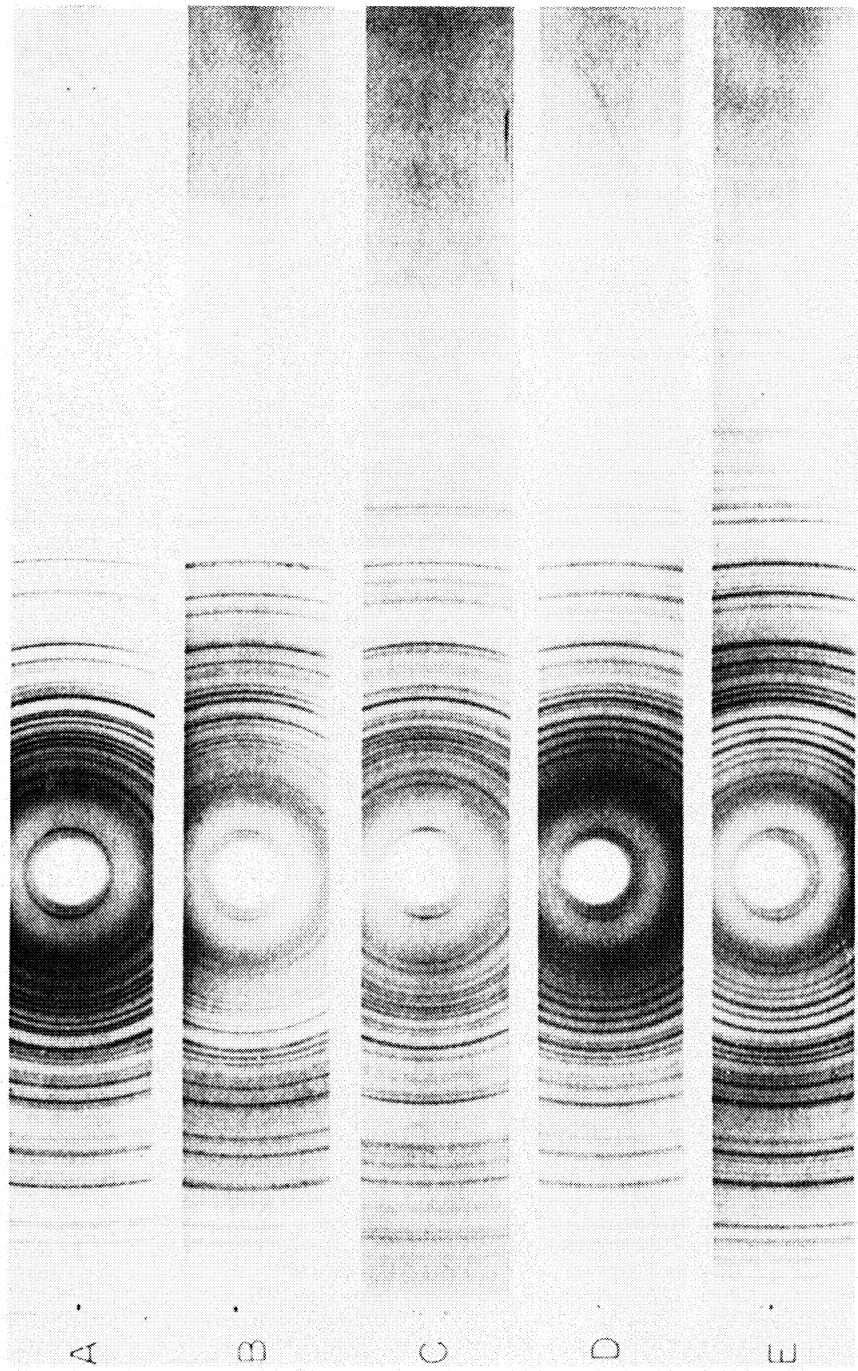
Fig. 7. X-ray powder photographs of polymorphs in muscovite-lepidolite series. A, normal muscovite, Varuträsk, Sweden; B, lithian muscovite, South Portland, Maine; C, 6-layer lepidolite, Gunnison County, Colorado; D, 1-layer lepidolite, Skuleboda, Sweden; E, 3-layer hexagonal lepidolite, Londonderry, Western Australia. (For another photograph, see Levinson, 1953, p. 98.)

TABLE XXXII

SPACINGS OF POLYMORPHIC FORMS

Cu Kα_1, λ = 1.53736 kX

2-layer lithian muscovite (monoclinic) South Portland, Maine No. 471		1-layer lepidolite (monoclinic) Topsham, Maine No. 467		3-layer lepidolite (hexagonal) Londonderry, Western Australia No. 978		6-layer lepidolite (monoclinic) Gunnison County, Colorado No. 514	
I	d-spacing	I	d-spacing	I	d-spacing	I	d-spacing
m	9.95	s	9.91	m	9.91	ms	9.89
m	5.01	s	4.98	m	4.97	m	4.99
m	4.45	w	4.94	m	4.66	m	4.49
vvw	3.95	vw	4.34	m	3.84	w	3.84
vvw	3.87	vw	4.12	m	3.58	m	3.61
w	3.71	vw	3.86	vs	3.31	m	3.47
w	3.46	s	3.61	s	3.09	m	3.31
ms	3.32	vs	3.33	s	2.86	m	3.19
m	3.21	s	3.07	w	2.651	m	3.07
m	2.98	s	2.87	s	2.571	m	2.88
s	2.84	m	2.675	mw	2.455	m	2.775
w	2.755	s	2.573	mw	2.375	vs	2.572
ms	2.571	m	2.468	vw	2.243	m	2.416
m	2.474	m	2.387	vw	2.186	vvw	2.248
m	2.387	vw	2.253	mw	2.126	vvw	2.190
vw	2.247	m	2.132	vw	2.052	vw	2.039
vvw	2.196	ms	1.988	m	1.984	s	1.985
mw	2.132	vvw	1.956	vvw	1.955	vvw	1.684
m	2.081	w	1.748	w	1.718	vvw	1.633
vw	1.951	vw	1.715	mw	1.643	vvw	1.572
w	1.742	m	1.646	w	1.611	m	1.506
vw	1.720	vvw	1.581	w	1.576	vvw	1.393
m	1.644	vvw	1.544	w	1.547	vvw	1.355
vvw	1.596	m	1.511	mw	1.511	vvw	1.319
vvw	1.557	vw	1.493	vvw	1.481	w	1.300
vvw	1.510	vvw	1.420	vvw	1.457	vvw	1.239
w	1.500	vvw	1.375	vvw	1.435		
w	1.487	vvw	1.352	vvw	1.411		
vvw	1.453	vw	1.337	w	1.341		
vw	1.427	vw	1.299	vw	1.295		
w	1.340	vvw	1.242	vvw	1.285		
w	1.296	vvw	1.199				
vvw	1.269	vvw	1.136				
vw	1.243						
vvw	1.220						
vvw	1.199						

The graph shows that micas with less than ca. 3.3 percent Li_2O have the normal muscovite structure; those with 3.4 to 4.0 percent Li_2O have transitional structures; those with 4.0 to 5.1 percent generally have crystallized with the 6-layer lepidolite structure; whereas those biaxial micas with more than 5.1 percent Li_2O generally have the 1-layer lepidolite structure. The uniaxial lepidolites as discussed previously have relatively large Li_2O contents, probably in the same range with those of the 1-layer form, and this structure may be explained by "twinning." Too few data are now available to place the lithian muscovite type of structure accurately in the series. However, the available evidence shows that it belongs somewhere between normal muscovite and 6-layer lepidolite. The above conclusions are graphically represented in Fig. 9.

Analyzed micas described by Berggren (1940, 1941) and Lundblad (1942) do not fit well into the conclusions represented by Fig. 9. The results obtained from X-ray studies of these micas are presented in Table XXXIII. Inconsistencies appear with regard to Berggren's (1940) micas in A,B,C, and Lundblad's (1942) No. 10 and No. 13. Micas C and No. 13, which are reported to have 3.9 and 5.7 percent Li_2O, respectively, have crystallized with the normal muscovite structure. As much as 5.7 percent Li_2O, or even 3.9 percent Li_2O, in normal muscovite appears improbable on the basis of present knowledge. It is possible that the lithium content of the mica samples varies so much that the portions supplied represent extreme structural variations of the analyzed materials. Several of the Swedish micas are optically nonhomogeneous, which may indicate the presence of several crystal phases, and one may even be contaminated with quartz (Lundblad, 1942). Under such circumstances an attempt to correlate polymorphism with chemistry would be fruitless. Baumhauer (1912) described several lepidolites with a wide range of 2E and several distinctive types of etch figure in single specimens. Both of these features probably indicate structural variations. Except for the Swedish micas the correlation is good; indeed, better than might be expected, considering the presence of structural variation in single books and the probable wide range in quality of the Li_2O determinations.

6. Discussion. There are at least two possible ways in which a theoretical series between muscovite and lepidolite can be constructed. One substitution consists of $2Li^{(6)} Si^{(4)} \rightleftharpoons Al^{(6)} Al^{(4)}$; the other is $Li^{(6)}Al^{(4)} \rightleftharpoons Si^{(4)}$ (the superscript numbers refer to coordination number). The former process leads to Winchell's (1942) polylithionite, whereas the latter leads to paucilithionite. Minor complications arise with the substitution of Fe^{+2}, Mn^{+2}, and Mg for Al^{+3}, but it is possible to determine without difficulty which of the above processes is involved, by inspection of the SiO_2 content. The silica content will be higher than in normal muscovite in the first case and lower in the second. It is possible that the two substitutions operate together in natural micas, but there appears to be preponderant reason for believing that the sequence muscovite-polylithionite dominates, for all true lepidolites that we have been able to study are higher in SiO_2 than muscovite. In the theoretical series muscovite-polylithionite $[2Li^{(6)}Si^{(4)} \rightleftharpoons$

1. Wodgina, Western Australia; No. 36, Rowledge (1945).
2. Tabba Tabba, Western Australia; No. 52, Rowledge (1945).
3. Varuträsk, Sweden; Analysis D, Berggren (1940).
4. Wodgina, Western Australia; No. 6, Rowledge (1945).
5. Tabba Tabba, Western Australia; No. 6, Rowledge (1945).
6. Karibib, South Africa, 2.5% Li_2O, courtesy of Foote Mineral Company.
7. Wodgina, Western Australia; No. 2, Rowledge (1945).
8. Manitoba, Canada; No. 1, Stevens (1938).
9. Karibib, South Africa; 3.0% Li_2O, courtesy of Foote Mineral Company.
10. Wodgina, Western Australia; Murray and Chapman (1931).
11. Tabba, Western Australia; Murray and Chapman (1931).
12. Ravensthorpe, Western Australia; Murray and Chapman (1931).
13. Bear Claim, Manitoba; Spec. XXXIII, Ellsworth (1932).
14. Pala, Calif.; No. 2, Stevens (1938).
15. Pala, Calif.; No. 3, Stevens (1938).
16. San Diego Co., Calif.; No. 4, Stevens (1938).
17. Pala, Calif.; No. 5, Stevens (1938).
18. Ubini, Western Australia; Murray and Chapman (1931).
19. Poona, Western Australia; Murray and Chapman (1931).
20. Pala, Calif.; No. 6, Stevens (1938).
21. Ohio City, Colorado; No. 7, Stevens (1938).
22. Pala, Calif.; No. 12, Stevens (1938).
23. San Diego Co., Calif.; No. 8, Stevens (1938).
24. Mesa Grande, Calif.; No. 9, Stevens (1938).
25. Antsongombato, Madagascar; Mauguin (1928B).
26. Wakefield, Canada; No. 10, Stevens (1938).
27. Mesa Grande, Calif.; No. 13, Stevens (1938).
28. Maharitra, Madagascar; Mauguin (1928B).
29. San Diego Co., Calif.; No. 15, Stevens (1938).
30. Antsongombato, Madagascar; No. 16, Stevens (1938).
31. Greenland; No. 17, Stevens (1938).
32. Usakos, Southwest Africa; Jakob (1927).
33. Kimito, Finland; Pehrman (1945).
34. Calgoorie, Western Australia; No. 14, Stevens (1938).
35. Londonderry, Western Australia; Murray and Chapman (1931)

The structures of all the above micas, with the exception of numbers 21, 22 and 26, have been determined by the authors. In the case of the three exceptions the structures reported by Hendricks and Jefferson (1939) have been used.
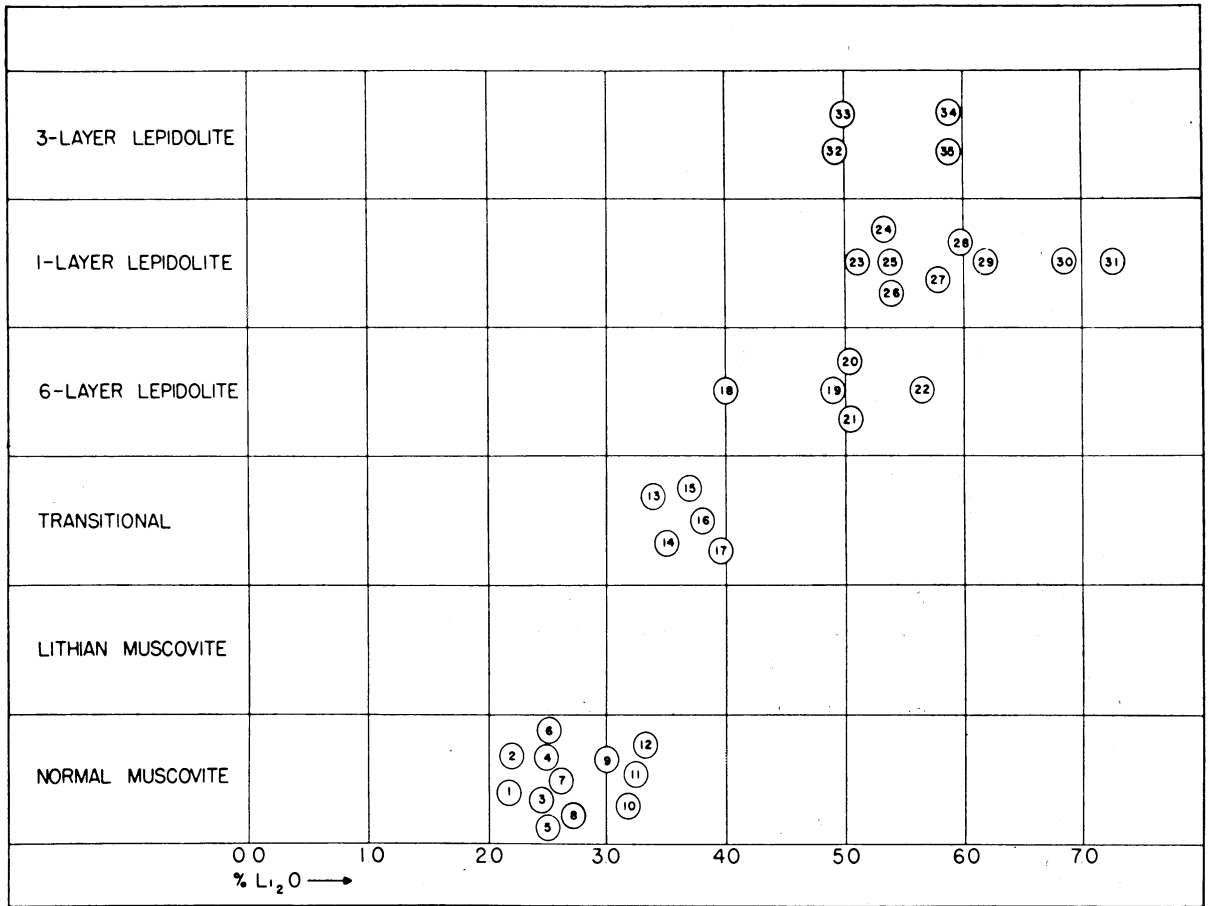
Fig. 8. Plot of the Li_2O contents of 35 micas in muscovite-lepidolite
series against the various polymorphs.
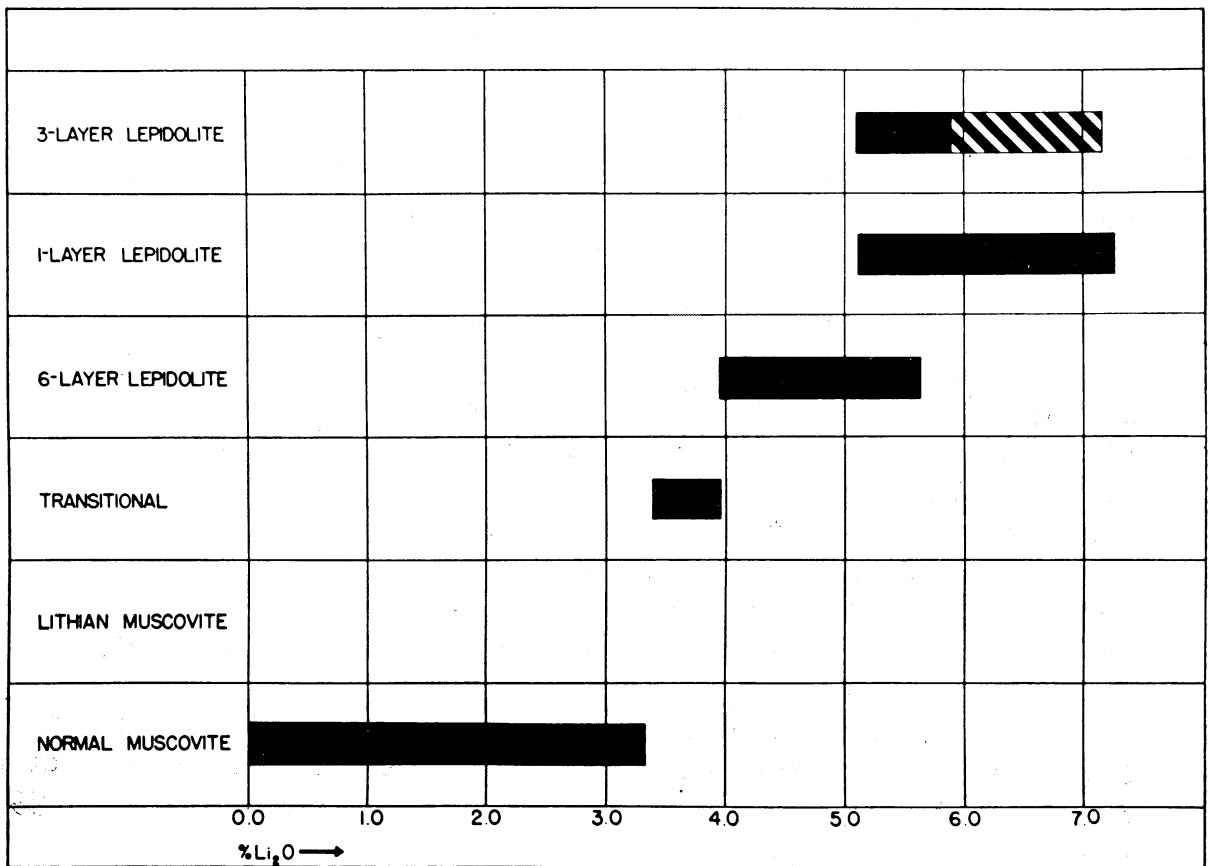
Fig. 9. Idealized representation of relationship between Li_2O content
and polymorphism in the muscovite-lepidolite series.

$Al^{(6)}Al^{(4)}$] which appears to be represented in nature, an increase in the Si content is accompanied by an increase of Li. In order for the overall charges to be balanced, two Li ions are needed in six-fold positions for each replaced aluminum. It is concluded, therefore, that inasmuch as two Li ions cannot occupy the space of one Al ion, the additional Li ion must occupy one of the vacant octahedral positions in the muscovite structure. The higher the Li content (i.e., the closer the approach to polylithionite), the more vacant positions are occupied; thus there exists a structural (though discontinuous) as well as a chemical series between heptaphyllite muscovite and octophyllite lepidolite.*

TABLE XXXIII

STRUCTURE OF MICAS DESCRIBED BY BERGGREN (1940, 1941)
AND LUNDBLAD (1942)

Analysis	% Li_2O	Structure	Structure
A	5.95	6-layer lepidolite	Varuträsk
B	4.35	1-layer lepidolite	Varuträsk
C	3.9	normal muscovite	Varuträsk
D	2.45	normal muscovite	Varuträsk
E	1.80	normal muscovite	Varuträsk
G	0.73	normal muscovite	Varuträsk
H	0.69	normal muscovite	Varuträsk
I	0.22	normal muscovite	Varuträsk
J	0.76	normal muscovite	Varuträsk
K	1.10	normal muscovite	Varuträsk
L	1.1	normal muscovite	Varuträsk
No. 10	4.55	1-layer lepidolite	Varuträsk
No. 13	5.7	normal muscovite	Utö
No. 14	5.5	6-layer and 1-layer lepidolite	Rozena

Further evidence for the existence of the muscovite-lepidolite series is found in:

*Actually the term lepidolite is awkward because it embraces 3 different polymorphs. The intended meaning of lepidolite in the phrase "muscovite-lepidolite series" is that of the ideal Li-rich mica with the composition of polylithionite ($KLi_2AlSi_4O_{10}F_2$) whose octahedral positions are completely filled.

1. The interpretation of the structure of lithian muscovite as a less distorted normal muscovite structure owing to a filling of the structural holes by Li ions (see discussion of lithian muscovite).

2. The observation that the Li_2O content of micas (Fig. 8) in the muscovite-lepidolite series ranges from 0 to 7.26 percent and that therefore the above mentioned accompanying substitutions $[2Li^{(6)}Si^{(4)} \rightleftarrows Al^{(6)}Al^{(4)}]$ probably take place.

3. The interpretation of Winchell's (1942) observation (that most lepidolites are deficient in Li_2O) as indicating that these micas do not have complete occupation of the vacant octahedral positions by Li ions; no muscovite has been found (by X-ray methods) interlayered with lepidolite except some lithian muscovite.

4. Stevens' (1938, p. 618) and Hendricks and Jeffersons' (1939, p. 761) calculations showing that the number of atoms with octahedral coordination in the micas analyzed by Stevens (1938) range from 2.48 to 2.98 (see Table XXX).

In our discussions of biotite and phlogopite it will be pointed out that no such similar chemical or structural series exists between heptaphyllite muscovite and either octophyllite biotite or phlogopite, as has been suggested by various investigators.

It has been suggested that the evidence presented above also may indicate that the extent of solid solution in the muscovite-lepidolite series is very limited. The transitional micas that appear near the middle of the series might be composed of two phases in such proportions that their average composition contains about 2.55 atoms with octahedral coordination. This concept is difficult to accept owing to the fact that several micas close to the middle of the series, such as Stevens (1938) No. 1 which contains 2.48 atoms in octahedral coordination, contain, from X-ray and optical studies, only one uniform phase.

7. Summary. The discovery of the lithian muscovite type of structure, the interpretation of the structural origin of the 3-layer hexagonal lepidolite polymorph and good correlation between Li_2O content and the various polymorphs shows that chemical and structural transitions occur between Li-free muscovites and Li-rich lepidolites. It seems probable, however, that the relationship between a single-ion variation and polymorphism in a structure as complex as the micas is somewhat oversimplified. Overlapping undoubtedly takes place and it may be possible to get even more accurate results by calculating the number of atoms with octahedral coordination against the

various polymorphs; at present too few complete analyses and specimens are available to attempt this approach.

The study shows also that structural variations in single lepidolite crystals are common and can be coupled with compositional variations. The correlation may prove valuable in estimating the approximate Li_2O content of the mica without a chemical analysis. Normal muscovite may have as much as 3.3 percent Li_2O. Micas with 3.4 to 4.0 percent Li_2O are generally characterized by poor crystal development and anomalous optical properties owing to their transitional structures. Lepidolites with 4.0 to 5.1 percent Li_2O generally crystallize as the 6-layer monoclinic polymorph whereas those with more than 5.1 percent Li_2O have usually crystallized as the 1-layer monoclinic polymorph. Hexagonal lepidolite is commonly associated with 1-layer monoclinic polymorph, and both have essentially identical compositions.

C. A New 3-Layer Monoclinic Lepidolite Polymorph

A specimen of lepidolite from Skuleboda, Sweden (No. 476) was found to include a very small amount of uniaxial (2V as large as approximately 5°) material. The majority of the lepidolite is biaxial (2V = 37°) and has crystallized as the 1-layer polymorph. The biaxial and uniaxial portions occur in the different sections of the same sheets and are megascopically indistinguishable. The boundary between the two phases is microscopically sharp. X-ray studies of the uniaxial portion reveal the presence of an unreported 3-layer monoclinic polymorph.

Zero-level Weissenberg photographs have been taken about all a- and pseudo a-axes (120° from true a-axis) as well as b- and pseudo b-axes of this polymorph. A plane of symmetry in the position of b* as well as c* was recorded on only one of the photographs; therefore this pattern is considered as having been obtained from rotation about the true a-axis. Coupled with information from other Weissenberg photographs the crystal system is established as monoclinic. The Weissenberg pattern obtained by rotation about the 0-level a-axis is identical with 0-level a-axis photographs obtained from the 3-layer hexagonal polymorph. A 0-level pseudo a-axis Weissenberg is shown in Fig. 10. Among others, 0-level b-axis (Fig. 11) and 1-level a-axis (Fig. 12) photographs show some differences from the corresponding patterns of the 3-layer hexagonal polymorph. Differences along the 13ℓ reciprocal lattice line between the 3-layer monoclinic lepidolite and 3-layer hexagonal polymorphs may be seen by comparing Fig. 12 with the 1-level a-axis Weissenberg photographs illustrated by Axelrod and Grimaldi (1949). For all practical purposes the illustrations given by Axelrod and Grimaldi (1949) are identical with those which can be obtained from the 3-layer hexagonal polymorph originally described by Hendricks and Jefferson (1939). Similar differences are also obtained comparing any reciprocal lattice row in which k is divisible by
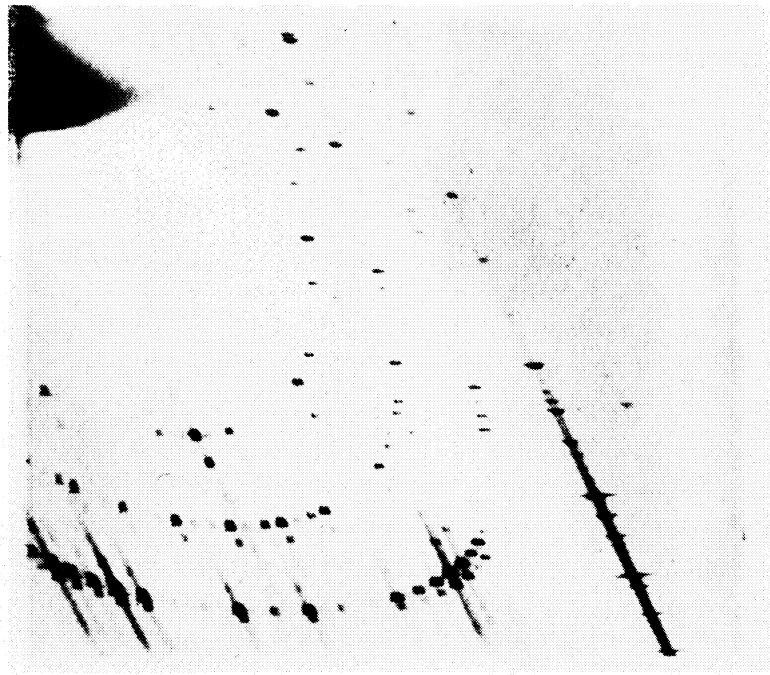
Fig. 10. 0-level pseudo a-axis Weissenberg photograph
obtained from 3-layer monoclinic lepidolite.
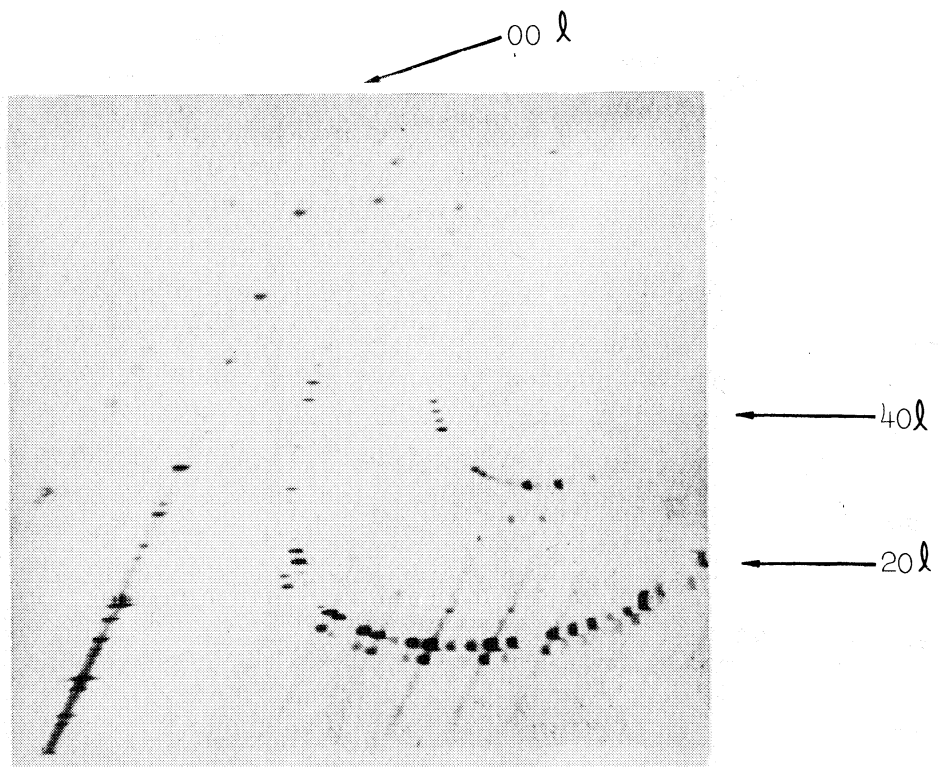
Fig. 11. 0-level b-axis Weissenberg photograph obtained from 3-layer monoclinic lepidolite.
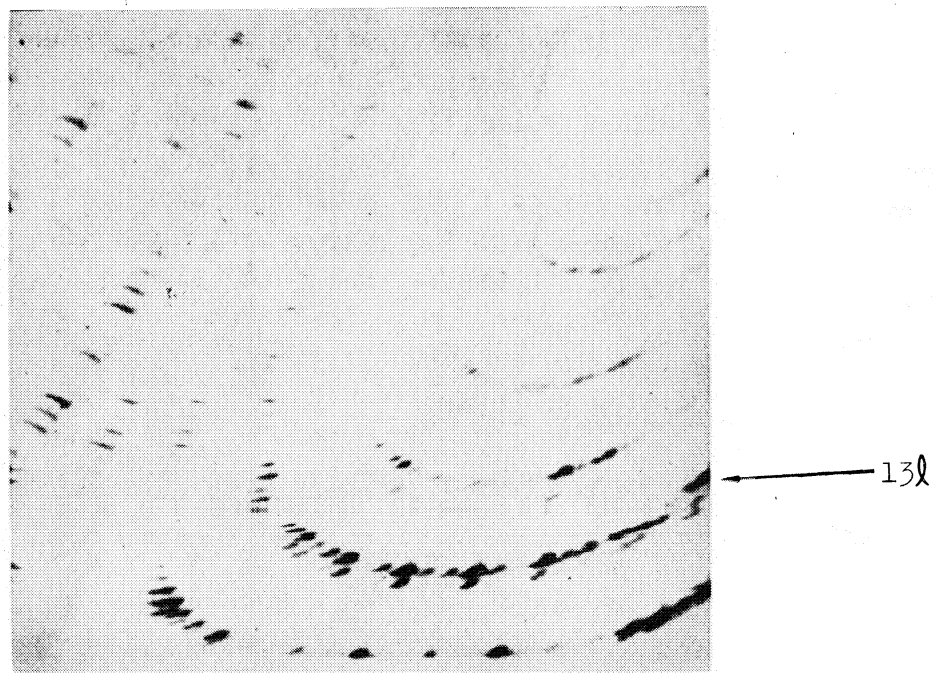
Fig. 12. 1-level a-axis Weissenberg photograph obtained from 3-layer monoclinic lepidolite.

3 (hkℓ, k = 3n; including h0ℓ reflections). Indexing showed that all reflections with h + k odd and (00ℓ) with $\ell \neq$ 3n were absent. The possible monoclinic space groups with the information thus far obtained are Cm, C2, and C2/m.

In comparing a- and b-axis photographs of the new form and the 1-layer monoclinic lepidolite (space group Cm) a very interesting relationship is observed. When the photographs are superimposed, every third reflection of the new form corresponds exactly with a reflection of the 1-layer polymorph. In between, two additional reflections will almost always be found on photographs of the new polymorph. This relationship also applies to photographs obtained by rotation about the pseudo a- and b-axes as well. Therefore it is logical to assume that the 3-layer monoclinic lepidolite contains the same symmetry elements as the 1-layer polymorph and thus has also crystallized in the monoclinic space group Cm. The unit cell dimensions are (approximately):

$$a_o = 5.2 \text{ Å}$$

$$b_o = 9.0 \text{ Å}$$

$$c_o = 30.0 \text{ Å}$$

$$\beta = 90° (\pm 30')$$

X-ray powder data of the 3-layer monoclinic lepidolite are given in Table XXXIV. In comparing the d-spacings and intensities with those given for the other mica polymorphs (Table XXXII) the existence of the new polymorph is substantiated. Also in Table XXXIV a quantitative spectrochemical analysis is given of the biaxial lepidolite (1-layer) which is found in the same book with the new structural variety. The Li_2O content of this specimen, however, does not correlate well with the results obtained for other micas in the muscovite-lepidolite series. The relationship of the new 3-layer monoclinic polymorph to other members of the muscovite-lepidolite series remains to be deciphered. It appears, however, that this polymorph is extremely rare.

D. Optical Properties Of Lepidolite

1. Indices, 2V, and Variation and Composition

α = 1.525 - 1.548   2V = 0 - 58° (negative)
β = 1.551 - 1.580 $\alpha \wedge c$ = 0 - -3°
γ = 1.554 - 1.586 $\beta \wedge a$ = 0 - -3°
$\gamma - \alpha$ = 0.018 - 0.038 γ = b

Optic plane normally parallel to (010).

TABLE XXXIV

SPECTROCHEMICAL AND X-RAY DATA ON LEPIDOLITE FROM SKULEBODA, SWEDEN (No. 476)

Analysis of 1-layer lepidolite*

SiO_2	=	48.
Al_2O_3	=	30.
K_2O	=	11.1
Fe_2O_3	=	.22
MgO	=	.01
Na_2O	=	.32
MnO	=	1.1
BaO	=	.0008
Rb_2O	=	.80
Li_2O	=	4.1
SnO_2	=	.006
Ga_2O_3	=	.016
TiO_2	=	.005
Sc_2O_3	=	----
SrO	=	.006
Cr_2O_3	=	----
CaO	=	.002
V_2O_5	=	.002
Cs_2O	=	.12
F	=	6.5

X-ray data on 3-layer monoclinic lepidolite**

Cu $K_{\alpha 1}$ = 1.53736kX

Intensity	d-spacing
s	9.91
m	4.98
m	4.54
w	3.87
m	3.56
s	3.32
w	3.18
vw	3.11
vw	2.95
vw	2.87
vw	2.83
ms	2.60
vvw	2.54
w	2.44
vvw	2.33
vw	2.26
vw	2.17
vw	2.09
vvw	2.02
m	1.99
vw	1.71
w	1.67
vw	1.63
vw	1.60
vvw	1.55
w	1.51
vvw	1.42
vvw	1.35
vw	1.30
vvw	1.28
vvw	1.24

**Measurements by C. H. Hewitt

*Spectrochemical analysis by C. E. Harvey

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The relationship between variation in optical properties and the chemical composition is complex, but the values of the indices of refraction do not depend upon the content of Li, but rather upon the amounts of ferric iron, ferrous iron and manganese that may be present. Lepidolites that are low in these three elements have indices of refraction and birefringences in the lower portions in the above listed ranges, whereas lepidolites containing appreciable amounts of ferric iron particularly, or of some ferric iron plus ferrous iron and manganese, have indices and birefringences that lie within the upper part of the above listed ranges. Apparently there is also a slight decrease in the size of 2V with increasing ferric iron or with increasing ferric iron plus ferrous iron and manganese.

Optical constants were measured on individual flakes whose structures first had been determined by the Weissenberg method. The results are presented in Table XXXV. The indices of refraction were measured on the Abbé refractometer or were obtained by the immersion method and are reproducible to $\pm$ 0.001. The Mallard method was used in the determination of 2E, and 2V was calculated. The only other determinations of the optical properties of lepidolite polymorphs have been by Hendricks and Jefferson (1939). Their results, though scanty, are in close agreement with those of this paper and are presented below. The values are averages and the numbers in parentheses indicate the number of published measurements used in calculating the averages.

6-layer lepidolite

α = not reported β = not reported γ = 1.559 (3)   2V = 32° (3)

3-layer hexagonal lepidolite

α = 1.525 (1) β = not reported γ = 1.558 (1)   2V = 0° (1)

1-layer lepidolite

α = 1.533 (1) β = not reported γ = 1.559 (12)   2V = 41° (12)

The lepidolite polymorphs are not distinguishable on the basis of optical properties alone; it follows therefore that their general chemistry cannot be deduced from the optical constants. A trend is that the indices, and particularly 2V, of the 1-layer polymorph tend to be slightly higher than those of the 6-layer polymorph, but the overlap is broad. This can be accounted for by the optical inactivity of the Li-ion. Although variations in Li content influence the layer stacking pattern, changes in optical constants are more sensitive to variations in Fe^3, Ti, Fe^2 and Mn, both in muscovite and the lepidolites. The indices of lepidolite vary within books (note values obtained from spec. 514 and 452 in Table XXXV). However, the indices of lithian muscovite are in the range of normal lepidolites whose values are considerably

TABLE XXXV

INDICES OF REFRACTION AND 2V OF LEPIDOLITES
AND LITHIAN MUSCOVITE

Specimen Number	Locality	α	β	γ	2V
	6-layer lepidolite				
502	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.555	1.560	30°
514(2)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.553	1.557	30°
514(3)	Opportunity Pegmatite, Ohio City, Colorado	1.533	1.555	1.560	33°
514(4)	Opportunity Pegmatite, Ohio City, Colorado	1.533	1.557	1.560	33°
514(5)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.555	1.558	--
514(6)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.553	1.560	33°
505(b)	Brown Derby Pegmatite, Ohio City, Colorado	1.530	1.556	1.560	34°
967(a)	Stewart Mine, Pala, California	1.530	1.552	1.559	33°
967(b)	Stewart Mine, Pala, California	1.529	1.554	1.558	34°
970(a)	Stewart Mine, Pala, California	1.534	1.555	1.559	33°
452(a)	Varuträsk, Sweden	1.532	1.556	1.561	33°
452(b)	Varuträsk, Sweden	1.535	1.556	1.561	35°
452(c)	Varuträsk, Sweden	1.532	1.552	1.557	37°
	3-layer hexagonal lepidolite				
539(f)	Pope mining claim, Southern Rhodesia	-	1.568	1.568	0°
539(g)	Pope mining claim, Southern Rhodesia	-	1.566	1.566	0°
	1-layer lepidolite				
514(k)	Opportunity Pegmatite, Ohio City, Colorado	-	1.556	1.558	-
464(b)	Newry, Maine	-	1.556	1.558	50°
471(b)	South Portland, Maine	1.533	1.553	1.560	41°
476(b)	Skuleboda, Sweden	-	1.562	1.564	-
476(c)	Skuleboda, Sweden	-	1.564	1.566	37°
535(c)	Dogon Daji, Nigeria	-	1.557	1.562	47°
539(a)	Pope Mining Claim, Southern Rhodesia	-	1.562	1.565	38°
539(b)	Pope Mining Claim, Southern Rhodesia	-	1.562	1.566	37°
	2-layer lithian muscovite				
556(b)	Tördal, Norway	-	1.553	1.556	34°
556(c)	Tördal, Norway	1.532	1.554	1.558	34°
465(a)	Newry, Maine	1.533	1.555	1.559	34°
465(f)	Newry, Maine	1.534	1.555	1.561	37°
471(a)	South Portland, Maine	1.529	1.551	1.556	32°
679(a)	Usakos, S. W. Africa	1.534	1.559	1.566	-

Specimens numbered such as 514(2) through 514(6), or 452(a) through 452(c) represent measurements obtained from different flakes in the same book.

below those of normal muscovite. It is well known that indices are very sensitive to a small amount of certain isomorphous substitutions. Winchell (1942) showed that practically all lithia micas show variations of the indices within single flakes but attributed the phenomena to other causes.

2. <u>Variation of Color with Composition</u>. It has been demonstrated by Heinrich and Levinson (1953) that the rose muscovites owe their color to the essential absence of Fe^2 and the dominance of Mn over Fe^3. Under such conditions it is possible for the Mn to be oxidized to the higher valence (Mn^3) in which state it becomes a powerful pigmenting agent, even though, present in only very small amounts. They state, (p. 46):

> "In lepidolites, the color variations also are related to fluc-
> tuations in the Fe/Mn ratio. The role of Mn in the coloration
> of lepidolite has been stressed, for example, by Hintze, 1897
> and by Doelter, 1917. As stated by Shibata (1952, p. 160),
> 'Lepidolite is pink or violet, which is due to the color of
> manganese. Lepidolite is sometimes colorless when it is poor
> in MnO. Lithium is not a color agent....' Those lepidolites
> that are colorless or of very pale colors usually have Fe:Mn
> $\cong$ 1:1; those that are gray normally have Fe > Mn, whereas
> the purple lepidolites contain more Mn than Fe."

In our work we have been able to confirm the above conclusions in greater detail on the basis of new spectrochemical analyses of lepidolites. From the data presented in Table XXXVI,XXXVII, and XXXVIII, the following generalizations can be drawn:

Mn:Fe (total Fe)

greater than $\cong$ 6:1		deep shades of purple
between	$\cong$ 3:1 and 5:1	medium shades of purple
between	$\cong$ 1:1 and 3:1	colorless to pale tints of purple
between	$\cong$ 1:1 and 2:1	colorless to gray (where amount of Fe is less than 1 percent)

Where the Fe content rises much above 1 percent, it apparently masks the effect of equal or even larger amounts of Mn and results in a brownish color (see specimens 1143,1016,990 in Tables XXXVII and XXXVIII). The effect of small amounts of Ti seems considerable in some cases. Ti apparently adds a brown tint in specimens No. 504 and No. 514 (Table XXXVI).

Accurate color descriptions of lepidolites as well as other micas are difficult to obtain. Physical factors which influence color are size of books, impurities, state of aggregation and grain size. The colors listed in Tables XXXVI, XXXVII, and XXXVIII are as accurate as we are able to ascertain. The specimens were compared under the same light conditions. It is instructive

to compare our color descriptions with those of Stevens (1938) on his analyzed micas made available for our study (Table XXXVIII). It would also be constructive to compare the variations in chemical analyses of selected oxides as listed in Table XXXVII.

TABLE XXXVI

RELATIONSHIP BETWEEN COLOR AND COMPOSITION IN LEPIDOLITES; NEW ANALYSES*

Number	Locality	MnO	Fe_2O_3	TiO_2	Color	Remarks
464 a	Newry, Me.	.10	.06	--	pale purple	coarse-grained
464 b	" "	.11	.10	--	"	fine-grained
476	Skuleboda, Sweden	1.1	.22	.005	deep purple	
483	Newry, Me.	.09	.20	--	gray	
488	Pala, Calif.	.10	.03	--	pale purple	
499	Brown Derby, Colorado	.23	.05	.015	medium purple	
500	Bazooka peg, Colorado	.13	.09	--	pale purple	
503	Brown Derby, Colorado	.45	.06	.012	deep purple	
504	Brown Derby, Colorado	1.7	.17	.03	deep purplish brown	

(464 a and 464 b: same specimen)

*Spectrochemical analyses by C. E. Harvey

TABLE XXXVI (cont.)

Number	Locality	MnO	Fe$_2$O$_3$	TiO$_2$	Color	Remarks
508	Brown Derby, Colorado	.08	.06	--	pale purple	
511	Brown Derby, Colorado	.05	.05	--	very pale purple	curved
514 a	Brown Derby, Colorado	1.2	.27	.005	medium brownish purple	two analyses of same specimen
514 b	Brown Derby, Colorado	6.0	.60	.01	medium brownish purple	
535	Nigeria	.46	.04	--	deep purple	
539 a	S. Rhodesia	5.5	1.1	.045	deep purple	biaxial zone
539 c	S. Rhodesia	6.0	2.2	.08	deep purple	uniaxial zone
991	Auburn, Maine	.39	.69	--	colorless	
1097	Rozena	.37	.08	--	deep purple	

TABLE XXXVII

RELATIONSHIP BETWEEN COLOR AND COMPOSITION IN LEPIDOLITES; REANALYSIS OF LEPIDOLITES REPORTED IN THE LITERATURE*

Number	Locality	MnO	Fe_2O_3	TiO_2	Color	Reference	Remarks
521	Varuträsk	.35	.10	--	medium purple	this report	Li_2O = 3.8
521	Varuträsk "	.28	.10	0.00	---	Berggren (1940) Anal. C	Li_2O = 3.9, FeO = 0.00
531	Varuträsk	.10	.04	--	gray	this report	Li_2O = 4.1
531	Varuträsk "	.00	.17	0.00	gray-white	Lundblad (1942) Anal. 10	Li_2O = 4.55, FeO = 0.00
532	Uto	.18	.18	--	white	this report	Li_2O = 4.2
532	Uto	.28	.15	0.00	---	Lundblad (1942) Anal. 13	Li_2O = 5.7, FeO = 0.02
533	Rozena	.46	.08	--	deep purple	this report	Li_2O = 2.1
533	Rozena	.52	.06	0.00	---	Lundblad (1942) Anal. 14	Li_2O = 5.5, FeO = 0.00
1143	Quebec	2.0	2.3	.12	pale brown	this report	Li_2O = 4.2
1143	Quebec	4.19	2.52	--	---	Ellsworth, (1932) Spec. XXXII	Li_2O = 5.44

*Spectrochemical analyses by Charles E. Harvey

TABLE XXXVIII

RELATIONSHIP BETWEEN COLOR AND COMPOSITION IN LEPIDOLITES;
SPECIMENS IN OUR LABORATORY FOR WHICH ANALYSES ARE RECORDED IN THE LITERATURE

Number	Locality	MnO	FeO	Fe_2O_3	TiO_2	Color	Reference
966	Ramona	2.06	.21*		.02	pink	Stevens (1938) No. 15
967	Pala, Calif.	.92	.04		tr.	medium purple	do. No. 3
969	Antsongombato	.72	.12		.03	medium purple	do. No. 16
970	Pala, Calif.	.52	.16		tr.	very pale purple	do. No. 6
971	Mesa Grande	.14	.11		tr.	pale purple	do. No. 8
972	Mesa Grande	.17	.04		tr.	medium purple	do. No. 9
973	Mesa Grande	.61	.07		.01	medium purple	do. No. 13
975	Pala, Calif.	.65	.05		tr.	medium purple	do. No. 5
977	San Diego Co.	.28	.04		tr.	medium purple	do. No. 4
978	W. Australia	.76	.23		.06	pale purple	do. No. 14
990	Usakos	1.48	0.00	2.34	tr.	pale brown	Jakob (1927)
1016	Kimito	.75	1.52	1.55	0.00	pale brown	Pehrman (1945)
1259	Maharitra	.45	--	.03	--	deep purple	Mauguin (1928B)
1260	Antsongombato	.24	--	.52	--	colorless	Mauguin (1928B)

*All Fe is reported as FeO by Stevens (1938) in the micas listed in this table.

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II. OCCURRENCE

A. Nonpegmatite

There are very few authenticated occurrences of lepidolites outside of pegmatites. The presence of lepidolites in the hydrothermal contact aureole of granites has been suggested by Harker (1932, p. 123).

Lepidolite also has been reported from a few granites, aplites, and high temperature veins and lodes, particularly those containing tin and tungsten minerals. That these micas are lithium-bearing has been shown in some cases, but few have been demonstrated to be lepidolites in composition or in structure.

B. Pegmatite

1. Localization. In pegmatites, lepidolite and lithian muscovites occur in:

a. Fracture-fillings, not uncommonly with cleavelandite, quartz, and tourmaline.

b. Replacement bodies, also commonly with quartz, albite (incl. cleavelandite), and a variety of accessory constituents.

Some investigators believe that lepidolite also can form as a primary constituent of inner zones (Cameron, et al., 1949, p. 68), but the number of such examples, even if they are truly of magmatic origin, is doubtless small indeed.

The major lepidolite areas of the world are:

United States:

Maine - Black Mtn., Rumford and Mt. Mica; Paris, Me.
Colorado - Brown Derby, Gunnison County
New Mexico - Harding Mine, Taos County
California - Stewart Mine, Pala

Southeastern Manitoba, Canada

Varuträsk, Sweden

Haut Vienne, France

Karibib, South-west Africa

Pilbara area, Western Australia

Madagascar

Portuguese East Africa

Southern Rhodesia

2. Macrostructural Features. a. General: Within individual pegmatites the grain size of lepidolite may vary enormously; although usually within a single replacement body, the grain size is relatively uniform. The grain size of lepidolite ranges from microscopic, which results in megascopically massive or structureless aggregates, to very coarse books, 10 to 12 inches across. Some of these larger books display structural defects identical with those found in muscovite-A, herringbone, wedge, and concentric structure. Mineral inclusions also may be common, as are fractures and rulings. Wavy and rippled books are very common, and even small books with completely flat sheets are rare.

b. Overgrowths: Overgrowths of lepidolite on muscovite have been found in pegmatites at several localities. The lepidolite commonly has formed as a narrow rim around a broad core of muscovite. The contact between the two may be sharp and show the general euhedral crystal outline of the muscovite core, or it may be irregular and appear to be somewhat corroded. In one type a narrow intermediate band of lepidolite in parallel position and having a common basal cleavage plane with the muscovite is found in direct contact with the muscovite and a third zone of granular lepidolite forms the extreme margin of the crystal. This latter type of lepidolite is highly twinned in tiny rhombic units. In a few cases pink fibrous muscovite coats the exterior of the crystal (Bowman, 1902).

Under the microscope, the twinned lepidolite aggregate does not extinguish as a unit. Because of the small size of the lepidolite units it is almost impossible to obtain the relationship of percussion figures and optic planes between the two species near the boundary. However, three successful determinations were made that show the optic planes of the two species to be either parallel or at an angle of 30°. The results of a study of a group of specimens of lepidolite overgrown on muscovite are given in the following list:

Specimen No.	Location	Notes
417	Eight Mile Park, Colorado	granular lepidolite rim, straight boundary
461	Auburn, Maine	single-crystal lepidolite rim, common basal cleavage, border irregular and corroded, optic planes parallel
467	Topsham, Maine	granular lepidolite rim, boundary straight in some sections and corroded in others
468	Topsham, Maine	granular lepidolite rim, boundary corroded
469	Topsham, Maine	granular lepidolite rim, boundary corroded
470	Topsham, Maine	granular lepidolite rim, boundary corroded
539	Southern Rhodesia	broad sheet of muscovite with inner rim of uniaxial muscovite and outer rim of biaxial lepidolite, all three zones have common cleavage, optic planes at 30°
651	Auburn, Maine	granular lepidolite rim
652	Auburn, Maine	single-crystal lepidolite rim, boundary straight, common basal cleavage
656	Auburn, Maine	single-crystal rim of lepidolite, boundary straight, common basal cleavage, optic planes at angle of 30°
713	Topsham, Maine	muscovite core, thin rim of single crystal lepidolite, outside rim of granular lepidolite, boundaries corroded, muscovite and thin rim of lepidolite have common cleavage
717	Auburn, Maine	granular lepidolite rim, corroded boundary

Reports of overgrowths from Maine have been made by Clarke (1888), Wolff and Palache (1902), Bastin (1911), and Baumhauer (1912). Overgrowths have been found at Paris, Auburn, Mount Apatite, and Minot. Generally, the lepidolite is fibrous or granular around a core of muscovite. A few occurrences of crystallographically continuous lepidolite on muscovite have been reported from the Wade and Pulsifer gem quarries, Androscoggin County, Maine. One diamond-shaped book of muscovite a foot across with a border zone of lepidolite 4 inches wide was found there.

The occurrences at Haddam Neck, Connecticut, have been studied by Bowman (1902) and Sterrett (1923). The columnar mica crystals occur with smoky quartz, albite, microcline, cookeite, and tourmaline. The outer surface of the mica crystals has a fibrous appearance caused by a thin layer of fibrous pink muscovite. The fibers are parallel to the long axis of the crystal. Cleavage in the fibers is perpendicular to their long axis. The lilac lepidolite surrounds a rhombic or hexagonal core of green-white muscovite. All three zones have a continuous cleavage. Under the microscope, the lepidolite does not extinguish completely because of the superposition of layers of material in twin position.

Jahns and Wright (1951) report lepidolite rims fringing a green core of muscovite. The two micas are crystallographically continuous and have a common basal cleavage. Heinrich (1948) has observed similar overgrowths in the Meyers Quarry pegmatite, Eight Mile Park, Colorado.

A regular overgrowth of lepidolite on muscovite has been reported by Scharizer (1886, 1887B) from Schüttenhofen, Germany. A rhombic core of muscovite is surrounded by a broad crystallographical continuous rim of pink lepidolite. Three separate irregular units were found in one specimen. In two of these units the optic plane is normal to the optic plane of the muscovite; in the third unit, the optic plane makes an angle of 30° with the optic plane of the muscovite.

Pehrman (1945) has described an interesting and rather complex overgrowth relationship between muscovite and lepidolite among the micas from Kimito, Finland. Several zones of lepidolite are involved.

Overgrowths of lepidolite on biotite are rare and have been reported from only a few localities; no reports of optical relationships have been found. Landes (1925) describes a 6-mm-wide zone of lepidolite around biotite plates from gneiss xenoliths in the granite pegmatites of central Maine. He attributes the lepidolite to reaction between the biotite xenoliths and the lithia-rich magma.

Clarke (1888) reports an overgrowth from Cape Ann, Massachusetts, in which a crystal of black lepidomelane is surrounded in part by a border of small crystals of a "dark greenish-black lithia mica, presumably cryophyllite". Analyses of the two micas are as follows:

	Border Granular Cryophyllite	Core Lepidomelane
SiO_2	52.17	31.69
Al_2O_3	16.39	11.93
Fe_2O_3	4.11	8.06
FeO	6.08	30.35
MnO	0.32	0.21

CaO	Trace	0.23
MgO	Trace	0.05
Li_2O	5.03	Trace
Na_2O	0.60	1.54
K_2O	10.54	8.46
H_2O	1.43	4.25
F	7.02	Trace
TiO_2	-----	3.42

3. _Compositional Variation in Individual Pegmatites and Districts._
Table XXXIX lists five new analyses of paragenetically distinct lepidolites
from the Brown Derby pegmatite, Gunnison County, Colorado. Despite the fact
that they differ markedly in grain size, texture, association and somewhat
in color, the compositional differences are relatively small. The major
differences occur in Mn, Na, H_2O^+ and F; minor in Li. Analyses 7 and 10 are
by two other analysts of our lepidolite 504.

Table XL lists new spectrochemical analyses of three lepidolites
and one lithian muscovite from Newry, Maine; again the principle differences
are in Mn, Na, F and Rb.

Table XLI lists reanalyses of lepidolites from the Varuträsk peg-
matite, Sweden, analyzed by Berggren; the agreement between the two sets of
results is good.

In Table XLII are reported three analyses of lepidolites from two
other pegmatites in the same district as the Brown Derby. Numbers 514a and
514b, from the same crystal, differ considerably from one another (notice
MnO content) and also from 500, as well as from the Brown Derby lepidolites
(Table XXXIX). Thus it appears that paragenetically distinct lepidolites may
differ considerably in composition or in other cases they may be rather
similar.

TABLE XXXIX

ANALYSES OF LEPIDOLITES FROM BROWN DERBY PEGMATITE,
GUNNISON COUNTY, COLORADO

Oxide	Specimen Nos.*						
	499	503	504	508	511	7	10
SiO_2	49.97	51.99	49.88	54.82	50.50	49.58	49.19
Al_2O_3	27.46	23.59	24.04	23.79	26.54	23.87	24.81
TiO_2	.01	.02	.08	.01	.02	0.06	0.08
Fe_2O_3	.02	.02	.14	.07	.02	-	$\left\{ 0.24 \right.$
FeO	.02	.02	.00	.07	.01	.21	
MnO	.22	.44	2.20	.04	.04	2.78	2.51
CaO	.005	.002	.004	.002	.003	none	trace
MgO	.01	.015	.03	.01	.01	none	0.05
BaO	.0004	.0003	.0004	.0003	.0004	n.d.	n.d.
SnO_2	.019	.024	.021	.026	.023	n.d.	n.d.
SrO	.006	.006	.005	.006	.006	n.d.	n.d.
Li_2O	4.35	5.30	5.24	4.81	4.83	5.05	5.10
Na_2O	.90	1.64	1.10	.52	.52	0.57	0.52
K_2O	9.05	9.00	9.35	8.50	9.04	10.14	10.25
Rb_2O	2.70	2.60	2.50	2.50	2.95	1.62	1.78
Cs_2O	.25	.15	.11	.16	.25	.09	0.19
H_2O+	1.54	.62	.77	1.00	1.59	1.22	$\left\{ 1.21 \right.$
H_2O-	.32	.36	.39	.28	.31	.51	
F	6.22	7.79	7.94	6.45	6.69	7.49	6.89
O = F	2.62	3.28	3.34	2.72	2.82	3.15	2.90

*Nos. 499, 503, 504, 508, 511, Analyst: R. L. Craig, Electrotechnical
Laboratory, U. S. Bureau of Mines, Norris, Tenn.; BaO, SnO_2, and SrO
determined spectrochemically by C. E. Harvey.

No. 7. R. E. Stevens, Am. Mineral., 23, p. 615 (1938).

No. 10. A. N. Winchell, Am. Mineral., 27, p. 115 (1942).

TABLE XL

SPECTROCHEMICAL ANALYSES* OF MICAS FROM NEWRY, MAINE

Oxide	Specimen Nos.			
	464a	464b	465	483
Fe_2O_3	.06	.10	.23	.20
MgO	.01	.015	.01	.02
MnO	.10	.11	.64	.09
TiO_2	---	---	---	---
CaO	.004	.004	.37	.05
SrO	---	---	---	---
BaO	---	---	.0004	.0002
Na_2O	.62	.33	.40	.50
Li_2O	4.3	4.0	4.1	3.9
Rb_2O	.38	.12	.52	.36
Cs_2O	.08	.10	.24	.10
Ga_2O_3	.018	.008	.007	.010
Sc_2O_3	---	---	---	---
Co_2O_3	---	---	---	---
Cr_2O_3	---	---	---	---
SnO_2	.023	.015	.001	.023
V_2O_5	---	---	---	---
F	6.3	5.8	5.2	6.8

464a: coarse-grained lepidolite
464b: fine-grained lepidolite from same specimen as 464a.
465: lithian muscovite
483: lepidolite

*Analyses by C. E. Harvey

TABLE XLI

REANALYSIS* OF TWO LEPIDOLITES FROM VARUTRÄSK, SWEDEN ANALYZED BY BERGGREN

Oxide	Specimen Nos.	
	521	531
Fe_2O_3	.10	.04
MgO	.008	.007
MnO	.35	.10
TiO_2	---	---
CaO	.002	.003
SrO	.006	.008
BaO	.0004	---
Na_2O	.45	.21
Li_2O	3.8	4.1
Rb_2O	.48	.70
Cs_2O	.09	.19
Ga_2O_3	.013	.01
Sc_2O_3	---	---
Co_2O_3	---	---
Cr_2O_3	---	---
SnO_2	.016	.013
V_2O_5	---	---
F	3.2	4.8

521 is reanalysis of Berggren (1940), Analysis C.
531 is reanalysis of No. 10 reported in Lundblad (1942): analyzed by Berggren.

*Spectrochemical analyses by C. E. Harvey

TABLE XLII

SPECTROCHEMICAL ANALYSES OF LEPIDOLITES FROM TWO OTHER PEGMATITES
IN BROWN DERBY DISTRICT

Oxide	Specimen Nos.		
	514a	514b	500
Fe_2O_3	.27	.60	.09
MgO	.008	.02	.15
MnO	1.2	6.0	.13
TiO_2	.005	.01	---
CaO	.002	.001	.032
SrO	.006	.002	.009
BaO	.0004	---	.0006
Na_2O	.47	.48	.72
Li_2O	4.4	4.0	3.0
Rb_2O	.73	.46	.80
Cs_2O	.10	.10	.07
Ga_2O_3	.011	.016	.03
Sc_2O_3	---	---	---
Co_2O_3	---	---	---
Cr_2O_3	---	---	---
SnO_2	.005	.006	.01
V_2O_5	---	---	---
F	4.5	5.9	5.8

514a: Untwinned, well-developed portions of lepidolite crystal from
Opportunity pegmatite, Gunnison Co., Colorado.

514b: Twinned, poorly developed portions of same book as 514a.

500: Bazooka pegmatite, Gunnison Co., Colorado.

PART IV. PHLOGOPITE

I. MINERALOGY

A. Composition

1. General. In phlogopite, $K_2(Mg,Fe)_6(Si_6Al_2)O_{20}(OH)_4$, Na can substitute for K up to nearly K:Na = 1:1 (Harada, 1936 and Heinrich, 1946); also minor Rb,Cs and Ba for K. Fe_2 is almost always present but Mg predominates. Fe^3,Mn, and Ti usually present in small amounts. The Si: tetrahedral Al ratio varies somewhat and may be larger than 6:2. Heinrich (1946) has shown the relationship between composition and geologic occurrence. Synthetic phlogopite is characterized by F in place of (OH). Manganophyllite may be classed as phlogopite normally with Mg>Mn>Fe and in some types with some Fe^3 and Mn^3 as well·

A complete series exists between phlogopite and biotite. There is no well-defined, natural, compositional boundary between ferroan phlogopite and magnesian biotite. If a division is required, we suggest that where the ratio of Mg:Fe>4:2 the mineral should be classed as phlogopite.

Prider (1940) reports a phlogopite from a phlogopite-leucite-lamproite from Western Australia which contains 8.97 percent TiO_2. The formula based on the analysis is:

$$(K,Na,Ba,Ca)_{2.06}(Mg,Fe^2,Fe^3,Ti)_{5.84}(Si,Al,Ti,)_{8.00}O_{20}(OH,F)_{2.16}$$
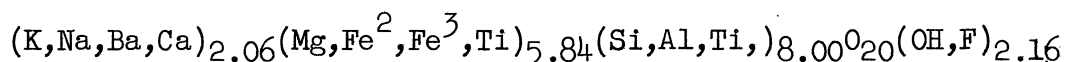
Prider (1940) notes that the low (OH,F) content is probably not due to loss of volatiles during crystallization at shallow depths and implies that the mineral may have formed from an extremely 'dry' magma. He also observes that other comparable micas with high Ti have low hydroxyl content. Tetrahedral positions are occupied by 0.15 atoms of Ti, as Si and Al alone are insufficient to fill these positions; 0.84 atoms of Ti occupy octahedral positions. A discussion of the role of Ti in the mica structure is presented under biotite chemistry.

2. Mahadevite. It has been established that no complete series (solid solution) exists between muscovite and either phlogopite or biotite. In considering these types of micas, Winchell's (1925, 1927, etc.) classification of heptaphyllite and octophyllite is correct. Ramaseshan (1945) and Venkayya (1949), however, have described a mica for which they propose the new species name mahadevite, which they believe to be intermediate

between muscovite and phlogopite. This conclusion is based upon both chemical and optical data. They note that the high MgO content (13 to 17 percent) eliminates the possibility of the mica being muscovite, whereas the high Al_2O_3 content (27 to 30 percent) eliminates phlogopite. The formula for this mica according to Hey (1950) is near $(K,Na)_{0.97}(Al,Fe,Mg)_{2.66}(SiAl)_4(O,OH)_{12}$. By means of percussion figures the optic plane was determined as being perpendicular to (010). (We have found this procedure unreliable).

Through the courtesy of Dr. H. S. Yoder we have been fortunate in obtaining a small amount of mahadevite for X-ray study. From 0-, 1- and 2-level a-axis Weissenberg photograph we find that this mica has crystallized as the 1-layer monoclinic polymorph, with intensities similar to those of other phlogopites. We also found the optic plane to be parallel with (010), as is natural with 1-layer micas, and 2V to be smaller ($<5°$) than the value (approximately 14°) indicated by Ramaseshan (1945) and Venkayya (1949). The physical appearance of the specimen is that of a typical phlogopite and there is no interlayered muscovite to account for the high Al_2O_3 content reported.

Our conclusion is that mahadevite is probably an Al-rich, Mg-, Fe^2-poor phlogopite, close to Winchell's (1935) end-member eastonite, $K_2(Mg_5Al)$ $(Si_5Al_3)O_{20}(OH)_4$.

B. Structure

1. **Polymorphism.** Hendricks and Jefferson (1939) report phlogopite as occuring in 3 polymorphs: the 1- and 2-layer octophyllite monoclinic, and the 3-layer hexagonal polymorphs. This is the only X-ray study of polymorphism in phlogopites reported in the literature.

The structures of approximately 80 phlogopite crystals from about 50 different specimens (28 chemically analyzed) were determined by means of the Weissenberg method. All but five have crystallized with the 1-layer monoclinic structure. Of those five, three have crystallized as the 2-layer monoclinic polymorph, whereas the remaining two have crystallized with 3-layer hexagonal structure (Table XLIII).

TABLE XLIII

2- AND 3-LAYER PHLOGOPITES

Number	Location	Reference	Structure
1062 (a and b)	Madagascar	Jakob and Parga-Pondal (1932) No. 65	2-layer monoclinic
1064 (a and b)	Madagascar	Jakob and Parga-Pondal (1932) No. 63	2-layer monoclinic
1261 (b and c)	Madagascar	Mauguin (1928B)	3-layer hexagonal
1220 (a)	Hull, Quebec	Not analyzed	2-layer monoclinic
1227 (a)	Labelle Co., Quebec	Not analyzed	3-layer hexagonal

It is noteworthy that three of the above specimens are from Madagascar, but that at least four other specimens of Madagascar phlogopite have crystallized with the common 1-layer monoclinic structure. A third crystal obtained from the material supplied by Professor Mauguin (No. 1261 (a)) has crystallized as the 1-layer monoclinic form; there probably is little difference in the composition of these several crystals from the same specimen. The specimens of known composition that have crystallized as the 2- and 3-layer forms do not appear to be chemically aberrant. In fact, the compositions of all the Madagascar varieties are practically identical.

A few of the phlogopite patterns show diffuse scattering of varying intensity. No correlation of diffuse scattering with polymorphism or chemistry was obtained.

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 specimen supplied by him. Weissenberg photographs indicate that the structure is the common 1-layer monoclinic form. However, from personal communication with Professor Pieruccini we learn that two distinct varieties of phlogopite are to be found in his specimens. It is possible, therefore, that we did not X-ray the crystals he believes to be triclinic.

2. <u>Relation of Structure to Composition</u>. An attempt was made to correlate chemistry, polymorphism and paragenesis in the dark-colored micas. In all, approximately 60 analyzed biotites, phlogopites, and manganophyllites with varying compositions and different paragenesis were X-rayed. The results, incorporated in Table XLIV, indicates that there is no evident relationship between chemistry, polymorphism and paragenesis among the dark-colored micas. Furthermore, the fact that the three different reported polymorphic modifications found in phlogopites as a whole are found in the Madagascar phlogopites apparently rules out the possibility of temperature or pressure as a major factor; undoubtedly the conditions of formation were fairly uniform for these very similar micas.

C. <u>Optical Properties</u>

The refractive indices for phlogopite are:

α = 1.530 - 1.573 γ-α = 0.028 - 0.049

β = 1.557 - 1.617   2V = 0° - 12° (negative)

γ = 1.558 - 1.618   r<v weak to distinct

$\alpha \wedge c = 0° - 4°, \beta = b, \gamma \wedge a = 0° - 5°$

Optic plane parallel with (010) generally; only in 2-layer forms
(which are rare) is it perpendicular to (010).

In a few rare types 2V may be somewhat larger than the above indicated range. The indices and birefringences increase with Fe^2 and Mn, but change more rapidly for similar increments of Ti and Fe^3. Some phlogopites with much Ti or Ti plus Fe^{+3} have indices as high as α = 1.599, γ = 1.643.

Phlogopites vary in color from colorless to nearly black or brownish-black in hand specimen. In thin slices many of them are normally colorless, pale yellow, orange, buff or light reddish brown. The depth of the color increases with an increase of Fe^2 over Mg and particularly with increasing Fe^3 and Ti. The latter probably is responsible for the reddish tints that some phlogopites display.

TABLE XLIV

STRUCTURE OF ANALYZED BIOTITES, PHLOGOPITES, AND MANGANOPHYLLITES

ANALYZED BIOTITES

Number	Reference	Structure	Paragenesis	FeO	Fe2O3	MgO	TiO2
728	Glass (1935)	3-layer	pegmatite	26.72	2.87	n.d.	3.60
729	Stevens (1946)	1-layer	pegmatite	8.96	3.31	16.15	1.11
797	Grout (1924) No. 1	1-layer (+3-layer ?)	basic segreg. in granite	7.72	7.44	16.55	1.67
798	Grout (1924) No. 2	3-layer (mod. scattering)	granite	14.80	4.05	10.21	2.23
799	Grout (1924) No. 3	2-layer	granite	23.23	3.03	9.24	3.32
800	Grout (1924) No. 4	1-layer	granite	23.75	1.14	6.16	4.34
801	Grout (1924) No. 5	1-layer	peridotite	12.96	8.67	n.d.	1.50
994	vanBiljon (1940)	2-layer	granite	13.58	4.23	12.16	3.89
960	Pagliani (1949)	2-layer	mica schist	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	16.38	3.28	8.99	2.45
1011	Kawano (1942)	1-layer	lepidomelane-quartzfels	23.27	7.81	4.32	2.42
1084	Jakob (1931) No. 57	2-layer (weak scattering)	2-mica pegmatite	10.47	4.09	13.19	2.06
1085	Jakob (1931) No. 58	2-layer	2-mica pegmatite	9.72	2.24	14.22	1.64
1086	Jakob (1931) No. 59	(3-layer ?)	2-mica pegmatite	16.26	4.03	8.46	3.16
1087	Jakob (1931) No. 60	2-layer (weak scattering)	2-mica pegmatite	16.85	4.08	8.06	2.71
1088	Jakob (1931) No. 61	1-layer	lamprophre	15.84	5.03	11.17	1.95
1089	Jakob (1937)	1-layer	feldspar pegmatite	28.61	0.00	6.72	1.99

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TABLE XLIV (CONT.)

Number	Reference	Structure	Paragenesis	FeO	Fe2O3	MgO	TiO2
1117	Coats and Fahey (1944)	1-layer (mod. scattering)	pegmatite (siderophyl-lite)	30.16	tr.	0.22	0.02
1140	C. O. Hutton (unpubl.)	2-layer	from sands derived from granitic rocks, Monterey Pen., Calif.	14.49	9.30	5.80	3.47
1145	Hallimond (1947)	1-layer	marble	6.8	1.1	18.7	1.9
1257	Hutton and Seeyle (1947)	2-layer	pegmatite-like lense	14.41	3.92	11.11	3.02
1262	Mauguin (1928B)	1-layer (heavy scattering)	from Tschebarkul	12.77	7.09	13.30	1.16
1350	Yamada and Sugiura (1950)	1-layer (weak scattering)	pegmatite	3.49	0.67	24.24	0.64

TABLE XLIV (CONT.)

ANALYZED PHLOGOPITES

Number	Reference	Structure	Paragenesis	FeO	Fe_2O_3	MgO	TiO_2	F
1054	Jakob (1938)	? Very diffuse scattering	from Morawitza	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 (1932)*No. 73	1-layer	Revision of 55	2.52	0.00	25.45	2.80	0.00
1057	Jakob (1932) No. 70	1-layer	Rossie, N. Y.	4.79	0.00	22.30	4.07	0.12
1058	Jakob (1932) No. 69	1-layer	Hull, Quebec	2.49	0.71	24.60	1.74	2.04
1059	Jakob (1932) No. 68	1-layer	Burgess, Ontario	0.30	0.00	27.32	0.39	6.74
1060	Jakob (1932) No. 67	1-layer	Burgess, Ontario	1.50	0.00	26.14	0.86	2.37
1061	Jakob (1932) No. 66	1-layer	Isolo, Madagascar	3.28	0.00	25.29	2.19	1.15
1062	Jakob (1932) No. 65	2-layer (mod. scattering)	Ampandrandara, Madagascar	2.96	1.18	23.40	1.69	0.68
1063	Jakob (1932) No. 64	1-layer	Mandridano, Madag.	2.09	0.00	24.48	1.64	0.08
1064	Jakob (1932) No. 63	2-layer	Saharakara, Madag.	2.79	1.68	23.78	1.11	0.56
1065	Jakob (1932) No. 62	1-layer	Ambatoaba, Madag.	1.42	0.66	24.80	0.86	0.86
1066	Jakob (1928) No. 19	1-layer (heavy scattering)	Simplon-Tunnel	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 met. carbonate rock, S. W. Africa	2.72	0.97	25.05	0.66	0.58
1069	Jakob (1928) No. 23	1-layer	Vesuvius	0.58	1.92	28.18	1.27	—
1071	Jakob-not published	1-layer	Skräbböle, Pargas, Finland	0.71	1.01	27.80	0.12	2.12
1072	Jakob-not published	1-layer	Pargas, Finland	5.59	1.54	22.00	1.41	2.80
1073	Jakob-not published	1-layer	Patteby, Pargas, Finland	1.47	1.29	26.16	0.33	1.87

* Reported in Jakob and Parga-Pondal (1932)

TABLE XLIV (CONT.)

Number	Reference	Structure	Paragenesis	FeO	Fe2O3	MgO	TiO2	F
1074	Jakob-not published	1-layer	Ontala, Pargas, Finland	1.68	1.87	25.91	0.68	1.30
1075	Jakob-not published	1-layer	Skräbböle, Pargas, Finland	0.59	1.75	27.22	0.10	0.48
949	Dana (1892) Anal. 12 p. 633	1-layer	Rossie, N. Y.	7.62	1.12	21.47	1.16	4.00
1135	Pagliani (1940)	1-layer	in crystalline limestone	1.55	2.65	27.62	2.83	
1139	Hutton and Seeyle (1947)	1-layer	from marble, New Zealand	2.38	0.43	22.95	0.82	0.62
1252	Pieruccini (1950)	1-layer	pneumatalytic, Mt. Somma	7.89	Tr.	15.66	0.33	2.57
1261	(a) Mauguin (1928B) / (b and c) Mauguin (1928B)	1-layer / 3-layer hexagonal	Ambotoaba, Madagascar	2.30		24.42	0.74	
730	Prider (1940)	1-layer	leucite lamproite, West Australia	3.75	2.18	19.66	8.97	0.66
1325	Cross (1897)	1-layer	in Wyomingite	0.90	2.73	22.40	2.09	1.03

TABLE XLIV (CONCL.)

ANALYZED MANGANOPHYLLITES

Number	Reference	Structure	Paragenesis	FeO	Fe₂O₃	MgO	MnO	Mn₂O₃	TiO₂
1076	Jakob (1925, No.8)	1-layer	contact met.	0.00	16.94	26.79	4.52	0.00	0.00
1077	Jakob (1925, No.7)	1-layer	contact met.	2.54	0.00	29.22	Tr.	Tr.	0.00
1078	Jakob (1925, No.6)	1-layer	contact met.	0.00	3.95	22.60	0.00	8.30	Tr.
1079	Jakob (1925, No.5)	1-layer	Varmland, Sweden	0.00	4.68	21.18	9.25	2.96	0.00
1080	Jakob (1925, No.4)	1-layer	contact met.	0.00	2.97	24.80	0.00	5.77	0.55
1081	Jakob (1925, No.3)	1-layer	contact met.	0.00	2.68	26.65	0.00	4.07	0.41
1082	Jakob (1925, No.2)	1-layer	contact met.	0.00	0.91	29.28	0.00	3.92	0.00
1083	Jakob (1925, No.1)	1-layer	Varmland, Sweden	0.00	2.81	27.87	0.00	4.93	0.22

D. Inclusions

Much phlogopite contains microscopic needles of rutile as inclusions oriented along the principal crystallographic directions. These inclusions are the cause of the well known asterism of phlogopite. They appear to have formed as the result of exsolution similar to much of the oriented hematite in muscovite. The distribution of the inclusions is not always uniform, but may be zonal. Zones with abundant inclusions may alternate with inclusion-free zones.

E. Zoning

Phlogopites show both color and inclusion zoning commonly. Some examples include the following.

Orcel (1926) described a peculiar green phlogopite from Snake Creek, Utah, that displays oscillatory color zoning. A specimen in our mica collection shows alternating light- and dark-green zones. The dominant portion of the irregular crystal is a light yellowish-green with fine darker-green bands parallel with the prism and clinopinacoid. There are minor reeves normal to the color bands. Optical properties are as follows.

Orcel	This Report
n_p = 1.5529	α = 1.552
	β = 1.592
n_g = 1.5910	γ = 1.593
uniaxial	2V = 2-3°

Both zones have essentially the same optical properties.

A spectrochemical analysis of a specimen of the light-green material in our collections from the Snake Creek locality is presented in Table XLV.

Larsen (1941) reports color-zoned phlogopite of hydrothermal origin associated with aegirine and sodic amphibole in contact-metamorphosed limestone at Iron Hill, Colorado. The micas vary moderately in refractive indices. They are strongly zoned with a pale-brown, or commonly a green core, and one or more outer zones that may be lighter or darker than the core. Some of the outer zones have pleochroism which is the reverse of that normal in phlogopite. Larsen gives the following optical data:

TABLE XLV

SPECTROCHEMICAL ANALYSIS* OF LIGHT-GREEN PHLOGOPITE FROM SNAKE CREEK, UTAH

Oxide	No. 712
SiO_2	40.
Al_2O_3	12.
K_2O	12.
Fe_2O_3	2.3
MgO	29.
MnO	.13
TiO_2	.07
CaO	.08
SrO	--
BaO	.08
Na_2O	1.05
Li_2O	--
Rb_2O	--
Cs_2O	--
Ga_2O_3	.009
Sc_2O_3	--
Co_2O_3	.004
Cr_2O_3	.015
SnO_2	--
V_2O_5	--
F	.35

* Spectrochemical analysis by
  C. E. Harvey

TABLE XLVI

SPECTROCHEMICAL ANALYSIS* OF ZONED PHLOGOPITE, PERTH, ONTARIO

	1131a - Core	1131b - Rim
SiO_2	41.	40.
Al_2O_3	12.5	11.5
K_2O	11.	11.5
Fe_2O_3	3.6	3.1
MgO	26.	27.5
MnO	.06	.05
TiO_2	.85	.85
CaO	.07	.11
SrO	.006	.004
BaO	.46	.55
Na_2O	.75	.70
Li_2O	--	--
Rb_2O	--	--
Cs_2O	--	--
Ga_2O_3	.004	.004
Sc_2O_3	--	--
Co_2O_3	.0008	.0009
Cr_2O_3	.0027	.0026
SnO_2	--	--
V_2O_5	.017	.014
F	2.7	3.7

* Spectrochemical analysis by
  C. E. Harvey

Specimen IH - 2

core: α = 1.576 β = 1.613 γ = 1.613
   pleochroism
      α nearly colorless, β and γ faint brown

intermediate zone: α = 1.576 β = 1.613 γ = 1.614
   pleochroism
      α deep chestnut brown, β and γ nearly colorless

outer zone:
   pleochroism
      α deep chestnut brown, β and γ nearly colorless

Generally, the indices of refraction are the same for the different zones, but the axial angle may be a little larger in the outer zones.

A phlogopite (1369) from Thorne Township, Quebec, Canada, shows four concentric color zones. The specimen varies in color from a deep red-brown at the core to a light green-brown at the margin. The boundaries between zones become sharper toward the core. Optical data for each zone are as follows.

Zone	α	β	γ	2V	Color
1 (core)	1.575	1.622	1.622	1°- 2°	deep red-brown with many rutile inclusions
2	1.575	1.622	1.622	1°- 2°	deep red-brown with no inclusions
3	1.568	1.611	1.611	0°	red-brown, inclusion free
4 (margin)	1.559	1.599	1.599	0°	green-brown

From margin to core, each zone shows a progressive increase in indices and size of the optic angle.

Another phlogopite (1370), an irregularly shaped sheet from an unknown Canadian locality has a parallel arrangement of light-brown inclusion-free zones alternating with darker-brown zones containing numerous inclusions of rutile. Both zones are essentially uniaxial. The inclusion-free zones show no asterism and yield a sharp figure, whereas the zones bearing inclusions show strong asterism and yield an extremely diffuse figure. Indices of refraction for each zone are:

	α	$\beta = \gamma$
Clear zones	1.541	1.570
Zones bearing inclusions	1.544	1.578

Two analyses were made of a zoned, amber-colored phlogopite (1131) from Perth, Ontario, one from the core of the sheet, the other from the rim (Table XLVI). There is a slight difference in Fe, none in Ti, a distinct difference in F.

Numerous specimens of Canadian phlogopite available to us display color zones. Generally, the optical properties of the different zones in a single crystal are essentially the same. Wherever a variation was noted, the darker-colored zones or zones of inclusion have higher indices of refraction and a larger optic angle. More specimens of phlogopite tend to be darker toward the center and lighter toward the rim than the converse.

II. OCCURRENCE

A. Nonpegmatitic

Phlogopite or phlogopitic biotite occurs in ultramafic igneous rocks, particularly kimberlites. It also forms as a secondary constituent in many other types of peridotites, probably largely through autometasomatism or hydrothermal metamorphism. Phlogopite also is a primary constituent of certain lamprophyres, mafic and ultramafic potassic rocks, and a few rare extrusive types.

In metamorphic rocks phlogopite is formed in marbles or Mg-silicate gneisses by the regional metamorphism or contact metamorphism of argillaceous dolomites, or impure magnesian limestones. If the phlogopite contains abundant F, metasomatism is generally indicated as a means of introducing that element. However, not all phlogopites contain fluorine and the mica may form without additions to the system, if sufficient water is available. Phlogopite marbles are considered to belong to the amphibolite facies. Phlogopites occur also in various skarns in whose formation metasomatism has been important.

B. Pegmatitic and Vein Occurrences

Phlogopite deposits occur as veins consisting chiefly of phlogopite, apatite and calcite in pegmatite and pyroxenite in southern Quebec and eastern Ontario. Scapolite, actinolite, diopside and many rarer minerals occur as accessory constituents. The veins and concentrations occur in or adjacent to masses of pyroxenite that have been intruded by bodies of granitic, microcline-rich pegmatite. Landes (1938) believes that the deposits were formed from the hydrothermal phases of granitic pegmatites markedly contaminated at depth by dolomitic limestone.

Analagous deposits occur in Madagascar in pyroxenite but here apatite is not abundant (Savornin, 1934).

Other vein deposits of phlogopite, calcite, apatite, pyroxenite, and diopside transect impure marbles, amphibole schists, and amphibolites in the Sejudjanka region of the USSR where these rocks have been intruded by pegmatites and granite dikes (Ginsburg, 1930).

III. MANGANOPHYLLITE

A. Introduction

Manganophyllite was first described by Igelstrom (1872), his specimen having come from the iron and manganese mines at Pajsberg near Filipstadt in Sweden: "The manganophyllite is accordingly the manganese-rich mica. The one known up to now is placed near phlogopite or biotite. Perhaps one can take it for a manganese-rich biotite." Dana (1892) states: "In composition manganophyllite is a manganesian biotite, but varying widely in the relative amounts of manganese and other elements."

B. Structure

The only X-ray work reported in the literature on manganophyllite has been that of Hendricks and Jefferson (1939). They report a manganophyllite from Pajsberg, Sweden with a 2-layer monoclinic structure. We have found that all the Swedish manganophyllites described by Jakob (1925A) have the 1-layer monoclinic structure. A specimen from Franklin, N. J. (No. 1358) has a 2-layer structure, however. The Swedish manganophyllites that have a very small 2V (particularly Jakob, 1925A) No. 1 and 5) appear to approach the 3-layer hexagonal structure owing to an intimate "twinning" of thin sheets similar to that described for uniaxial lepidolites. The chemical analyses of these micas show no correlation between composition and polymorphism. Manganophyllites with the 1-layer form show extreme ranges in the Fe and Mn (for example, Fe_2O_3 = 0.00 to 16.94 percent; see also Table XLIV). Jakob (1925A) reports most of the manganese in these micas as Mn_2O_3.

C. Composition

The general formula for manganophyllite can be written as follows:

$$K_2(Mg_{5-4},Mn_{2-1},Fe^3_{0-0.5},Fe^3_{0-1}Mn^3_{0-1})(Si_6Al_2)O_{20}(OH)_4$$
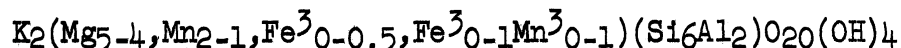
with a total of 6 atoms in octahedral positions.

Other possible isomorphous substitutions are minor Na, Ca, and Ba for K; and minor Ti for Si in tetrahedral positions or more probably in octahedral positions for Mg, Mn, or Fe^3.

In biotites small amounts of Mn are associated either with high ferric or ferrous iron. Some manganophyllites contain both Mn^2 and Mn^3,

others have only Mn^2, and in a few the manganese is chiefly divalent. Manganophyllites are low in iron and have a maximum of about 18 percent MnO plus Mn_2O_3. Phlogopites on the other hand have about a maximum of 30 percent MgO and less than 10 percent Fe and Fe_2O_3 combined.

Some investigators class manganophyllite as a variety of biotite, but owing to the small amounts of Fe^2 and large quantities of Mg, most manganophyllite should be classed as manganian phlogopite.

D. Optics

$$\alpha = 1.548 - 1.573 \qquad \gamma - \alpha = 0.024 - 0.040$$
$$\beta = 1.581 - 1.636 \qquad 2V = 0° - 34°$$
$$\gamma = 1.581 - 1.636$$

There appears to be no generalization which accurately represents the relationship between indices and other optical properties with composition. For example, Jakob's (1925A) analysis No. 8 with 16.94 percent Fe_2O_3 has the highest indices of any of his manganophyllites. However, Jakob's analysis No. 6 has indices approximately 0.03 lower and contains only 3.95 percent Fe_2O_3.

The γ indices of a few manganophyllites were plotted against 1/2 (Mn_2O_3 or MnO) + 2(Fe_2O_3 + TiO_2) by Heinrich (1946). The results indicated that the effect of the manganese on the index of refraction is less than that of the ferrous iron.

The most significant observation concerning the optical properties of the manganophyllites studied is that several of Jakob's (1925A) specimens (Numbers 3, 4, and 6) showed the optic plane perpendicular to (010). Practically 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.

E. Single Crystal Variation

A survey of the literature failed to uncover any information regarding structural or optical variations or color zoning in single crystals of manganophyllites. In our work we have found no cases of polymorphic variations or color zoning within single crystals of manganophyllites. Small-scale variations in the size of 2V within individual crystals have been observed, but are generally insignificant.

F. Occurrence

     In Table XLVII are presented a listing of all the occurrences of manganophyllite (including doubtful or incompletely described manganophyllites) we have been able to find in the literature.

G. Conclusions

1. Manganophyllite is a magnesium-manganese mica, with variable amounts of ferric and ferrous iron crystallizing, with 1-layer and 2-layer monoclinic structures.

2. Most manganophyllites could be described equally well as manganian phlogopites.

3. Many reported manganophyllites are of doubtful authenticity and need further study, particularly chemical analyses.

4. Most manganophyllites of undoubted authenticity occur in manganese deposits of contact metamorphic origin.

TABLE XLVII

OCCURRENCE OF MANGANOPHYLLITE

Location	Type Deposit	Host Rock	Assoc. Mn Minerals	Other Minerals	Reference	Remarks
Ada Co., Idaho	disseminated	pegmatite	-----	quartz, orthoclase	Kauffman, et al. (1950)	Probably a biotite; spectro-analysis given
Mason Co, Washington	lenses, pockets, and thin beds	volcanics, limestones, and argillites	bementite, neotocite and other Mn silicates and minerals	calcite, jasper	Park (1942) Pardee, et al. (1921)	
Kaso Mine, Japan	veins of basic composition	quartzite and slate	rhodonite, tephroite	Mn-bearing biotite, actinolite and other silicates and Ba-feldspar	Yoshimura (1939)	Called mangan phlogopite; chemical analysis given
Franklin, N. J.	disseminated	skarn	many Mn-bearing silicates	Zn and Fe minerals	Chester (1894) Palache (1935)	
Langban, Sweden	disseminated	skarn	many Mn-bearing silicates	Fe ores	Palache (1929) Jakob (1925A)	
Harstigen, Pajsberg and Jacobsberg, Sweden					For chem. anal. see: Jakob (1925A) Dana (1892)	Occurrence similar to that of Langban
Ankara, Turkey		sedimentary rocks	rhodonite, rhodochrosite, braunite		Wijkerslooth (1943)	Need further study
Central India	disseminated	mica schists	in Mn deposits		Fermor (1909)	Incompletely described
Postmasburg, South Africa		sedimentary rocks	psilomelane, pyrolusite, rhodonite		Hall (1926) Chudaba (1929)	Need further study

PART V. BIOTITE

I. MINERALOGY

A. Composition

$$K_2(Fe^2Mg)_{6-4}(Fe^3,Al,Ti)_{0-2}(Si_{6-5}Al_{2-3})O_{20-22}(OH,F)_{4-2};$$
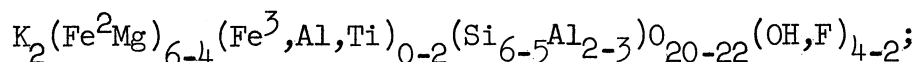
some Na,Ca,Ba,Rb, and Cs for K; Mn for Fe^2. Mg may be almost absent in some
varieties; others contain much Fe^3; in some Li may replace Al possibly leading
to zinnwaldite. Zambonini (1919) reports 14.33 percent CaO (see p. 42).
Freudenberg (1919) reports a biotite with 12.5 percent TiO_2. It has been
pointed out that a complete series exists between biotite and phlogopite; a
suggested division is discussed under phlogopite chemistry. For a discussion
of the role of titanium in biotite (and micas) and the variation of tetra-
hedrally coordinated Al/Si, the reader is referred to the section on the rela-
tion of composition and occurrence (p. 195).

B. Structure

1. Introduction. Hendricks and Jefferson (1939) report biotite
as occurring in five polymorphs:

1-layer monoclinic - space group Cm

$a = 5.3Å$ $b = 9.2Å$ $c_0 = 10.2Å$ $\beta = 100°$

2-layer monoclinic (octophyllite type) - space group C2/c

$a = 5.3Å$ $b = 9.2Å$ $c_0 = 20.2Å$ $\beta = 95°$

3-layer rhombohedral enantiomorphic hemihedral - space group
$C3_112$ or $C3_212$

$a = 5.3Å$ $c_0 = 30.0Å$ $\beta = 90°$

6-layer triclinic holohedral - space group $P\bar{1}$

$a = 5.3Å$ $b = 5.3Å$ $c = 60.0Å$

$\alpha = 90°$ $\beta = 90°$ $\gamma = 120°$

24-layer triclinic holohedral - space group $P\bar{1}$

$$a = 5.3\text{Å} \qquad b = 5.3\text{Å} \qquad c = 240.0\text{Å}$$

$$\alpha = 90° \qquad \beta = 90° \qquad \gamma = 120°$$

The only other polymorph reported in the literature is an incompletely described 18-layer triclinic biotite (Amelinckx, 1952B).

2. X-ray Technique. The X-ray study of biotites, and to a lesser extent of phologopites, is more difficult than that of muscovites and lepidolites. A crystal orientation procedure based on optical methods was perfected and used satisfactorily with the latter series. The positions of the crystallographic axes were determined by means of interference figures and extinction positions, for in muscovite and lepidolite the crystallographic axes correspond essentially to extinction positions. In phlogopite and biotite, however, the birefringence of (001) sections in often nil and 2V approaches zero; thus an optical orientation method is impractical.

To overcome these problems, Laue photographs were used to determine the positions of the desired axes. However, Laue photographs of some monoclinic specimens appear so nearly hexagonal that the true plane of symmetry is not easily determined (owing to the large crystal-to-film distance of our camera, few reflections were recorded). Weissenberg photographs about the two pseudo a-axes were often taken before the one about the true a-axis was obtained. An alternative method was used occasionally, whereby it was not necessary to take a Laue photograph. This consisted of producing a precussion figure on the cleavage flakes and choosing one of three rays at random as the rotation axis, with the hope that it would be the true a-axis and not one of the pseudo a-axes. But, as in the first method, several attempts usually were required before the structure could be determined accurately.

Zero-level Weissenberg photographs taken about one of the pseudo a-axes of the 2-layer monoclinic polymorph appear to be identical with the 0-level a-axis photograph of the 6-layer triclinic biotite illustrated by Hendricks and Jefferson (1939, p. 745). Likewise, 0-level pseudo a-axis photographs of the 1-layer monoclinic polymorph strongly resemble 0-level b-axis photographs of the 6-layer monoclinic polymorph reported only in lepidolites. Therefore extreme caution had to be practiced in deciding the structure of these micas from 0-level photographs. Diffuse scattering, particularly among the biotites, also made structure determinations difficult in several instances.

In some specimens the crystal development of the flakes was poor, or the only analyzed specimens available were powders. Under these conditions, single-crystal methods were not possible, and the powder X-ray method was

attempted. It was found that iron radiation was most suitable and even essential in studying biotites by the powder method. A filter was not used, for it increased the necessary exposure time by a factor of nearly three. Unfortunately the powder patterns of the common polymorphic forms are so similar that no distinctive patterns could be obtained. A monochromator, for longer exposures without background fogging, was not available.

3. New Structural Data. Although the structures of about 200 biotites have been determined, only a few definite conclusions are available. Attempts to correlate chemistry with structure have been unsuccessful, but information is available on the geographic and geologic distribution of various polymorphs. The reader is referred to Table XLIV in which the results of the X-ray study of 27 analyzed biotites indicates no apparent relationship between chemistry and polymorphism in the biotites (and phlogopites).

In the study of the biotites, the structures of several uncommon varieties were determined. The results are tabulated in Table XLVIII.

During this study neither the 6-layer nor the 24-layer triclinic biotites described by Hendricks and Jefferson (1939) were found. Our studies of the supposed type 6-layer biotite described by Hendricks and Jefferson (1939) from Sterling, New York (U.S.N.M. C3675) indicates the 1-layer monoclinic structure. Dr. H. S. Yoder (personal communication) has studied other material from the same specimen and has found the 2-layer monoclinic structure. Only the 1- and 2-layer monoclinic and the 3-layer hexagonal polymorphs were definitely established. In a few scattered instances patterns not assignable to any known polymorphs were recorded on Weissenberg photographs. Detailed study of these specimens will probably show that they consist of crystals with mixed structures rather than new polymorphs.

4. Variation of Structure in Pegmatite Districts. About 80 biotite specimens from about 15 pegmatite deposits in the southeastern United States were studied. A compilation of the results showed that about 50 have crystallized as the 2-layer monoclinic polymorph, 15 as the 3-layer hexagonal polymorph, but only 1 as the 1-layer monoclinic polymorph. Heavy diffuse scattering prevented accurate structural determination of the remainder. Diffuse scattering is characteristic of biotites from this area. The results of studies from other widely scattered pegmatite districts are summarized below:

Southern Norway - predominantly 1-layer monoclinic forms, but 2-layer forms abundant.

Canada: Bancroft, and Wilberforce Districts - predominantly 1-layer forms.

Colorado: Guffey District - predominately 1-layer forms.

TABLE XLVIII

STRUCTURE OF BIOTITE VARIETIES

Number	Name	Locality	Structure
732(a)	waddoite	Isle of Waddo	1-layer monoclinic
746(a)	lepidomelane (pterolite)	Brevig, Norway	2-layer monoclinic
752(a)	meroxene	Mt. Vesuvius	1-layer monoclinic
753(a)	meroxene (white)	Mt. Vesuvius	3-layer hexagonal
759(a)	meroxene	Mendham, N. J.	1-layer monoclinic
754(a)	calciobiotite	Italy	1-layer monoclinic
770(a)	annite	Rockport, Mass.	1-layer monoclinic
1116(a)	annite	Rockport, Mass.	1-layer monoclinic
775(a)	siderophyllite	Pikes Peak, Colo.	1-layer monoclinic
1117(a)	siderophyllite	Brooks Mt., Alaska	1-layer monoclinic (very diffuse scattering)
783(a)	cryophyllite	Rockport, Mass.	1-layer monoclinic
1112(a)	monrepite	Finland	1-layer monoclinic
1113(a)	eukamptite	Presburg, Hungary	1-layer monoclinic
1114(a)	bastonite	Bastogne, Belgium	2-layer monoclinic
1130(a)	haughtonite	Scotland	(?) - mixed structures

Geological environment may play an important role in the crystallization of biotite micas. The pegmatites of the southeastern United States (North Carolina, particularly) are of quartz diortic composition, whereas those of the Bancroft area are nepheline syenitic. It seems possible that biotites crystallizing from such different magmas might have structures characteristic of their particular environments, but this hypothesis needs considerable further study. As mentioned above, composition alone does not seem to be the exclusive factor. It was hoped that a knowledge of the temperature of crystallization of these micas would be very 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.

C. Optical Properties

1. Indices, 2V, Optic Plane; Variation with Composition.
The refraction indices of biotite are:

$$\alpha = 1.565 - 1.625 \qquad \gamma - \alpha = 0.040 - 0.060$$
$$\beta = 1.605 - 1.675 \qquad 2V = 0° - 25°$$
$$\gamma = 1.605 - 1.675 \qquad \text{(negative)}$$

2V is usually less than 10°, but a few rare varieties have 2V somewhat greater than 25°.

Dispersion: $r > v$ or $r < v$
$a \wedge c = 8° - -2°$, $\beta = b$, $\gamma \wedge a = 0 - 9°$.

The optic plane is usually parallel to (010), less commonly (only in 2-layer polymorphs) normal to (010). The 1-layer polymorph is the most common structure and invariably has its optic plane parallel to (010). The indices increase with Fe^2 and more rapidly with Fe^3 and Ti. For example, see the studies of Winchell and Winchell (1951, p. 374, Fig. 257) and that of Heinrich (1946), in which it was demonstrated that the effect of Fe^3 and Ti in increasing the indices of refraction is approximately twice that of Fe^2 and that the effect of Mn^3 was approximately one-half that of Fe^2. In biotites in which total iron remains constant, 1 percent TiO_2 increases refractive indices 0.0046 (Hall 1941). In some rare ferrian biotites γ may reach as high as 1.73 and $\gamma - \alpha = 0.080$.

2. Color; Variation with Composition.
Biotites are deeply colored, usually in shades of brown and green. Red-brown biotites owe their color mainly to combined titanium and ferric iron; greenish-brown

and brown biotites are colored mainly by Fe^2 with lower amounts of Ti and Fe^3; and blue-green biotites are colored mainly by Fe^2 (Hall, 1941 and Shibata, 1952A).

3. **Zoning.** Zoning in biotite has not been as widely recognized as in muscovite. This may be due to the much darker color of most biotites in which slight color differences would be hidden except in very thin sheets. Some examples of zoned biotites reported in the literature are listed.

Brögger (1921) reports optically zoned biotite from the Fen Region of southern Norway, in damkjernite, a porphyritic rock containing phenocrysts of biotite and pyroxene in a fine-grained groundmass of pyroxene, biotite, and magnetite with minor nepheline, microcline, and calcite. In hand specimen the biotite is bronze-brown, but under the microscope it is reddish in color. The biotite flakes are not six-sided, but usually irregular. Pleochroism of the core is from ruby-red or orange-red to bright yellow; pleochroism of the rim is from bright green to bright yellow or colorless. The biotite seems to be uniaxial.

Johannsen (1937) notes the occurrence of color-zoned biotite in minette, a lamprophyre of the syenite series, composed of bibtite phenocrysts in an orthoclase groundmass. The biotite is dark brown under the microscope and in both the phenocrysts and the groundmass it shows "a light center surrounded by an iron-rich opaque border."

Grout (1924) mentions that biotite in the Minnesota granites commonly shows a zonal structure.

Clarke (1887) describes a bronzy-black mica (siderophyllite), from Pikes Peak, Colorado, "...resembling phlogopite externally, ... but made up of a core composed of a soft rotten material, evidentally derived from the original mica, and surrounded by a broad black margin of the latter. Streaks of rusty alteration products reached into the margin in every direction." Analyses for both zones are given as follows.

	Margin	Center
H_2O	4.54	7.28
SiO_2	34.21	34.63
Al_2O_3	16.53	17.95
Fe_2O_3	20.12	31.25
FeO	14.17	3.01
MnO	0.91	0.34
CaO	0.48	0.81
MgO	1.34	1.08
Na_2O	1.43	0.89
K_2O	6.50	1.96
F	0.08	0.54

Coats and Fahey (1944) describe an optically zoned siderophyllite from an alaskite pegmatite near Brooks Mountain, Alaska. In thin section some of the plates show a core having a double refraction of 0.050 and 2V = 6° - 8° surrounded by a discontinuous rim of pale-blue mica having a double refraction of 0.028 and 2V = 37°.

Over 100 specimens of biotite were examined for zoning. None of the pegmatitic biotite showed any zoning. One thin section of minette from Freiberg, Saxony, contained phenocrysts of zoned biotite as described by Johannsen (1937). Apparently zoning in biotite is very uncommon.

II. OCCURRENCE

A. Igneous Rocks

Biotite occurs as an essential or accessory mineral of most types of intrusive igneous rocks, particularly pegmatites, aplites, granites, granodiorites, syenites, feldspathoidal syenites, monzonites, diorites and norites; less commonly in diabases, other gabbroic types, and in rarer types. In many of these it appears as a primary constituent, although in some, such as diorites and gabbros for example, it has apparently formed by deuteric reactions and can be seen as replacements of and overgrowths on such minerals as hornblende, augite or magnetite.

Biotite, as phenocrysts, also is widespread in felsic and intermediate extrusive rocks, and may show strong resorption mainly to iron oxide and pyroxene. Biotite is rare, however, as a matrix mineral in these rocks.

B. Metamorphic Rocks

Biotite is a widespread mineral of metamorphic rocks and is stable under a broad temperature-pressure range, forming apparently at relatively low temperatures and remaining stable into the granulite facies. In contact metamorphism it forms under low grade conditions in metamorphosed argillaceous, impure arenaceous, as well as in intermediate and acid igneous rocks and remains stable into the higher intensity environments.

The biotite zone is indicative of moderate-grade regional metamorphism in argillaceous rocks, but, if stress is abnormally low, biotite may form even at somewhat lower temperatures. The reaction between chlorite and muscovite yields biotite for such rocks as biotite schists, biotite-sericite schists, biotite-chlorite schists and albite-biotite schists. Once formed, biotite usually persists into higher-grade zones, appearing with garnet, kyanite, sillimanite, and cordierite, in schists, gneisses and granulites. More mafic igneous rocks and somewhat calcareous sediments also yield biotite-bearing types upon regional metamorphism -- biotite-plagioclase schists, biotite-hornblende schists, and biotite-epidote schists, for example.

C. Variation of Composition with Occurrence

Heinrich (1946) has shown that biotites of intrusive igneous rocks vary systematically in composition with the rock in which they were formed. This variation is chiefly an increase in the Fe^2/Mg ratio toward the more felsic rocks. Biotites rich in Fe^3 are strongly represented in alkalic rocks,

and also in extrusive igneous rocks where some of their Fe^2 was oxidized to Fe^3 by escaping gases. Others have traced the successive changes in individual elements in biotites. Chapman and Williams (1935), for example, in their study of the alkaline White Mountain magma series, have observed that in going from a gabbro to granite, the biotite will show a decrease in Si,Ti,Al, and particularly Mg with a corresponding substantial increase in Fe. Similar results are reported by Inoue (1950) on biotites from nepheline syenites and nepheline syenite pegmatite. She also notes that biotites from nepheline syenites tend to be higher in Mn. Nockolds and Mitchell (1948) in a comprehensive study of the non-alkaline Garabal Hill - Glenn Fyne complex show a decrease in Si,Ti, and Mg with an increase in Al,Fe^3, and Fe^2 in the biotites on progressing from pyroxene-mica diorite to porphyritic granodiorite. In their study of the trace elements they find a progressive increase in Ga,Mn, and Rb on passing from early to late biotite whereas Cr,V,Cu,Ni,Co, and Sr decrease in the same direction. Ba remains more or less constant in early biotites and decreases in the late stages; Li, on the other hand, increases rapidly in the early biotites and later remains more or less constant.

The role of Ti in the crystal lattice of the micas is discussed by Serdiuchenko (1948B). He concludes that at low temperatures and in an acid medium, Ti proxies for Mg, whereas at high temperatures and with an increase in the alkalinity of the medium, Ti may substitute for Si. Thus, in alkaline igneous rocks Ti may be found in the tetrahedral layer or may enter both the tetrahedral and octahedral layers. In nonalkaline igneous rocks Ti is exclusively in the octahedral layers and isomorphously replaces Mg. Biotites of feldspathic quartz-free rocks (mainly lamprophyres) occupy intermediate positions with relation to Ti (but closer to the position of Ti in alkaline rocks).

The presence and role of Ti^3 in phlogopite and biotite is recorded and discussed by Jakob and Parga-Pondal (1932) and Jakob (1937). However, they maintain that Ti is not found in the tetrahedral positions.

Most workers believe that titanium replaces magnesium. Several analyses, however, are recorded in the literature in which some of the Ti must be grouped with silicon and tetrahedral aluminum in calculating the formulas so that all tetrahedral positions are filled. Such a case is well illustrated in the study of a titaniferous phlogopite described by Prider (1940); Serdiuchenko (1948B) gives about 10 other examples of biotites and titanian biotites (wodanites). Serdiuchenko (1948B) also finds it necessary to include small amounts of Fe^3 to fill tetrahedral positions in the case of three lepidomelanes. In this manner the atomic requirements for the tetrahedral layer are completed with Ti (and Fe^3), since the total of aluminum and silicon is insufficient. Nockolds and Mitchell (1948) also discuss this problem and point out that practically all the aluminum in the biotites they studied (from diorites, granodiorites and granites) is tied up in tetrahedral positions.

Serdiuchenko (1948B) (in translation) quotes Fersman as reporting:

"Ti replaces Si in tetrahedra of the pyroxenes, amphi-
boles (chains) and micas (sheets) but not in feldspars or
zeolites (3 dimensional linkage). However, the crystallo-
chemical role of titanium in the lattice of minerals is not
determined only by the type of structure, but for the very
same minerals (for example micas, amphiboles and others) its
role changes with respect to the thermodynamic and geochemical
conditions of the genesis (temperature, pressure, oxidation potential,
pH)."

Machatschki (1930) states that at low temperatures Ti may not re-
place Si because of the differences in ionic radii of titanium and silicon
atoms. At high temperatures, however, it may replace Si, but the tendency
for exsolution of ilmenite or other titanium minerals appears to be great.
Bragg (1937) points out that Ti is exclusively found in sixfold coordination
and therefore it is very unlikely that in the micas it would replace silicon
in fourfold coordination. Ramberg (1948) believes that Ti is relatively
abundant in high-temperature metamorphic biotites and accounts for their
stability.

In our opinion the problem still remains unsolved. The accuracy of
the conclusions reached by Serdiuchenko (1948B) depends upon (1) whether it is
absolutely necessary for the tetrahedral positions to be filled (the octa-
hedral may be deficient), and (2) the accuracy of the analyses used in the
compilation..

In metamorphic rocks the data are still too few to permit much
correlation between composition and the temperature-pressure environment.
Undoubtedly, however, metamorphic biotites vary considerably in composition.

It has been found in a review of the literature that biotites formed
under conditions of simple thermal metamorphism generally have a much larger
quantity of Al in tetrahedral position than biotites formed under conditions
of regional metamorphism (Dr. A. Miyashiro, personal communication). This
fact appears to be in accord with the general tendency of minerals that formed
at high temperatures to contain more fourfold Al than those that formed at low
temperatures.

Harry (1950) has recently demonstrated that this is the case for some
igneous and metamorphic minerals, particularly amphiboles and pyroxenes, and
believes that the situation is analogous in biotites. He suggests that large
amounts of tetrahedrally coordinated Al may help stabilize biotite that formed
under physical conditions (high temperature) ordinarily promoting the

crystallization of simpler lattice-type minerals. This suggestion is based upon the observation that biotite has the largest amount of Al substitution for Si in a rock also containing pyroxene and hornblende in which all three minerals crystallized under the same pressure-temperature conditions.

III. PEGMATITIC BIOTITE

A. Localization

The occurrences of pegmatitic biotite may be grouped under

1. Wall-zone occurrences
2. Outer intermediate-zone occurrences
3. Fracture-fillings and fracture-controlled replacements
4. Exomorphic and endomorphic biotite.

Biotite of magmatic origin occurs normally in the outer zones of pegmatites. In wall zones the biotite may occur in elongate blades or plates that tend to be arranged normal to the wall-rock contacts. Not uncommonly these blades are "tangle foot", i.e., they contain a central strip of biotite and bordering parallel overgrowths of muscovite. The School Section pegmatite of Eight Mile Park, Colorado, contains such blades abundantly in the hanging-wall section of the wall zone. Many pegmatites of the southeastern districts contain unoriented books and plates of biotites in wall zones.

Smaller books, flakes and scales of biotite also are abundant in outer intermediate zones in many districts of New England and other regions. In such zones the biotite occurs with quartz, microcline, sodic plagioclase and, in some cases, muscovite as well.

Fracture-fillings of biotite are conspicuous features in some pegmatites, for example, those of Topsham, Maine; in the Big Flint pegmatite, Jackson County, North Carolina; in the Yard pegmatite, near Salida, Colorado, and in many pegmatites of other districts, especially some in Southern Norway and Sweden. The biotite crystals are commonly of large size, 1 to 20 feet, and usually appear in subparallel to slightly radial patterns. Dendritic arrangements also occur with a central large sheet as a stalk and smaller sheets as branches. In some districts with biotite of this type, rare-earth minerals, such as fergusonite, gadolinite, samarskite, etc., occur relatively abundantly, as crystals and grains between the biotite sheets. In a few deposits small books of muscovite have been formed between the larger parallel biotite sheets, normal to them.

The outer parts of some pegmatites contain wisps and schlieren of recrystallized coarse-grained parallel biotite derived by incorporation of blocks of biotite-rich wall rocks. Examples are the Ledford Cove pegmatite,

Franklin, North Carolina and the Big Ridge pegmatite near Waynesville, North Carolina. Most pegmatites that have been intruded into hornblende-rich rocks have developed prominent selvages of coarse biotite by reaction.

In a few pegmatites more than one generation of biotite has been observed, e.g., wall-zone plus fracture-filling biotites, but there is no information as to any compositional variation between the two types. It has been observed that pegmatites containing concentrations of green book muscovites in wall zones rarely contain any biotite, except some that may be exomorphic in origin. Biotite associated with or included in green book muscovite usually itself is deep olive green in color, whereas that intergrown or occurring with ruby muscovites is black or dark brown, probably with more Fe^3 and Ti and less Fe^2. Where abundant biotite occurs with muscovite in zones, the muscovite normally is largely free of iron-oxide inclusions.

In some districts biotite and schorl are mutually exclusive, either in wall zones or in the entire pegmatite.

B. Macrostructural Features

A systematic study of the structural features of book or sheet biotite has not been attempted. Biotite commonly forms in thin sheets or flakes from a fraction of an inch to many feet across. Thick books are relatively uncommon. Ruling marks thicker books in some deposits. Many sheets that crystallized along cracks follow the curvature of the fractures. Some very large sheets of this type show a diamond-shaped crosshatch pattern of markings resembling reeves but less closely spaced. Intergrowths with muscovite are common, normally with the biotite occupying the central part of the composite book or sheet. However, an overgrowth of central muscovite with framing biotite also is known. Concentric structure in biotite has been reported by Simpson (1912) from a pegmatite in Ubini, Western Australia.

PART VI. ZINNWALDITE

I. MINERALOGY

A. Composition

General composition: The composition of zinnwaldite may be expressed by the following general formula:

$$K_2(Fe^2_{1-2},Li_{2-3},Al_2)(Si_{6-7},Al_{2-1})O_{20}(F_{3-2},OH_{1-2}).$$
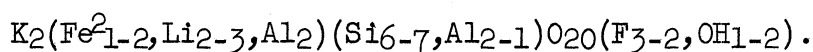
The (Al,Fe,Li) group usually contains six atoms but may be considerably deficient. The possible ionic substitutions that can take place are as follows:

For Si: One-fourth of the Si may be replaced by Al; minor Ti may also be substituted in this position.

For Al: Ti,Fe^2,Fe^3,Mn,Mg, and Li.

For K: Na,Ba,Rb,Cs, and minor Ca.

Trace elements: Glass (1935) in studying zinnwaldite from Amelia, Virginia, reports the largest variety of trace elements ever recorded in a zinnwaldite: B,Be,Sn,Zn,Rb,Li,Cs,Ti; Cs_2O = 0.10 percent and Rb_2O = 1.04 percent. Other trace elements in zinnwaldites are P,He,Mg,Mn^3,Ga,Ba,Sc,Tl, and Y (Rankama and Sahama, 1950).

The chemical similarity between zinnwaldite, lepidolite, and biotite are presented in Table XLIX.

TABLE XLIX

CHEMICAL RELATION OF ZINNWALDITE TO LEPIDOLITE AND BIOTITE

	Typical Zinnwaldite %	Typical Lepidolite %	Typical Biotite %
SiO_2	48	50	35
Al_2O_3	20	23	20
FeO	8	--	}18
Fe_2O_3	2	--	
Li_2O	4	5	--
F	7	7	--

Zinnwaldite is, in general, chemically closer to lepidolite than biotite, with the important exception of Fe.

B. Structure

Three structural varieties of zinnwaldites are now known:

(a) 1-layer monoclinic,

(b) 2-layer monoclinic muscovite-type, and

(c) 3-layer hexagonal.

Hendricks and Jefferson (1939) report zinnwaldites with the 1-layer structure from Amelia, Virginia; Zinnwald, Bohemia; and Brambach, Saxony. They also report a 3-layer hexagonal structure zinnwaldite from Amelia, Virginia. The X-ray data of the zinnwaldites studied by us is tabulated in Table L. Eight samples studied have crystallized as the 1-layer polymorph, five as the 3-layer polymorph, and one as a 2-layer polymorph very similar to that of normal muscovite.

An excellent Weissenberg photograph obtained from a flake of zinnwaldite (Spec. 1251) from Ubini, West Australia, (Murry and Chapman, 1931) indicates the 2-layer monoclinic structure with the presence of (06ℓ) reflections with ℓ odd (Fig. 13). The significance of these reflections has been discussed in detail by Hendricks and Jefferson (1939), and in this report with regard to lithian muscovite. The significant point is that (06ℓ) reflections with ℓ odd are present in the 2-layer muscovite (heptaphyllite) type of micas, and their presence indicates considerable distortion from the ideal mica structure. Hendricks and Jefferson (1939) believe that the muscovite-like micas produce these reflections because of an incomplete filling of the octahedral positions which results in the distortion. Muscovite has only two-thirds of the octahedral positions filled, whereas the ideal octaphyllite micas have all these positions occupied. Therefore, the presence of 06ℓ reflections with ℓ odd recorded on the Weissenberg photograph of Spec. 1251 implies that there is distortion in the structure, and that this zinnwaldite approaches the heptaphyllites in structure. This appears to be the only specimen of zinnwaldite which has been definitely shown to be more closely related structurally to heptaphyllite micas than to the octaphyllites. Noteworthy is the fact that the deficiencies in the Al,Fe,Li (octahedral) position have been reported. Although Lemke, et al. (1952) report the occurrence at Amelia, Virginia, of many books intermediate between zinnwaldite and muscovite with some gradations from muscovite centers to zinnwaldite borders, our examination of this material shows only a very narrow gradational zone, if any.

TABLE L

NEW STRUCTURAL STUDIES OF ZINNWALDITE

Identification Number	Polymorphic Form	2V*	Color	Locality	Remarks
778	1-layer	Medium	Green	Bohemia	Borrowed from Harvard
779	1-layer	ND	Pale green-brown	Zinnwald	Yale No. 1504
1329	1-layer	Large	Amber brown	Bohemia	Ontario Museum
1330	3-layer	ND	Dark brown	Altenberg	Rabenglimmer, Ontario Museum
1331	1-layer	ND	Green-black	Bohemia	Ontario Museum
780	1-layer	ND	Green-black	Fredrickstown, Mo.	American Museum
1128	1-layer	ND	Rust brown	Martha Mine, Pongb, Bolivia	U. S. Nat'l. Museum R8089
1251	2-layer	ND	Pale brown	Ubini, W. Australia	Murry and Chapman (1931)
1324	3-layer	ND	Dark brown	Brown Derby, Colorado	
1337 (a)	3-layer	Small	Brown	Brown Derby, Colorado	Core zone
1337 (b)	3-layer	Small	Dark brown	Brown Derby, Colorado	Margin Zone
786	1-layer	Medium	Pale brown	Amelia, Va.	Harvard
787 (a)	3-layer	ND	Pale brown	Amelia, Va.	Analyzed by Glass (1935)
1242 (a)	1-layer	Medium	Brown	Amelia, Va.	outer edge zone
1242 (b)	3-layer	Small	Dark brown	Amelia, Va.	inner zone
1389 (a)	3-layer	Small	Brown	Amelia, Va.	Margin Zone; Lemke et al., (1952)

*Large 2V means greater than 40°
Medium 2V means 15° -40°
Small 2V means 0° -15°
ND means not determined but assumed to be Medium
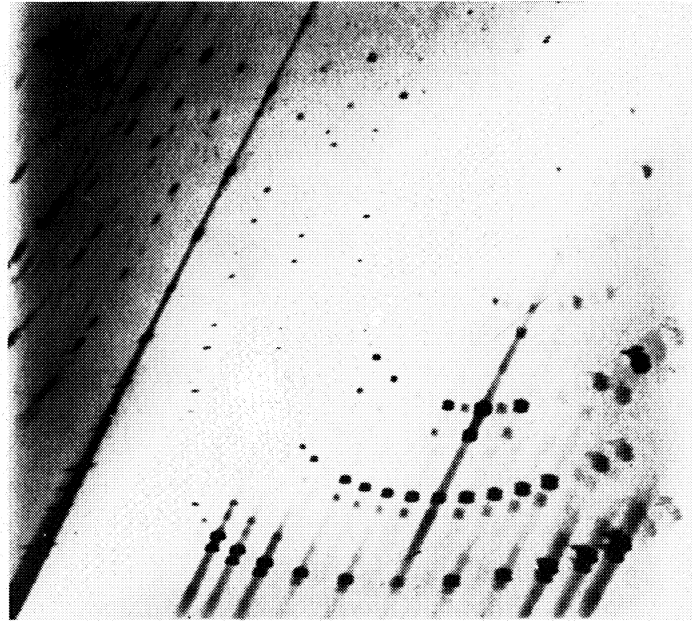
Fig. 13. 0-level a-axis Weissenberg photograph of
zinnwaldite with 2-layer monoclinic structure.
Compare with Figs. 5 and 6.

Murray and Chapman (1931) reported only a partial analysis (the alkalis) of the Ubini material. The Li_2O content of 1.13 percent is extremely low for a zinnwaldite and further studies may show that we are not dealing with a true zinnwaldite. Nevertheless, the color is very similar to other zinnwaldites and from the darkening of the Weissenberg films it is assumed that the Fe content is considerable; certainly more than high Fe, normal muscovites.

The itensities of the (06ℓ) reflections with ℓ odd (Fig. 13; compare with Figs. 5 and 6) from the specimen of zinnwaldite from Ubini (No. 1251) are of an order of magnitude somewhere intermediate between normal muscovite and lithian muscovite (apparently closer to normal muscovite). The intensities of these reflections and other selected $(0k\ell)$ reflections are listed in Table LI.

TABLE LI

APPROXIMATE OBSERVED INTENSITIES
OF SOME $(0k\ell)$ REFLECTIONS OF 2-LAYER FORMS

Plane	Normal Muscovite	2-Layer Zinn-waldite	Lithian Muscovite
020	W	W-	a
022	MW	MW	VW
026	a	VW	VW
045	a	VVVW	VW
061	W	W-	VW
065	VW	VW	VVW
066	W	W	VVW
067	VW	VW	VVW
069	W	VW	VVW

C. Optical Properties

In Table LII are listed the optical constants of zinnwaldites as reported in the literature. The range of values is as follows:

α = 1.535 - 1.558

β = 1.570 - 1.589

γ = 1.572 - 1.590

2V = 0° - 70° —— Average 25° - 30°

Axial plane: parallel with (010)

Color: gray to amber-brown

Sp. Gravity: 2.916 - 3.018

Unfortunately only three analyzed zinnwaldites have precise optical determinations; this prevents any definite correlation of optical data with composition. There does, however, appear to be a general increase in the indices with an increase in iron content.

D. Zoning and Overgrowths

Baumhauer (1879) reported a zoned zinnwaldite from Zinnwald, Bohemia. According to his descriptions, crystals of zinnwaldite showed zones of different widths normal to the A structure. These zones were yellow, whereas the rest of the crystal was colorless to gray. He postulated that the zoning was due to chemical differences. Upon examination under polarized light, the various zones were found by him to have the same absorption formulas. Sollas (1889) reports a zinnwaldite found in the granite of Mourne Mountains, Ireland, which shows a zonal structure with a dark-green center having an axial angle of 44°04' and a border zone having an angle of 52°06'.

Sterrett (1923, p. 133) reports zoning in a zinnwaldite from the Palermo Mine, Grafton County, New Hampshire: "The sheets of this mica show a clear brown core and a greenish-blue exterior about the color of indicolite tourmaline. These colors are arranged parallel to planes of crystallization, and the blue contains thin zonal growths of the brown."

A zoned crystal from Amelia, Virginia (spec. 1242) upon examination showed optical and structural variations. The dark-brown inner zone had a 2V of 3 to 4°, β = 1.584, and γ = 1.586, and had crystallized as the 3-layer form. The marginal zone had a 2V of 28°, β = 1.583; and γ = 1.588, and crystallized as the 1-layer form. It has been shown in the lepidolites that the 3-layer and 1-layer forms have almost identical chemical compositions; it is believed that the 3-layer form results from a twinning of the 1-layer form. This twinning also probably accounts for the structural variation in this zinnwaldite, as the chemistry of the two zones probably varies very little.

A systematic X-ray study of several zoned zinnwaldites from the Brown Derby, Colorado, pegmatite has shown that polymorphic variation does not necessarily exist within markedly zoned crystals. These micas have a light outer zone and a dark core. Both zones have the same optical constants; β = 1.584, γ = 1.585, and 2V = 0 - 2°, and both have crystallized as the 1-layer type.

Lemke et al. (1952 p. 129) report that many books intermediate between zinnwaldite and muscovite occur in the Amelia District of Virginia and that some grade from muscovite centers to zinnwaldite borders. They conclude that the zinnwaldite formed later than muscovite and in some books formed rims around cores of muscovite.

TABLE LII

OPTICAL PROPERTIES OF ZINNWALDITES
(From the literature)

Locality	α	β	γ	Axial Angle	Color	References
Zinnwald, Bohemia	1.551	1.578	1.581	2V = 29-30°	Brown	Larsen and Berman, 1934
Zinnwald, Bohemia	1.5435 -1.5450	1.5729 -1.5737	1.5750 -1.5757	2V = 30-32°		Winchell, 1942
Zinnwald, Bohemia	1.541	1.571	1.573	2V = 28°47'	Light Gray	Jakob, 1927
Zinnwald, Bohemia	1.5511	1.5777	1.5812	2V = 14°48' 2E = 47°30'		Kunitz, 1924
Zinnwald, Bohemia	1.539		1.564	2V = 30°		Hendricks and Jefferson, 1939
Altenberg, Saxony	1.5572	1.5850	1.5876	2V = 10°21' 2E = 36°20'		Kunitz, 1924
Fichtelgebirge, Bavaria				2E = 47°10'		Dürrfeld, 1909
Brambach, Saxony			1.572	2V = 25-30°		Hendricks and Jefferson, 1939
Volhynia, Russia			1.587	2V = 13-25°		Buryanova, 1940
Erongo Schlucht, S. W. Africa		1.573 +0.002	1.576 +0.002	2V = 31° ± 3°		Frommurze, Gevers, and Rossouw, 1942
Umberatana, South Australia		1.57		2V = 10°		Mawson and Dallwitz, 1945
Wakefield, Quebec	1.5357 -1.5440	1.5596 -1.5671	1.5628 -1.5701	2V = 30-39°		Winchell, 1942
New Ross, Nova Scotia				2V = 70° (?)	Amber Brown	Walker and Parsons, 1924
Amelia, Virginia	1.550 -1.558	1.580 -1.589	1.580 -1.590	2V = 0-33°	Bronze to Gray	Glass, 1935
Amelia, Virginia	1.550		1.584	2V = 0°		Hendricks and Jefferson, 1939

We have examined some material from Amelia supplied by W. R. Griffitts in order to determine the extent of gradation between zinnwaldite and muscovite. The heavily iron-stained specimen ($No.$ 1389) consists of an incomplete crystal which shows definite zoning. The boundary between the muscovite core and zinnwaldite rim appears megascopically sharp and distinct owing to the brownish tint of the zinnwaldite. Under the microscope the boundary is, in the main, sharp but jagged, as determined by differences in birefringence under crossed nicols. In a few small areas the boundary, on the basis of birefringence and size of the optic angle, appears gradational. However, the gradational zone is narrow, and its lateral extent is limited. The structure of the muscovite is 2-layer (normal). The zinnwaldite has the 3-layer hexagonal structure; the optic angle is small (at the most, $10°$) and variable. We have been unable to isolate any of the small "gradational areas" for X-ray studies. In general, however, the specimen represents an overgrowth rather than a gradation.

Lemke et al. (1952) have described overgrowths of zinnwaldite on muscovite at the Morefield mine, Amelia, Virginia. Zinnwaldite overgrowths on green biotite are reported by Shibata (1952B) in pegmatites of the Naegi district, Japan, and zinnwaldite overgrowths on muscovite were noted by the same investigator (Shibata, 1952B) occurring in pegmatites at Naegi and Tanokamiyama.

II. OCCURRENCE

Zinnwaldite is found chiefly in granite pegmatites and cassiterite veins associated with greisen. Table LIII lists the localities and type of deposit for most of the zinnwaldite occurrences described in the literature.

The survey of the literature describing the occurrences of zinnwaldite leads to the following generalizations concerning the paragenesis of zinnwaldite:

1. Zinnwaldite is associated with granitic magmas, especially with pegmatites, greisens, and granites.

2. Rocks containing zinnwaldite usually contain one or more of the following minerals: topaz, cassiterite, lepidolite, feldspar (cleavelandite), beryl, tourmaline, tantalite, columbite, monazite, spodumene, and fluorite.

3. Zinnwaldites in pegmatites are usually associated with Na-Li replacement units.

4. Zinnwaldite probably crystallizes after muscovite and/or biotite but before lepidolite (Shibata, 1952B).

TABLE LIII

OCCURRENCES OF ZINNWALDITE

Location	Type deposit	Associated minerals	Reference	Remarks
Zinnwald, Bohemia	greisen	cassiterite, topaz, albite	Beck (1905) Konta (1950)	Similarly at Altengerg (as rabenglimmer) and Erzgebrige
Bavaria Germany	pegmatite	quartz, albite, orthoclase	Mieleitner (1921)	At Fuchsbau Forest
Cornwall, England	greisen	cassiterite, topaz, albite	Dana (1892)	Also at St. Just
Makonu, Rhodesia	greisen		Zealley (1917)	
Hunan Prov., China	greisen	cassiterite, lepidolite	Ming and Chang (1935)	
Italy	Baveno granite	siderophyllite, lepidolite	Gallitelli (1936)	
York Region, Alaska	greisen	cassiterite, fluorite, topaz	Collier (1905)	
Umbertana, S. Australia	pegmatite		Mawson and Dallwitz (1945)	
Tourmaline Hill, S. Australia	greisen	feldspar, quartz	Mawson and Dallwitz (1945)	
Ubini, Western Australia	pegmatite	lepidolite	Murray and Chapman (1931)	Also other localities in Western Australia
Amelia, Virginia	pegmatite	topaz, albite, beryl	Glass (1935) Lemke et al. (1952)	

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TABLE LIII (CONT.)

Location	Type deposit	Associated minerals	Reference	Remarks
Brown Derby, Colorado	pegmatite	albite, quartz, tourmaline	Heinrich and Levinson (1953)	
White Picacho dist., Arizona	pegmatite	lepidolite, spodumene	Jahns (1952)	
Rockport, Mass.	granite	quartz, feldspar	Cooke (1867)	Also Cape Ann
Pointe du Bois, Manitoba	pegmatite	lepidolite, spodumene	Heinrich and Levinson (1953)	
Karibib, S. W. Africa	pegmatite	feldspar, tantalite, tourmaline	Frommurze et al. (1942)	Similarly at other localities: Omaruru
Stavoren Tin Fields, S. Africa	greisen	cassiterite	Wagner (1921)	
Greenland	pegmatite	albite, aegirite	Flink (1902) Dana (1892)	At Narsarsuk and Kangerdluarsuk
Russia	pegmatite	topaz	Buryanova (1940)	At Volhynia, Lavitinsky, and Turkestan Mt. Range
Tordal, Norway	pegmatite	amazonite, topaz, fluorite, cassiterite	Oftedal (1942)	
Madagascar	pegmatite	lepidolite	Lacroix (1922)	At Antsongombato, Antaboaka, etc.
Japan-Korea	pegmatite	lepidolite, spodumene, cassiterite, fluorite, topaz	Shibata (1952B) Harada (1936) Iimori and Yoshimura (1926)	

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PART VII. MISCELLANEOUS MICAS

I. PARAGONITE

$(Na,K)_2Al_4(Si_6Al_2)O_{20}(OH)_4$; chemically very similar to muscovite except with Na in place of K; K in very subordinate amounts. Schaller and Stevens (1941) have pointed out that the series muscovite-paragonite is not completely represented in nature. They believe there is no longer any doubt as to the validity of paragonite as a separate species even though designation of many alleged soda micas as paragonite was apparently based on faulty analytical determinations.

As the optical properties of paragonite, Winchell and Winchell (1951) give:

$$\alpha = 1.564 - 1.577$$
$$\beta = 1.599 - 1.605$$
$$\gamma = 1.600 - 1.609$$
$$2V = ca. \ 40°$$

Paragonite cannot be distinguished from muscovite on the basis of optical properties alone.

As for its structure, Bannister (1943) has determined the following cell dimensions for paragonite which indicate the close similarity to the normal 2-layer muscovite structure:

$$a = 5.12Å \qquad b = 8.87Å \qquad c \sin\beta = 18.95Å$$

On synthetic material, Gruner (1942) found:

$$b_o = 8.90Å \qquad c_o = 19.33$$

Gruner (1942) suggests that paragonite is uncommon because Na ions are so small that they will not fit readily into the K positions of the micas. In twelvefold coordination positions, bonding with Na is extremely weak.

Paragonite is reported as occuring in schists and phyllites, in some with kyanite, also as an alteration of corundum. Jahns (personal communication) has found paragonite in a pegmatite.

II. ROSCOELITE

Roscoelite has been generally accepted as a vanadiferous muscovite in which V has replaced much of the Al. This conclusion was reached by Clarke on the basis of 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 similarities to muscovite, such as the position of the optic plane normal to (010), were noted. Indices, however, are high: α = 1.610 β = 1.685 and γ = 1.704 ($\pm$ 0.003, sodium light). 2E = 42° to 69°. Pleochroism is fairly strong: γ = green-brown, β = olive-green and α = olive-green. Absorption, $\gamma > \beta > \alpha$.

Atomic ratios calculated by Wells and Brannock (1946) on the basis of ten chemical analyses show that V and Al combined (with the exception of the Al required to fill tetrahedral positions) total two atoms with octahedral coordination in the mica sheet. This is the theoretical number for a hepta-phyllite (muscovite-type) mica. The isomorphous replacement also seems probable on the basis of ionic radii: Al^{3+} = 0.57kX, V^{3+} = 0.65kX.

The first published results of X-ray work on roscoelite that we have been able to find are embodied in a statement in Fischer (1942, pp. 376-377):

"The principal vanadium mineral is micaceous, and it has heretofore been thought to be roscoelite, the vanadium mica; but as a result of recent X-ray studies by Sterling Hendricks of the Department of Agriculture, it is now thought to belong to the imperfectly understood hydrous-mica group of clay minerals." This tentative statement apparently has been widely accepted and the micaceous vanadium mineral of the Colorado Plateau area has subsequently been referred to as vanadium hydromica upon several occasions by geologists studying these deposits. The extent of its dissemination may be gauged by its appearance in a Russian article (Kultiassov and Dubinkina, 1946). Since the results of our studies on roscoelite were not in agreement with this identification, we wrote to Dr. Hendricks for further details. In his reply he states:

"I probably made an error in the roscoelite. The sample used was an extremely small one (actually too small to work with) and the powder diffraction photographs were poor. The statement was based on an apparent value of d(001) in excess of 10.0Å."

The only other reported structural work is a statement in Wells and Brannock (1946, p. 121) attributed to Dr. J. M. Axelrod, which indicates that

X-ray powder patterns of roscoelite have " ... lines very close to those of muscovite, paragonite, and biotite." However, studies by Fischer (1937, p. 929) and by Alice D. Weeks (personal communication) seem to indicate the presence of another vanadiferous micaceous mineral, possibly of chloritic nature, in the V ores of the Placerville district of the Colorado Plateau province.

In the University of Michigan Mineralogical Collections only a few fine-grained samples of roscoelite were available, so that the study was begun by means of the powder method. Powder photographs were taken of three specimens from California localities. All of these have spacings that correspond closely to those of the 1-layered monoclinic mica polymorph (Table LIV, column 3); they are slightly, but consistantly, larger than those of the 1-layer lepidolite polymorph (p.141). Specimens of type roscoelite from the Stockslager mine were obtained from the U. S. National Museum and Harvard Mineralogical Museum. Powder photographs of this type material also gave the 1-layer polymorph pattern. On a single flake suitable for Weissenberg study from this locality, a 0-level a-axis photograph indicated the presence of the 1-layered monoclinic structure, with the optic plane parallel with the side pinacoid. Samples of two recently analyzed roscoelites, one from Paradox Valley, Colorado, described by Wells and Brannock (1946), and the other from the Placerville District, Colorado, described by Fischer, Haff, and Rominger (1947) were obtained from Dr. Axelrod and are the same ones on which he based his above cited statement in Wells and Brannock (1946, p. 121). Powder photographs of these specimens likewise gave 1-layer patterns with no values of d(001) in excess of 10.0Å.

Two specimens of alleged roscoelite-bearing ore were loaned by Prof. E. N. Goddard for study. One is from the King Wilhelm mine near Jamestown, Colorado; the other from the Buena mine in the same district. Lovering and Goddard (1950, p. 264) report for the Jamestown district that, "Associated with the ore in the Rip Van Dam, King Wilhelm, and Gladiator veins are small amounts of roscoelite ..." Roscoelite in this district also has been noted by Bray (1942, p. 787), who correlates the presence of this mica in veins with the relatively large amounts of V in the tertiary granodiorite to which the veins are genetically related. Genth (1876) has described and analyzed roscoelite from the nearby Magnolia district in Boulder County, Colorado.

The Jamestown specimens consist of thin vein coatings of quartz, fluorite, pyrite, tellurides, and a very fine-grained gray-green mica, most of which is intimately intergrown with fine-grained quartz. Single flakes, about 1 mm in diameter, were separated from the specimen from the King Wilhelm mine, and micaceous powders were obtained from both specimens. Weissenberg photographs establish that the single flakes have the 2-layer monoclinic structure of normal muscovite. The powder X-ray photographs show strong quartz contamination, and the polymorphic type is not determinable. Mrs. Alice S. Corey of our laboratory could not substantiate the presence of V in these micas by means of the test described by Axelrod (1946).

TABLE LIV

X-RAY POWDER DATA FOR ROSCOELITE AND BARIUM MUSCOVITE

Muscovite Nagelschmidt (1937)		Roscoelite California Cu Kα, λ = 1.53736kX		Barium Muscovite Franklin, N. J. Cu Kα, λ = 1.53736kX			
9.98	s	9.91	s	9.80	m	1.635	vw
5.00	s	4.57	m	6.56	m	1.586	vvw
4.49	s	4.38	w	5.70	vw	1.569	vw
3.91	w	4.15	w	4.91	vw	1.530	vvw
3.73	w	3.66	m	4.56	w	1.516	vvw
3.50	m	3.34	s	4.43	w	1.497	w
3.33	vs	3.12	m	4.20	vvw	1.454	vvw
3.20	m	2.90	vw	3.76	m.	1.419	vvw
3.00	m	2.70	w	3.60	w	1.383	vw
2.88	m	2.60	m	3.52	w	1.356	vvw
2.80	m	2.426	w	3.42	m	1.340	vw
2.57	vs	2.355	vw	3.31	s	1.208	vvw
		2.286	vvw	3.23	m	1.193	vw
2.475	w br	2.221	vvw	2.99	m	1.153	vvw
		2.167	vw	2.89	m	1.137	vw
2.385	m	2.041	vvw	2.83	vvw	1.107	vvw
2.28 }	w	2.007	vw	2.76	m	1.078	vvw
2.19 }		1.665	vw	2.66	vvw	1.056	vvw
2.134	s	1.603	vvw	2.56	s		
1.995	vs	1.524	m	2.487	vvw		
				2.422	vvw		
1.730	vw			2.376	vvw		
1.651	s br			2.314	vw		
1.546	vw			2.226	vvw		
1.523	vw			2.165	w		
1.500	s			2.123	vw		
1.450	vw			2.056	vvw		
1.421	vw			1.990	m		
1.356 }	m			1.937	vvw		
1.337 }				1.848	vvw		
1.296	m			1.795	m		
1.247	m			1.669	vvw		

Roscoelite has been found in the ores of Cripple Creek (Lindgren and Ransome, 1906, p. 128) also associated with quartz, fluorite, and calaverite. Unfortunately, no analyses of this material are available.

Several other occurrences of roscoelite or vanadium-bearing muscovite have been reported. The vanadiferous muscovite (1.08 percent V_2O_3) from Schmiedefeld, Saalfeld in Thuringia, Germany, (Jung, 1937) has the normal 2-layer monoclinic structure, as is shown by his X-ray powder spacings (p. 40). It occurs with pyrite in quartz veins in an iron deposit. Chatterjee (1931) reports roscoelite in quartz veins with tourmaline and rutile and in schists containing muscovite, rutile, roscoelite, and kyanite in the Bhandara district of India. He states (p. 297), "The roscoelite mica occurs in the kyanite-bearing rocks in small nests. It is pale olive-green and slightly pleochroic; and its optical properties correspond to those of muscovite. Tests for nickel and chromium failed, while vanadium was found to be present. This mineral is also found in certain quartz veins." Efforts to obtain specimens of roscoelite from this locality were unsuccessful.

Service (1943) has described a green vanadiferous mica (1.7 percent V_2O_5) from cavities of the Nsuta manganese deposits of the Gold Coast and another green mica (0.62 percent V_2O_3) from a quartz vein in the same deposit. Again an attempt to secure some of this material was unsuccessful.

The vanadium mica of the vanadium deposits of the Kara-tau Mountains in Kazak, U.S.S.R. has been described as roscoelite by Koslov and Sokolov (1944) and by Turin (1944) and later described as vanadium-oellacherite by Kultiassov and Dubinkina (1946). But as we have shown under oellacherite, the homogeneity and identity of this material are doubtful.

Other reported localities for roscoelite include Kalgoorlie, Western Australia (Lindgren, 1933, p. 680) also with native gold and tellurides, and west of Pretoria, Transvaal, in magnetite ores (Wagner, 1928, p. 95-96) of the Daspoort horizon, where it lies within the contact aureole of the Bushveld Complex.

Apparently the normal 2-layer monoclinic muscovite structure can accommodate the substitution of significant amounts of V for Al without any detectable change. Those micas with the 1-layer roscoelite structure contain 12 percent V_2O_5 or more. We believe that this structural change accompanies the large-scale substitution of V for Al. The few available analyses seem to indicate that there is no complete series between vanadian muscovite and roscoelite. It would perhaps be instructive to determine the structure of the vanadiferous mica from the Magnolia district, Colorado (Genth, 1876), which contains 7.78 percent V_2O_3.

Since roscoelite is both chemically and structurally distinct from other micas, it can no longer be regarded as a vanadiferous muscovite, but must be given the status of an independent species.

III. TAENIOLITE

Taeniolite is an extremely rare member of the mica group whose formula is $K_2Mg_4Li_2Si_8O_{20}F_4$. It is characterized by the absence of essential aluminum, and for this reason is unique among the micas. Mg and Li alone appear to occupy octahedral positions. The high SiO_2 and F along with a substantial Li_2O (approximately 3.5 percent) content make it appear related to octophyllite lepidolite; future investigations may show this mineral to be a magnesian lepidolite in which Mg has replaced Al.

An occurrence of taeniolite from Magnet Cove, Arkansas, is described by Miser and Stevens (1938). They also discuss the relation of taeniolite to other micas. The optical properties of the Arkansas material are:

$$\alpha = 1.522, \; \beta = \gamma = 1.553, \; \gamma-\alpha = 0.031; \; 2V = \text{near } 0°$$

Hendricks and Jefferson (1939) demonstrated that taeniolite from this locality has the 1-layer monoclinic structure.

Taeniolite is found in veins and pockets with quartz, dickite, rutile and other minerals at this locality and also at Narsarsuk, Greenland, and also possibly in the Kola Peninsula.

PART VIII. PARAGENESIS OF PEGMATITIC MICAS

I. SEQUENCE OF CRYSTALLIZATION OF MICAS

A. Zonal Growths

In the muscovites, the common type of color zoning consists of darker-brown cores and lighter-brown or greenish-brown outer zones. From the relation of color to composition, this indicates a decrease in Fe^3 and Ti outward and probably an increase in Fe^2 outward. Analyses 8a and 8b, Table LV, representing a zoned muscovite from the Jasper mine are typical.

	8a green margin	8b ruby core
Total Fe as Fe_2O_3	2.9	2.4
MgO	.45	.75
TiO_2	.05	.30

The inner parts of muscovite crystals that show zoning normally are richer in hematite indicating the greater availability of Fe^3 and the greater ability of muscovite to incorporate it during the early growth stages.

Zoned biotites are less common, but some examples have been analyzed by Shibata (1952A) from pegmatites in the Naegi district, Japan. Green outer zones have somewhat more Fe^2, slightly less Fe^3, and markedly less Ti than brown cores.

	green margin	dark-brown core
Fe_2O_3	7.44	7.73
FeO	24.12	22.41
MgO	0.29	0.62
TiO_2	0.08	0.76

TABLE LV

SPECTROCHEMICAL ANALYSES* OF MUSCOVITES FROM JASPER PEGMATITE
(FRANKLIN-SYLVA DISTRICT, N. C.)

	6 ruby inner part of wall zone	8a green margin	8b ruby core
		zoned-book outer part of wall zone	
Fe_2O_3	3.2	2.9	2.4
MgO	1.7	.45	.75
MnO	.04	.07	.07
TiO_2	.32	.05	.30
CaO	.014	.002	.001
SrO	.001	----	----
BaO	.08	.04	.07
Na_2O	.67	.75	.90
Li_2O	----	----	----
Rb_2O	----	----	----
Cs_2O	----	----	----
Ga_2O_3	.017	.016	.02
Sc_2O_3	.003	----	----
Co_2O_3	.0002	----	.0004
Cr_2O_3	.0003	.0001	.0003
SnO_2	.004	.004	.004
V_2O_5	.014	.001	.01
F	----	----	----

*Spectrochemical analyses by C. E. Harvey

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B. Overgrowths Between Micas

Overgrowths between mica species are commonly of the types:

1. Muscovite overgrowths on biotite.

2. Lepidolite overgrowths on muscovite.

3. Lepidolite overgrowths on biotite.

4. Zinnwaldite overgrowths on muscovite.

5. Zinnwaldite overgrowths on biotite.

Reverse relations, such as biotite on muscovites and muscovite on lepidolite are known but are rare for the pairs.

C. Localization in Pegmatites

Biotites of magmatic origin occur mainly in wall zones or outer intermediate zones. In wall zones they are earlier, normally, than any accompanying muscovite. Wall-zone and intermediate-zone biotite is earlier, of course, than core-margin muscovite. Hydrothermal biotite occurs as fracture fillings but very rarely in replacement units. Where biotite of the fracture-fillings type occurs associated with fracture-filling or replacement muscovite, the latter is the younger.

Shibata (1952A) has analyzed several paragenetically distinct biotites from the Iisaka pegmatite, Japan:

	Older brown biotite in graphic granite zone	Younger green biotite, fracture controlled, associated with fergusonite
Fe_2O_3	12.89	11.72
FeO	20.06	23.73
MgO	1.21	0.14
TiO_2	0.81	0.53

The younger biotite is lower in Fe^3, Mg, and Ti but higher in Fe^2.

Muscovites of wall zones and outer zones are older than those of core-margin units, which in turn, are older than muscovite formed hydro-thermally along fractures and by replacement. Table XIX illustrates the differences in wall-zone (ruby) and core-margin (green) muscovites from the Kiser Pegmatite. The younger muscovite has higher total Fe and lower Ti.

The muscovites of the Franklin-Sylva district, North Carolina, are almost entirely of magmatic origin, and most of them are wall-zone micas. The micas of the Petaca district, New Mexico occur chiefly in replacement. units and are in the main of hydrothermal origin (Jahns, 1946). Table LVI contrasts their average compositions.

Again, the chief differences are in Fe, Mg, Mn, and also in Li and F. Na, Ti and some of the trace elements also show variation.

Liashchenko (1940) has analyzed two muscovites from the Biriussa mica mine in eastern Siberia (Table LVII). Number 1 is a normal pegmatitic muscovite; No. 2 is a fine-grained secondary muscovite that forms as pseudo-morphs replacing the book muscovite (No. 1).

The TiO content of No. 2 is abnormal and may be presumed to be due to inclusions of a Ti mineral.

It is to be noted that the secondary muscovite is phengitic, is slightly deficient in K, and is very low in $Fe^3 + Fe^2$.

As early as 1887, Scharizer (1887B) analyzed the several micas from the Schüttenhofen pegmatite.(Table LVIII).

He concluded that the youngest is richest in SiO_2, poorest in divalent bases, and is also the lightest in color. In the outer zone, muscovite of type I also forms overgrowths on biotite so that the complete sequence is:

Biotite ⟶ muscovite I ⟶ muscovite II ⟶ lepidolite.

He believed that the type II muscovite is poor in Fe^3 because tourmaline formed with it, which utilized that element.

Shibata (1952A) has deciphered the paragenetic sequences of micas in several districts:

1. Naegi district, Japan

                    ──────────────→   younger

      brown biotite

          green biotite

              protolithionite ── zinnwaldite ── lepidolite

                  muscovite ─ ─ ─ ─ ─ ─ ─ ─ ─ ─ ─ ─ ─ ─ ─ sericite yellow mica

2. Bunsen pegmatite, South Kankyō-dō, Korea

                    ──────────────→   younger

      biotite

          radial muscovite ── zinnwaldite ── white ──── pink
                                            lepidolite lepidolite

3. Sakihama, Japan   See Table LIX.

        Heinrich and Levinson (1953) have cited evidence to show that in
many cases rose muscovite is younger than lepidolite.

TABLE LVI

COMPARISON OF AVERAGE COMPOSITION* OF MUSCOVITES FROM
FRANKLIN-SYLVA, N. C. AND PETACA, N. MEX., DISTRICTS

	Franklin-Sylva North Carolina	Petaca New Mexico
Fe_2O_3	2.77	4.24
MgO	1.12	.12
MnO	.06	.47
TiO_2	.17	.105
CaO	.015	.005
SrO	.0009	absent
BaO	.19	.006
Na_2O	.83	1.08
Li_2O	absent	.46
Ga_2O_3	.017	.041
Sc_2O_3	.0014	.0002
Co_2O_3	.0002	.0001
Cr_2O_3	.0002	.00001
SnO_2	.0018	.014
V_2O_5	.0066	.0004
F	.029	1.00

*Based on spectrochemical data by C. E. Harvey

TABLE LVII

TWO MUSCOVITES ANALYZED BY LIASHCHENKO (1940)

	1	2
SiO_2	44.01	51.07
TiO_2	0.57	4.52
ZrO_2	---	0.03
Al_2O_3	34.23	29.29
Cr_2O_3	n.d.	---
Fe_2O_3	1.09	0.45
FeO	---	0.20
MgO	1.35	1.36
CaO	0.79	---
K_2O	10.45	7.71
Na_2O	0.38	---
H_2O+	5.78	5.24
H_2O-	1.62	0.56
CO_2	0.18	0.16
	100.45	100.59

TABLE LVIII

MICAS FROM THE SCHÜTTENHOFEN PEGMATITE ANALYZED BY SCHARIZER (1887B)

	I muscovite, brown, oldest	II muscovite yellow	III lepidolite youngest
SiO_2	43.67	44.08	49.25
Al_2O_3	36.69	36.83	25.26
Fe_2O_3	2.10	0.48	---
FeO	0.55	0.74	0.84
MnO	tr	0.25	0.85
K_2O	8.57*	11.10	13.85*
Na_2O	1.95	0.20	0.35
Li_2O	tr	0.37	5.38
CaO	--	0.20	--
H_2O+	4.35	4.98	1.76
H_2O-	1.15	1.17	--
F	0.35	0.19	5.68
	99.38	100.59	103.22
$O = F =$	0.14	0.08	2.38
	99.24	100.51	100.84

*Includes Rb_2O and Cs_2O

TABLE LIX

TWO MICAS FROM SAKIHAMA, JAPAN ANALYZED BY SHIBATA (1952A)

	Zinnwaldite (core margin)	Lepidolite (Core, in vugs)
SiO_2	45.98	46.86
TiO_2	0.22	0.14
Al_2O_3	18.04	21.25
Fe_2O_3	3.56	1.01
FeO	6.00	0.16
MgO	1.50	0.52
MnO	2.46	7.55
CaO	1.80	0.78
Na_2O	0.25	0.22
K_2O	9.91	9.48
Li_2O	4.57	5.20
Rb_2O	---	1.36
Cs_2O	---	0.04
H_2O+	0.86	0.25
H_2O-	1.14	1.28
F	5.66	8.31
	102.08	104.41
$O \equiv F$	2.38	3.66
	99.70	100.75

II. CHEMICAL EVOLUTION

Shibata (1952B) concludes that there exist three lines of descent in the evolution of pegmatitic micas:

1. biotite $\longrightarrow$ lepidomelane

   (Mg-Fe^2 $\longrightarrow$ Fe^2-Fe^3)

2. muscovite $\longrightarrow$ lithia mica

3. biotite $\longrightarrow$ lithia mica

He also notes (p. 160) that, "In later stage minerals found in pegmatites the ratio of MnO to FeO increases and at the same time lithium increases especially in mica, etc. When magnesium almost diminishes in later stage-minerals, manganese takes its place. Relative stage of magmatic differentiation is represented by the ratio of $FeO/(MgO + FeO)$ at the earlier stage and by $MnO/(FeO + MnO)$ at the later one." With these general conclusions the writers are in agreement, as was expressed by Heinrich (1952). Shibata also states (p. 160), "Ferric iron can be derived from ferrous one by oxidation by pegmatitic gases and so the ratio of ferric to ferrous iron has no constant relation, otherwise ferric iron increases in minerals of the later stage at the expense of ferrous iron." The writers agree that ferrous iron can be converted to ferric by means of the escape of pegmatitic gases, but conclude that such oxidation is more likely to take place in the outer parts (wall zones) of pegmatites. Thus wall-zone muscovites contain more Fe^3 (and exsolved Fe_2O_3) than core-margin types and biotites of outer zones will also probably be shown to contain more Fe^3 than core-margin types (most biotites included in green core-margin muscovites are themselves green) and more Fe^3 than fracture-filling biotite.

Table IX summarizes the average contents of the various oxides in the micas analyzed for this study.

In the muscovites the chemical evolution normally involves the changes indicated in Table LXI.

Normally the sequence:muscovite $\longrightarrow$ lithian muscovite $\longrightarrow$ lepidolite is accompanied by:

TABLE LX

AVERAGE CONTENTS* OF VARIOUS OXIDES IN MICAS

	Muscovite	Lepidolite	Rose Muscovite
Fe_2O_3	2.90	0.48	0.11
MgO	0.93	0.04	0.04
MnO	0.11	1.16	0.33
TiO_2	0.13	0.01	0.002
CaO	0.007	0.007	0.004
SrO	0.0005	0.006	0.0004
BaO	0.114	0.0003	0.0006
Na_2O	1.02	0.41	0.87
Li_2O	0.09	3.77	0.32
Rb_2O	--	0.54	0.62
Cs_2O	--	0.08	0.11
Ga_2O_3	0.022	0.017	0.024
Sc_2O_3	0.0011	0.0006	---
Co_2O_3	0.0002	--	0.0004
Cr_2O_3	0.0001	0.000004	0.00004
SnO_2	0.0067	0.021	0.025
V_2O_5	0.0004	0.0001	0.00005
F	0.24	5.41	0.42

*Based primarily on spectrochemical analyses by C. E. Harvey

TABLE LXI

CHEMICAL EVOLUTION OF MUSCOVITE

| | Oldest → → → Youngest | | | |
	Outer-zone muscovite	Core-margin muscovite	Replacement muscovite	Fine-grained replacement muscovite ("sericite")
Fe^3	maximum	——— decreasing ——— →		
Fe^2	——— increasing ——→		maximum	← variable, generally low
Mg	maximum	——— decreasing ——— →		
Mn	——— increasing ——→		maximum	high
Ti	maximum	——→ generally decreasing ——→		
Ca	——— decreasing slightly ——————→			
Ba	maximum ←——— decreasing ———→			
Na	——— increasing ——→		maximum	← slight increase
Li	generally absent	low increasing →	maximum	low or absent
F	low	low → increasing	maximum	low and variable ←
OH	high	slight decrease ←	lowest →	generally high
Si	generally the same		marked increase →	maximum
K	← slight increase		← great increase	lowest

Ca, Sr, Co, Cr, Sn show little systematic variation; V, Sc

appear to be concentrated slightly in the older micas; Ga is

slightly higher in younger micas; Rb and Cs are absent in most

micas except the rose types.

1. An increase in Li,Mn,Sr,Rb,Cs,F.

2. A decrease in Mg, total Fe,Ti,Na,Ba.

3. A slight decrease in V(?),Cr,Co,Sc.

4. A slight increase in Sn(?).

5. Essentially no change in Ca,Ga.

Rose muscovites, which are generally later than lepidolites, differ from the latter in having still lower total Fe,Mn,Ti,Sr,Li,Sc, and F; and higher Na and perhaps Ga. Rb,Cs,Ca,Ba,Sn, and V are essentially the same. These appear to be the only muscovites that normally contain Rb and Cs.

In the sequence:zonal biotite $\longrightarrow$ fracture-filling biotite the data are relatively few, but the indicated accompanying changes are:

1. Decrease in Fe^3,Ti,OH(?).

2. Increase in Fe^2,Li(?),F(?)

The chemical changes that appear in successively younger generations of pegmatitic micas reflect in general, and in detail, the differentiation trends of pegmatitic magmas and their derivative hydrothermal solutions as well as the course of differentiation of magmas in general.

In the magmatic micas with decreasing temperature of crystallization Mg is supplanted by Fe^2, and the Fe^2 by Mn. The relatively high Fe^3 contents of older (higher-temperature) micas may be due to oxidation by gases escaping from the partly crystallized pegmatite magma. With decreasing temperatures less Ti and Ba can be incorporated in micas. Li,Rb,Cs, most Mn, and F do not enter appreciably into the structure of the magmatic micas but are rejected to the hydrothermal fraction, along with additional Si.

REFERENCES

Amelinckx, S. (1952A), Nature, 169, p. 580.

——————— (1952B), Natuurwet. Tijdschr. (Belgium), 34, pp. 73-75.

Axelrod, J. M. (1946), U.S.G.S. Bull., 950, pp. 19-23.

——————— and Grimaldi, F. S. (1949), Am. Mineral., 34, pp. 559-572.

Bannister, F. A. (1943), Min. Mag., 26, pp. 304-307.

Bastin, E. S. (1911), U.S.G.S. Bull., 445, p. 50.

Bauer, L. H., and Berman, H. (1933), Am. Mineral., 18, p. 30.

Baumhauer, H. (1879), Zeit. Krist., 3, pp. 113-121.

——————— (1900), Zeit. Krist., 32, pp. 164-176.

——————— (1903), Eclogae Geol. Helvetiae, 7, p. 354.

——————— (1912), Zeit. Krist., 51, pp. 344-357.

Beck, R. (1905), The Nature of Ore Deposits: p. 205.

Berggren, T. (1940), Geol. Fören. Förh., 62, pp. 182-193.

——————— (1941), Geol. Fören. Förh., 63, pp. 262-278.

Berman, H. (1937), Am. Mineral., 22, pp. 342-408.

Bowman, H. L. (1902), Min. Mag., 13, p. 98.

Bragg, W. L. (1937), Atomic Structure of Minerals: Cornell Univ. Press, Ithaca, N. Y.

Brammall, A., Leach, J. G. C., and Bannister, F. A. (1937), Min. Mag., 24, pp. 507-520.

Bray, J. M. (1942), Geol. Soc. Amer. Bull., 53, pp. 765-814.

Brögger, W. C. (1921), Die Eruptivgesteine des Cristianiagebietes: IV, p. 294.

Brooke, H. J., and Miller, W. H. (1852), An Elementary Introduction to Mineralogy by the Late William Phillips: London.

Brown, G., and Norrish, K. (1952), Min. Mag., 29, pp. 929-932.

Buryanova, E. Z. (1940), Mem. Soc. Russe Min., 69 (4), pp. 519-540.

Cameron, E. N., Jahns, R. H., McNair, A. H., and Page, L. R. (1949), Econ. Geol., Monograph 2.

Chapman, R. W., and Williams, C. R. (1935), Am. Mineral., 20, pp. 502-530.

Chatterjee, S. K. (1931), India Geol. Surv. Records, 65, pp. 285-305.

Chester, A. H. (1894), Trans. N. Y. Academy of Sciences, 13, p. 181.

——————— (1896), A Dictionary of the Names of Minerals: New York.

Chudaba, K. (1929), Cent. Min., Abt. A, pp. 11-18.

Clark, R. W., and Hunt, W. F. (1915), Cent. Min., pp. 666-668.

Clarke, F. W. (1887), Amer. Journ. Sci., 34, pp. 131-137.

——————— (1888), U.S.G.S. Bull., 42, pp. 11-27.

——————— (1889), Amer. Journ. Sci., 138, pp. 384-393.

——————— (1914), U.S.G.S. Bull., 588.

Coats, R. R., and Fahey, J. J. (1944), Am. Mineral., 29, pp. 373-377.

Collier, A. J. (1905), U.S.G.S. Bull., 229, p. 20.

Collins, R. S. (1942), Min. Mag., 26, pp. 254-259.

Cooke, J. P. (1867), Amer. Journ. Sci., 2nd Series, 43, pp. 217-230.

Cross, W. (1897), Amer. Journ. Sci., Ser. 4, 4, pp. 115-141.

Daimon, N. (1952), Jap. Journ. Ind. Chem., 55, pp. 694-695.

Dana, E. S. (1892), System of Mineralogy: 6th ed., New York.

——————— (1922), A Textbook of Mineralogy: 3rd ed., New York.

——————— (1926), A Textbook of Mineralogy: 3rd ed., New York.

Dana, J. D. (1854), System of Mineralogy: 4th ed., New York.

——————— (1867), Amer. Journ. Sci., 94, pp. 252-263.

Dana, J. D. (1868), <u>System of Mineralogy</u>: 5th ed., New York.

——————— (1875), <u>System of Mineralogy</u>: 5th ed., New York.

——————— (1882), <u>System of Mineralogy</u>: 5th ed., New York.

Daub, R. (1913), Neues Jahrb., (1), pp. 214-217.

Des Cloizeaux, A. (1862), <u>Manuel de Minéralogie</u>: Paris.

Doelter, C. (1914), <u>Handbuch der Mineralchemie</u>: 2, Part 2, p. 442.

Durrfeld, V. (1909), Zeit. Krist., 46, pp. 563-598.

Ellsworth, H. V. (1932), Canada Geol. Survey, Econ. Geol. Ser., No. 11.

Erämetsä, O., Sahama, T. G., and Kanula, V. (1943), Bull. Geol. Comm.
        Finlande, 128, pp. 80-88.

Eskola, P. (1914), Bull. Geol. Comm. Finlande, No. 40, p. 37.

Fermor, L. L. (1909), Mem. Geol. Surv. India, 37.

Fischer, R. P. (1937), Econ. Geol., 32, pp. 909-951.

——————— (1942), U.S.G.S. Bull., 936 P.

——————— Haff, J. C., and Romiger, J. F. (1947), Colorado Scientific
        Soc. Proc., 15, No. 3, pp. 117-146.

Flink, G. (1899), Meddelser om Grönland, 24. (Cit. in Neues Jahrb., Referata,
        pp. 29-30, 1902).

Ford, W. E. (1932), <u>A Textbook of Mineralogy, by E. S. Dana</u>: 4th ed.,
        John Wiley and Sons, Inc., New York.

Foster, M. D. (1952), Bull. Geol. Soc. Am., 63, pp. 1250-1251.

Frank, F. C. (1949), Discus. of Faraday Soc., No. 5, Crystal Growth, pp.
        48-54.

Freudenberg, W. (1919), Mitt. Badisch. Geol. Landesanst., 8, p. 317.

Frommurze, H., Gevers, T. W., and Rossouw, P. J. (1942), Geol. Survey
        South Africa, Explanation of Sheet No. 79, pp. 107-114.

Frondel, C. (1936), Am. Mineral., 21, pp. 777-799.

——————— (1940), Am. Mineral., 25, pp. 69-87.

——————— and Ashby, G. E. (1937), Am. Mineral., 22, pp. 104-121,

Furcron, A. S., and Teague, K. H. (1943), Georgia State Div. of Conservation, Geol. Surv. Bull., 48.

Gädeke, R. (1938), Chemie der Erde, 11, pp. 592-636.

Gallitelli, P. (1936), Peridico Min. Roma, 7, pp. 61-76.

Gennaro, V. (1925), Rend, Accad. naz. Lincei, Ser. 6, 2, pp. 508-510.

Genth, F. A. (1876), Amer. Journ. Sci., 110, pp. 32-35.

Ginsburg, J. (1930), Zeit. Praht. Geol., 38, pp. 24-30.

Glass, J. (1935), Am. Mineral., 20, pp. 741-768.

Grigoriev, D. P. (1935), Mem. Soc. Russe Min., 64, pp. 76-79.

———————————, (1936), Bull. Soc. Nat., Moscow, 17(4-5), pp. 14-30.

Grim, R. E., and Bradley, W. F. (1951), The Mineralogical Society, London, pp. 138-154.

————— Bray, R. H., and Bradley, W. F. (1937), Am. Mineral., 22, pp. 813-829.

Grout, F. F. (1924), Am. Mineral., 9, pp. 159-165.

Gruner, J. W. (1942), Am. Mineral., 27, pp. 131-134.

—————————— (1948), Am. Mineral., 33, pp. 679-691.

Hall, C. E. (1885), Penn. Geol. Survey Report of Progress C^5, Part I.

Hall, A. L. (1926), Proc. Geol. Soc. of South Africa, 29, pp. 17-46.

Hall, A. J. (1941), Am. Mineral., 26, pp. 29-33.

——————————, (1941), Am. Mineral., 26, pp. 34-41.

Hallimond, A. F. (1925), Min. Mag., 20, pp. 305-318.

—————————— (1926), Min. Mag., 21, pp. 25-33.

—————————— (1927), Am. Mineral., 12, pp. 413-414.

—————————— (1947), Min. Mag., 28, pp. 230-242.

Hanley, J. B., Heinrich, E. Wm., and Page, L. R. (1950), U.S.G.S., Prof. Paper 227, pp. 29, 40.

Harada, Z. (1936), Journ. Fac. Sci. Hokkaido Imp. Univ., Sapporo, Japan, 3, Ser. 4, pp. 221-362.

Harker, A. (1932), Metamorphism: London.

Harry, W. T. (1950), Min. Mag., 29, pp. 142-149.

Heinrich, E. Wm. (1946), Am. Journ. Sci., 244, pp. 836-848.

——————————— (1948), Am. Mineral., 33, pp. 420-448; 550-588.

——————————— (1950), Am. Mineral., 35, pp. 173-184.

——————————— (1951), Am. Mineral., 36, p. 634.

——————————— (1952), Bull. Geol. Soc. Amer., 63, p. 1261.

——————————— (1953), Am. Mineral., 38, pp. 68-87.

——————————— and Levinson, A. A. (1953), Am. Mineral., 38, pp. 25-49.

——————————— and Olson, J. C. (1953), U.S.G.S., Prof. Paper 248-G.

Hendricks, S. B., and Jefferson, M. E. (1939), Am. Mineral., 24, pp. 729-771.

Hess, F. L., and Fahey, J. J. (1932), Am. Mineral., 17, pp. 173-176.

Hey, M. H. (1950), An Index of Mineral Species and Varieties Arranged Chemically: British Museum, London.

Hillebrand, W. F., Turner, H. W., and Clarke, F. W. (1899), Amer. Journ. Sci., 157, pp. 451-458.

Hintze, C. (1897), Handbuch der Mineralogie: 2, pp. 516-656. Leipzig.

Holzner, J. (1936), Zeit. Krist., 95, pp. 435-450.

Hutton, C. O. (1940), New Zealand Journ. Sci. Tech., Sec. B, 21, pp. 330-333.

——————————— (1942), Trans. Roy. Soc. New Zealand, 72, pp. 53-68.

——————————— and Seeyle, F. T. (1947), Trans. Roy. Soc. New Zealand, 76, pp. 481-491.

Igelstrom, L. J. (1872), Neues Jahrb., p. 296.

Iimori, S., and Yoshimura, S. (1926), Chem. Soc. Japan, 1, pp. 215-219.

Inoue, T. (1950), Journ. Geol. Soc. Japan, 56, pp. 71-77.

Jackson, W. W., and West, J. (1930), Zeit. Krist., 76, pp. 211-227.

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Jackson, W. W., and West, J.   (1933), Zeit. Krist., 85, pp. 160-164.

Jacobson, G., and Webb, J. S.   (1946), Geol. Surv. of Nigeria, Bull. 17.

Jahns, R. H.   (1945), Geol. Soc. Amer. Bull., 56, p. 1170.

——————— (1946), New Mexico Bur. Mines Min.Res., Bull. 25.

——————— (1952), Bull. Univ. of Arizona, Ariz. Bur. Mines, 23, No. 5
       (Min. Tech. Ser. No. 46, Bull. No. 162).

——————— and Lancaster, F. W.   (1950), U.S.G.S., Prof. Paper 225.

——————— and Wright, L. A.   (1951), Calif. Div. of Mines, Special
       Report 7-A.

Jakob, J.   (1920), Helvetica Chimica Acta, 3, pp. 669-704.

——————— (1921), Zeit. Krist., 56, pp. 295-304.

——————— (1925A), Zeit. Krist., 61, pp. 155-163.

——————— (1925B), Zeit. Krist., 62, pp. 443-453.

——————— (1926), Zeit. Krist., 64, pp. 430-454.

——————— (1927), Schweiz Min. Petrog. Mitt., 7, pp. 139-141.

——————— (1928), Zeit. Krist., 69, pp.217-225.

——————— (1929A), Zeit. Krist., 69, pp. 403-410.

——————— (1929B), Zeit. Krist., 69, pp. 511-515.

——————— (1929C), Zeit. Krist., 70, pp. 493-496.

——————— (1929D), Zeit. Krist., 72, pp. 327-380.

——————— (1931), Zeit. Krist., 79, pp. 367-368.

——————— (1937), Schweiz Min. Petrog. Mitt., 17, pp. 149-153.

——————— (1938), Schweiz Min. Petrog. Mitt., 18, p. 473.

——————— and Parga-Pondal, I.   (1932), Zeit, Krist., 82, pp. 271-
       284.

Johannsen, A.   (1932), A Descriptive Petrography of the Igneous
       Rocks: II, Univ. of Chicago Press, p. 93.

——————— (1937), A Descriptive Petrography of the Igneous
       Rocks: III, Univ. of Chicago Press, p. 35.

Johannsen, A. (1938), A Descriptive Petrography of the Igneous
        Rocks: IV, Univ. of Chicago Press, p. 60.

Jung, H. (1937), Chem. Erde, 11, pp. 38-44.

Kauffman, A. J., Mortimore, D. M., and Hess, H. D. (1950), U. S. Bur.
        Mines, Dept.of Investig. 4721.

Kawano, Y. (1933), Proc. Imp. Acad. Tokyo, 9, pp. 613-616.

——————— (1942), Journ. Jap. Assoc. Mineralogists, Petrologists and
        Econ. Geologists, 27, pp. 283-90.

Kennard, T. G., and Howell, D. H. (1941), Am. Mineral., 26, p. 405.

Kenngott, A. (1860), Ubersicht der Resulate Mineralogischer Forshungen:
        Leipzig.

Kerr, P. F., and Hamilton, P. K. (1949), Prelim. Report No. 1, Amer.
        Petrol. Instit. Proj. 49, Clay Mineral Standards, Columbia Univ.,
        New York.

——————— Anderson, T. P., Rasor, C. A., Pill, R. J., Hamilton, P. K.,
        Wright, H., and Cohen, W. J. (1951), United States Atomic Energy
        Commission, RMO-797.

Knopf, A. (1929), U.S.G.S., Prof. Paper 157, p. 38.

Kokscharov, N. V. (1875), Materialien zur Mineralogie Russlands: 7,
        pp. 222-344, St. Petersburg.

Konta, J. (1950), Rozpravy Ciski Akad., Cl. II, 60, No. 13, 17pp.

Koslov, N. A., and Sokolov, V. A. (1944), Akad. Nauk. Kazakstan S.S.R.
        Izv., 2-3, pp. 82-99.

Kultiassov, S. V., and Dubinkina, R. P. (1946), Mem. Soc. Russe Min.,
        75 (3), pp. 187-192.

Kunitz, W. (1924), Neues Jahrb., Beilage-Band, 50, pp. 365-413.

Lacroix, A. (1922), Mineralogie of Madagascar: Vol. I, p. 476.

Landes, K. K. (1925), Am. Mineral., 10, p. 371.

——————— (1938), Am. Mineral., 33, pp. 359-390.

Larsen, E. S. (1941), U.S.G.S., Prof. Paper 197-A, p. 53.

——————— andBerman, H. (1934), U.S.G.S. Bull., 848, pp. 163-
        237.

Lemke, R. W., Jahns, R. H., and Griffitts, W. R. (1952), U.S.G.S., Prof. Paper 248-B.

Lester, J. G. (1946), Am. Mineral., 31, p. 77.

Levinson, A. A. (1953), Am. Mineral., 38, pp. 88-107.

Liaschenko, C. P. (1940), Mem. Soc. Russe Min., 69 (4), pp. 477-479.

Linck, G. (1901), Zeit. Krist., 35, pp. 315-320.

Lindgren, W. (1933), Mineral Deposits: McGraw-Hill Book Company, Inc., New York.

——————— and Ransome, F. L. (1906), U.S.G.S., Prof. Paper 54.

Lovering, T. S., and Goddard, E. N. (1950), U.S.G.S., Prof. Paper 223.

Lundblad, B. (1942), Geol. Fören. Förh., 64, pp. 55-60.

Macgregor, A. M. (1945), Min. Mag., 27, pp. 157-165.

Machatschki, F. (1930), Cent. Min., Geol. und Paleon., Abt. A, pp. 255-267.

Marginac, C. (1847), Suppl. Bibl. Universe. Genève, Arch. Sci. Phys. Nat., 6, pp. 293-304.

Mauguin, C. (1927), Compt. Rend., 185, pp. 288-291.

——————— (1928A), Compt. Rend., 186, pp. 879-881.

——————— (1928B), Bull. Soc. Fr. Min., 51, pp. 285-332.

Maurice, C. S. (1940), Econ. Geol., 35, pp. 57-67.

Mawson, D., and Dallwitz, W. B. (1945), Trans. Roy. Soc. South Australia, 69, pp. 22-49.

Meixner, H. (1939), Ann. Naturhist. Museums Wien, 50, pp. 694-703.

Merkulova, G. A. (1939), Mem. Soc. Russe Min., 68 (4), pp. 559-561.

Mieleitner, K. (1921), Zeit. Krist., 56, pp. 90-94.

Ming, H. M., and Chang, K. (1935), Mem. Nat. Research Inst. Geol. Acad. Sinica, Naking, No. 15, pp. 15-72.

Miser, H. D., and Stevens, R. E. (1938), Am. Mineral., 23, pp. 104-110.

Murdoch, J., and Webb, R. W. (1938), Am. Mineral., 23, pp. 349-355.

Murray, D. G., and Chapman, F. E. (1931), Journ. Roy. Soc. West Australia, 17, pp. 151-155.

Nagelschmidt, G. (1937), Zeit. Krist., 97, pp. 514-521.

Nockolds, S. R., and Mitchell, R. L. (1948), Trans. Roy. Soc. Edinburgh, 61, pp. 533-575.

Noll, W. (1932), Naturwissenschaften, 20, p. 283.

Ödman, O. H. (1938), Geol. Fören. Förh., 60, H. 2.

———————— (1950), Sveriges Geol. Undersokning, Ser. C, Arsbok, 44, No. 2.

Oftedal, I. (1942), Norsk Geologisk Tidsskrift, 22, pp. 1-14.

Olson, J. C. (1942), U.S.G.S. Bull., 931 P, p. 379.

Orcel, M. J. (1926), Bull. Soc. Fr. Min., 48, pp. 362-366.

Pagliani, G. (1937), Atti. Soc. Ital. Sci. Nat. Mus. Civ. Milano, 76, pp. 13-16.

———————— (1940), Atti. Soc. Ital. Sci. Nat. Mus. Civ. Milano, 79, pp. 20-22.

———————— (1949), Atti. Soc. Ital. Sci. Nat. Mus. Civ. Milano, 88, pp. 191-198.

Palache, C. (1929), Am. Mineral., 14, pp. 43-47.

———————— (1935), U.S.G.S., Prof. Paper 180.

Pardee, L. T., Larsen, E. S., and Steiger, G. (1921), Journ. Wash. Acid. Sci., 11, No. 2.

Park, C. F. (1942), U.S.G.S. Bull., 931-R.

Partridge, F. C. (1937), Trans. Geol. Soc. South Africa, 39, pp. 457-460.

Pauling, L. (1930), Proc. Nat. Acad. Sci., 16, pp. 123-129.

Peacock, M. A., and Ferguson, R. B. (1943), Univ. of Toronto, Geol. Ser., No. 48, pp. 65-82.

Pecora, W. T., Klepper, M. R., and Larrabee, D. M. (1950), U.S.G.S. Bull., 946-C.

Pehrman, G. (1945), Acta Acad. Abo. math. phys., 15 (2), p. 57.

Penfield, S. L. (1893), Amer. Journ. Sci., 46, pp. 288-295.

Pieruccini, R. (1950), Atti. Soc. Toscana Sci. Nat. Mem., 57, Ser. A, pp. 145-175.

Pogue, J. E. (1911), Zeit. Krist., 49, p. 455.

Postel, A. W., and Adelhelm, W. (1944), Am. Mineral., 29, pp. 279-290.

Powell, R. W., and Griffiths, E. (1937), Proc. Roy. Soc., Ser. A, 163, pp. 189-198.

Prider, R. T. (1940), Min. Mag., 25, pp. 373-387.

Prior, G. T. (1908), Min. Mag., 15, p. 385.

Rabbitt, J. C. (1948), Am. Mineral., 33, pp. 263-323.

Ramaseshan, S. (1945), Proc. Indian Acad. Science, 22, Sec. A, pp. 177-181.

Ramberg, H. (1948), Econ. Geol., 43, p. 553.

Rammelsberg, C. (1862), Zeit. Deut. Geol. Gesell., 14, pp. 758-764.

——————— (1878), K. Akad. Wissen., Berlin, Monatsberichte, pp. 613-631.

——————— (1879), K. Akad. Wissen., Berlin, Monatsberichte, pp. 833-853.

Ramsdell, L. S. (1947), Am. Mineral., 32, pp. 64-82.

Rankama, K., and Sahama, T. G. (1950), Geochemistry: Univ. of Chicago Press.

Reusch, E. (1869), Pogg. Ann. Phys. Chem., 138, pp. 628-638.

Rinne, F. (1925), Zeit. Krist., 61, pp. 113-124.

Rowledge, H. P. (1945), Annual Report, Government Mineralogist Analyst and Chemist, Western Australia, for 1943.

Roy, R. (1949), Journ. Amer. Ceram. Soc., 32, p. 202.

Sandberger, F. (1875), Neues Jahrb. für Mineralogie, pp. 624-626.

Sandréa, A. (1950), Bull. Soc. Fr. Min., 73, pp. 446-453.

Savornin, A. (1934), Soc. Fr. Min., Bull. 57, pp. 130-139.

Schafhäutl, K. F. (1842), Lieb. Ann., 44, p. 40.

——————— (1843), Lieb. Ann., 46, p. 325.

Schaller, W. T. (1916), U.S.G.S. Bull., 610, pp. 139-140.

————————— (1930), Am. Mineral., 15, pp. 566-574.

————————— (1950), Min. Mag., 29, pp. 405-415.

————————— and Henderson, E. P. (1926), Am. Mineral., 11. pp. 5-16.

————————— and Stevens, R. E. (1941), Am. Mineral., 26, pp. 541-545.

Scharizer, R. (1886), Zeit. Krist., 12, pp. 1-17.

————————— (1887A), Zeit. Krist., 13, pp. 15-24.

————————— (1887B), Zeit. Krist., 13, pp. 449-473.

Scheerer, T. (1862), Zeit. Deutsch. Geol. Gesell., 14, pp. 23-150.

Serdiuchenko, D. P. (1948A), Akad. Nauk. S.S.S.R., Doklady, 59 (3), pp. 545-548.

————————— (1948B), Akad, Nauk. S.S.S.R., Doklady, 59 (4), pp. 739-742.

Service, H. (1943), Gold Coast Geol. Surv., Mem. No. 5.

Shannon, E. V. (1920), Am. Mineral., 5, pp. 84-86.

Shibata, H. (1952A), Tokyo Bundrika Daigaku, (Tokyo Univ. Lit. Sci.) Geol. Min. Inst., Science Reports, Sec. C, 2, No. 10-11.

————————— (1952B), Tokyo Bundrika Daigaku (Tokyo Univ. Lit. Sci.) Geol. Min. Inst., Science Reports, Sec. C, 2, No. 12.

Simpson, E. S. (1912), Geol. Survey West. Australia, Bull. 48, pp. 95-99.

————————— (1927), Journ. Roy. Soc. West. Australia, 13, pp. 37-49.

————————— (1932), Journ. Roy. Soc. West. Australia, 18, pp. 61-74.

Sollas, W. J. (1889), Proc. Roy Irish Acad., Sec. 3, I, pp. 379-380.

Spence, H. S. (1930), Am. Mineral., 15, p. 438.

Starkl, G. (1883), Jahrb. der K.-K. Geol. Reichsanstalt, Vienna, 33, pp. 653-656.

Sterrett, D. B. (1923), U.S.G.S. Bull., 740, pp. 12, 65, 133.

Stevens, R. E. (1938), Am. Mineral., 23, pp. 607-628.

Stevens, R. E. (1946), U.S.G.S. Bull., 950, pp. 101-119.

Stoll, W. C. (1950), U.S.G.S., Prof. Paper 229, p. 62.

Sudo, T. (1949), Bull. Chem. Soc. Japan, 22, pp. 25-33.

Sugi, K. (1940), Mem. Fac. Sci., Kyusyu Imp. Univ., Ser. D, (1), p. 7.

Svanberg, L. F. (1844), Konglig. Veten. Akad. Stock. Öfver., 1, p. 219.

Taddei, C. (1940), Schweiz Min. Petrog. Mitt., 20, pp. 247-252.

Tchirvinskii, P. N. (1948), Mem. Soc. Russe Min., 77 (3), pp. 246-9.

Thiébaut, J. L. (1925), Contribution a l'etude des Sediments Argilo-
        calcaires du Bassin de Paris: Nancy. Cit. in Min. Mag., 24, p. 621.

Thomson, T. (1828), Annals of the Lyceum of Natural History of New
        York, 3, p. 47.

——————————⸴ (1836), Outlines of Mineralogy, Geology, and Mineral
        Analysis: 2 volumes, London.

Tolansky, S. (1948), Multiple-Beam Interferometry of Surfaces and
        Films: The Clarendon Press, Oxford.

Tschermak, G. (1877), Zeit. Krist., 2, pp. 14-50.

——————————— (1878), Zeit. Krist., 3, pp. 122-167.

Turin, B. A. (1944), Akad. Nauk. U.S.S.R. Izv., Ser. Geol. 2.

vanBiljon, S. (1940), Trans. Geol. Soc. South Africa, 42, pp. 123-219.

Venkayya, E. (1949), Proc. Indian Acad. Science, 30, Sec. A, pp. 74-77.

Volk, G. W. (1939), Am. Mineral., 24, pp. 255-266.

Wagner, P. A. (1921), Trans. Geol. Soc. S. Africa, 24, p. 73.

——————————— (1928), Geol. Surv. Union of South Africa, Mem. 26.

Wahl, W. (1925), Fennia, 45 (No. 20), 127 pp.

Walker, T. L., and Parsons, A. L. (1924), Univ. of Toronto Studies,
        Geol. Series, No. 17, pp. 46-50.

——————————————————————————— (1926), Univ. of Toronto Studies,
        Geol. Series, No. 22, pp. 5-14.

Webb, R. W. (1939), Am. Mineral., 24, pp. 123-129.

Weinschenk, E. (1896), Zeit. Krist., 26, p. 471.

Wells, R. C., and Brannock, W. W. (1946), U.S.G.S. Bull., 950, pp. 121-127.

Wherry, E. T. (1915), Proc. U. S. Nat. Mus., 49, pp. 463-467.

Whitmore, D. R. E., Berry, L. G., and Hawley, J. E. (1946), Am. Mineral., 31, pp. 1-21.

Wijkerslooth, P. (1943), Maden Tetkik ve Arama Eustitusu Mecmuasi, Ankara, 8, pp. 91-101.

Winchell, A. N. (1925), Amer. Journ. Sci., 209, pp. 309-327; 415-430.

———————— (1927), Am. Mineral., 12, pp. 267-279.

———————— (1932), Am. Mineral., 17, pp. 551-553.

———————— (1935), Am. Mineral., 20, pp. 773-779.

———————— (1942), Am. Mineral., 27, pp. 114-130.

———————— and Winchell, H. (1951), Elements of Optical Mineralogy: Part II, New York.

Wolff, J. E., and Palache, C. (1902), Zeit. Krist., 36, pp. 438-448.

Wood, W. A. (1937), Proc. Roy. Soc., Ser. A, 163, pp. 199-204.

Woodward, H. H. (1951), Am. Mineral., 36, pp. 869-883.

Wright, F. E. (1914), Amer. Journ. Sci., 38, pp. 305-308.

Wülfing, E. A. (1886), Ber. Deutschen Chem. Gesell., 19, pp. 2433-2438.

Yamada, H., and Sugiura, K. (1950), Journ. Jap. Assoc. Mineralogists, Petrologists and Econ. Geologists, 34, pp. 122-128.

Yoshimura, T. (1939), Journ. Fac. Sci., Hokkaido Imp. Univ., Ser. 4, Vol. 4, No. 3-4, pp. 313-452.

Zambonini, F. (1919), Mem. Descr. Carta Geol. Italia, 7, Part 2, p. 124.

———————— (1922), Rend. Accad. naz. Lincei, Ser. 5, 31 (1), pp. 147-151.

Zealley, A. E. V. (1917), Proc. Rhodesia Sci. Assoc., 16, pp. 17-23.