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QUARTERLY REPORT

NATURAL MICA STUDIES

(Covering period December 1, 1952, to February 28, 1953)

By

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INTRODUCTION

During the quarterly period December 1, 1952, to February 28, 1953, several additional phases of research on the natural micas have been completed. These include a study of the zinnwaldites and the problem of zoning and overgrowths in all mica species. The work on both topics consisted of an extensive survey of the literature supplemented by new data obtained from specimens in the University of Michigan mica collections.

With the acquisition and completion of x-ray studies on several critical specimens, the manuscript "Studies in the mica group; The structure of the high-silica muscovites (phengites)" by E. Wm. Heinrich and A. A. Levinson was completed. Work has also been completed on the study of roscoelite and oelacherite, and the manuscript for this study is practically complete. Electron photo-micrographs of several alleged specimens of barium-and mangan-muscovites were taken and are now being interpreted. It is planned to submit both papers for publication in the near future.

Since the publication of the conclusions on the relationship between chemistry and composition in the muscovite-lepidolite series, interest in our work has been aroused at the Corning Glass Works, Corning, New York. Recently a Corning representative visited our laboratory to discuss the subject with particular reference to their problem of obtaining a source of lepidolite with a consistent  $\text{Li}_2\text{O}$  content for glass manufacture. As a result of these discussions the Corning Company will analyze several specimens in the muscovite-lepidolite range, and we shall study them structurally.

During this quarterly period a study of the world-wide distribution, geologic relationships, and paragenesis of all known lepidolite and lithium-bearing muscovite ( $> 2.0\% \text{Li}_2\text{O}$ ) deposits has been completed. Emphasis was

placed on paragenesis and on recommendations for potentially economically valuable deposits.

It was hoped that the x-ray study of the dark-colored micas would be completed by the end of the quarter, but about 20 biotites from extrusive rocks still remain to be studied. The structures of all analyzed specimens, however, have been determined and are discussed in this report.

During the next quarter, in addition to completion of the biotite study, the subject of mica symplectites will be investigated. Thin sections are now being made of about a dozen specimens showing symplectic intergrowths. Work will also begin on several initial phases of the final report. This will include cataloguing the many x-ray films and compiling data. A comprehensive study of manganophyllites is tentatively scheduled for completion, and if the results of the final group of spectrochemical analyses become available, a study of trace elements in micas may be ready.

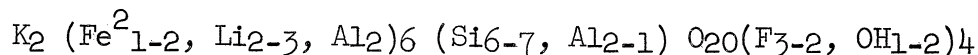
#### STRUCTURE OF ANALYZED BIOTITES, PHLOGOPITES, AND MANGANOPHYLLITES

In the First Annual Report, mention was made of the work being carried on in an attempt to correlate chemistry, polymorphism, and paragenesis in the dark-colored micas. This work has been completed with the determination of the structures of the approximately 60 analyzed micas in this category. Most specimens have been described in the literature and were received from foreign investigators. The results of the study are presented in Table 1. A survey of the data will indicate that there is no evident relationship between chemistry, polymorphism, and paragenesis among the dark-colored micas. In the near future an attempt will be made to correlate the cell dimensions with chemistry.

#### A SUMMARY OF THE MINERALOGY AND PARAGENESIS OF ZINNWALDITE

##### Crystal Chemistry

General Composition. The composition of zinnwaldite may be expressed by the following general formula:



The (Al, Fe, Li) group may be considerably deficient. The possible ionic substitutions that can take place are as follows:

TABLE 1

## STRUCTURE OF ANALYZED BIOTITES, PHLOGOPITES, AND MANGANOPHYLLITES

## ANALYZED BIOTITES

Number	Reference	Structure	Paragenesis	FeO	Fe <sub>2</sub> O <sub>3</sub>	MgO	TiO <sub>2</sub>
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	vanBiljón (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

TABLE 1 (cont.)

Number	Reference	Structure	Paragenesis	FeO	Fe <sub>2</sub> O <sub>3</sub>	MgO	TiO <sub>2</sub>
1117	Coats and Fahey (1944)	1-layer (mod. scattering)	pegmatite (siderophyllite)	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 Seelye (1947)	2-layer	pegmatite-like lense	14.41	3.92	11.11	3.02
1262	Mauguin (1928)	1-layer (heavy scattering)	from Tischebarkul	12.77	7.09	13.30	1.16
1350	Yamada and Sugirua (1950)	1-layer (weak scattering)	pegmatite	3.49	0.67	24.24	0.64

TABLE 1 (cont.)

## ANALYZED PHLOGOPIITES

Number	Reference	Structure	Paragenesis	FeO	Fe <sub>2</sub> O <sub>3</sub>	MgO	TiO <sub>2</sub>	F <sub>2</sub>
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	Skråbböle, Pargas, Finland	0.58	1.92	28.18	1.27	2.12
1071	Jakob-not published	1-layer	Finland	0.71	1.01	27.80	0.12	2.80
1072	Jakob-not published	1-layer	Pargas, Finland	5.59	1.54	22.00	1.41	1.87
1073	Jakob-not published	1-layer	Patteby, Pargas, Finland	1.47	1.29	26.16	0.33	1.87

TABLE 1 (cont.)

Number	Reference	Structure	Paragenesis	FeO	Fe <sub>2</sub> O <sub>3</sub>	MgO	TiO <sub>2</sub>	F <sub>2</sub>
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	Skråbböle, 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 Seelye (1947)	1-layer	from marble, New Zealand	2.38	0.43	22.95	0.82	0.62
1252	Pieruccini (1950)	1-layer	pneumatolytic, Mt. Somma	7.89	Tr.	15.66	0.33	2.57
1261	(a) Mauguin (1928) (b and c) Mauguin (1928)	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
731								
1325	Cross (1897)	1-layer	in Wyomingite	0.90	2.73	22.40	2.09	1.03

TABLE 1 (cont.)

## ANALYZED MANGANOPHYLLITES

Number	Reference	Structure	Paragenesis	FeO	Fe <sub>2</sub> O <sub>3</sub>	MgO	MnO	Mn <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>
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
726	Kauffman, et al. (1925)	1-layer	pegmatite						
					-----	Spectrographic	-----		



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<sup>2</sup>, Fe<sup>3</sup>, 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<sub>2</sub>O = 0.10%, and Rb<sub>2</sub>O = 1.04%. Other trace elements reported in zinnwaldites are P, He, Mg, Mn<sup>3</sup>, Ga, Ba, Sc, Tl, and Y (Rankama and Sahama, 1950).

### Polymorphism

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, Bavaria; and Brambach, Saxony. They also report a 3-layer hexagonal structure zinnwaldite from Amelia, Virginia. The x-ray data of 14 zinnwaldites studied in the Mineralogical Laboratory of the University of Michigan are tabulated in Table 2. 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. The significance of these reflections has been discussed in detail by Hendricks and Jefferson (1939) and Levinson (1953). 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. These reflections should be absent in the ideal mica structure on the basis of structure-factor calculations. 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 a distortion, the extent and nature of which are not known. Muscovite has only two-thirds of the octahedral positions filled, whereas the ideal heptaphyllite micas have all these positions occupied. Therefore, the presence of  $06l$  reflections with  $l$  odd recorded on the Weissenberg photograph of Spec. 1251

TABLE 2

## NEW STRUCTURAL STUDIES OF THE ZINNWALDITES

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
780	1-layer	ND	Green-black	Fredrickstown, Mo.	American Museum
786	1-layer	Medium	Pale brown	Amelia, Va.	Harvard
787(a)	3-layer	ND	Pale brown	Amelia, Va.	Analyzed by Glass (1935)
1128	1-layer	ND	Rust brown	Martha Mine, Pongo. Bolivia	U. S. Nat'l Musuem R8089
1242(a)	1-layer	Medium	Brown	Amelia, Va.	outer edge zone
1242(b)	3-layer	Small	Dark brown	Amelia, Va.	inner zone
1251	2-layer	ND	Pale brown	Ubini, W. Australia	Murry and Chapman (1931)
1324	3-layer	ND	Dark brown	Brown Derby, Colorado	
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
1337(a)	3-layer	Small	Brown	Brown Derby, Colorado	Core zone
1337(b)	3-layer	Small	Dark brown	Brown Derby, Colorado	Margin zone

\*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

implies that there is distortion in the mica structure and that this zinnwaldite approaches the heptaphyllites in structure. This appears to be the only specimen of zinnwaldite which can be definitely shown to be more closely related structurally to heptaphyllite micas than to the octaphyllites. Noteworthy are the facts that the deficiencies in the Al, Fe, Li (octahedral) position have been reported, and that Lemke, et al. (1952) report the occurrence at Amelia, Virginia, of many books intermediate between zinnwaldite and muscovite and some showing gradations from muscovite centers to zinnwaldite borders. This would be expected if zinnwaldite were to approach a heptaphyllite composition.

The intensities of the  $06l$  reflections with  $l$  odd 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  $0kl$  reflections are listed in Table 3.

TABLE 3

APPROXIMATE OBSERVED INTENSITIES  
OF SOME ( $0kl$ ) REFLECTIONS OF 2-LAYER FORMS

Plane	Normal Muscovite	2-Layer Zinnwaldite	Lithium 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

Optics

In Table 4 are listed the optical constants of zinnwaldites as reported in the literature. The range of values is as follows:

$$\alpha = 1.535-1.558$$

Axial plane: parallel with (010)

$$\beta = 1.570-1.589$$

Color: gray to amber-brown

$$\gamma = 1.572-1.590$$

Sp. Gravity: 2.916-3.018

$$2V = 0-70^\circ$$

Average 25-30°

An attempt was made to correlate the optical data with the chemistry. Unfortunately only three analyzed zinnwaldites have optical determinations sufficiently precise for plotting, which prevents any definite conclusion. There does, however, appear to be a general increase in the indices with an increase in iron content.

Single-Crystal Variations

Baumhauer (1879) reported a zoned zinnwaldite from Zinnwald, Bavaria. According to his descriptions, crystals of zinnwaldite showed zones of different widths normal to an "A" structure. These zones were yellow, whereas the rest of the crystal was colorless to gray. He postulated the zoning to be due to chemical differences. Upon examination under polarized light he found the various zones to have the same adsorption formulae. 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  $44^\circ 04'$  and a border zone having an angle of  $52^\circ 06'$ .

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  $3-4^\circ$ ,  $\beta = 1.584$ , and  $\gamma = 1.586$ , and crystallized as the 3-layer form. The marginal zone had a  $2V$  of  $28^\circ$ ,  $\beta = 1.583$ ; and  $\gamma = 1.588$ , 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

TABLE 4

## OPTICAL PROPERTIES OF ZINNWALDITES

Locality	$\alpha$	$\beta$	$\gamma$	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'		Durfeld, 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 <u>+0.002</u>	1.576 <u>+0.002</u>	2V = 31° <u>+3°</u>		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

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 2 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 strongly zoned crystals. The zoning of these micas is a color zoning with a light outer zone and dark inner zone. Both zones gave the same optical results:  $\beta = 1.584$ ,  $\gamma = 1.585$ , and  $2V = 0.2^\circ$ . There are neither optical changes nor structural changes with respect to the zoning in zinnwaldite specimens from this locality.

No reference to a chemical study of any of the zoned crystals was found in the literature, although most writers agree that slight chemical differences are probably the main reason for zoning in the zinnwaldites.

Lamke, Jahns, and Griffitts (1952) 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 concluded that the zinnwaldite formed later than muscovite and in some books formed rims around cores of muscovite.

#### General Paragenesis

A 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 (Shibate, 1952).

ZONING IN MICAS

Many species of mica, particularly those of pegmatitic origin, display zoning of various types in physical, optical, or structural properties within a single crystal. Several of these properties are interrelated. Primary types include those formed during the period of crystallization, whereas secondary types of zoning result from changes occurring after the period of crystallization. The information on different types of zoning in micas given below has been drawn both from the literature and from the examination of specimens in our mica collections.

Color Zoning

The type of zoning that is the most easily recognized is color zoning, which may or may not be accompanied by significant differences in indices,  $2V$ , etc., in the various zones. Cleavage flakes of mica often show concentric bands displaying slightly different colors. These bands are parallel with the prism faces (110) in the diamond-shaped crystals, parallel with the prism and clinopinacoid in the pseudo-hexagonal crystals, or parallel with the margin of an irregular crystal. Five different varieties have been noted. They are:

1. Core and Margin. Two distinct zones parallel to the crystal outline or to an irregular outline. The zonal boundary may be sharp or gradational. Visible on the cleavage faces.
2. Oscillatory Zoning. Alternating bands of two different colors. Generally the boundaries are sharp. Zones are parallel with a diamond or pseudo-hexagonal shape. Visible on cleavage faces.
3. Varicolored Zoning. Several shades of color are involved. From core to margin, each zone tends to become darker or in some specimens lighter than the preceding zone. Visible on the cleavage faces.
4. Three-Dimensional Color Zoning. Two shades of color are involved. The cleavage plates at one end of a crystal are one color, whereas those at the other end are of another color. The two colors grade into one another along the length of the c-axis.
5. Crosshatch Zoning. This type appears as linear streaks associated with two sets of reeves or as a plaid pattern resulting from intersecting spots and patches. Visible on the cleavage faces.

### Optical Zoning

Optical zoning without accompanying color zoning is inconspicuous and hence not easily recognized. Variation in indices of refraction, birefringence, size of optic angle, or optic orientation indicate the presence of optical zoning. Since each differently colored zone generally has slightly different optical properties, optical zones often correspond to color zones.

### Megastructural Zoning

Megastructural zoning results from the systematic distribution of reeves, ruling, "A" structure, or wedging. Reeves are lines or shallow corrugations that lie in the plane of cleavage and are oriented perpendicular to the prismatic or clinopinacoid faces. "A" structure has two sets of reeves intersecting at  $60^\circ$  along the edges of a V-shaped fragment. The interior portion of the "A" may or may not show other types of zoning. Some "A" mica will have color zones developed normal to the reeves. Rulings are parting planes perpendicular to the prismatic or clinopinacoid faces. Wedging is caused by interlayering of unequal-sized sheets and is commonly associated with "A" mica. In many books these imperfections do not extend over the entire width of the crystal but are concentrated either in central or in marginal parts.

### Polymorphic Zoning

The most difficult zoning to detect is that showing variation in polymorphic types. Generally, it is marked by an irregular variation in the crystal structure across the basal cleavage face.

### Inclusion Zoning

Zoning of inclusions is seen either as inclusion-filled cores surrounded by inclusion-free margins, as an alternation of inclusion-rich zones with inclusion-free zones, or as irregularly distributed zones of inclusions. All three types are evident on cleavages. The inclusions occur as needles, flattened crystals, or irregular spots and are commonly crystallographically oriented. As many as 30 different minerals have been reported as inclusions in mica. Frondel (1940) states that the inclusions may have been trapped on a crystal face and buried by continual growth of the mica crystal. Some inclusions are thought to be the result of exsolution. Still others may be secondary.



### Three-Dimensional Zoning

Variation in color along the c-axis has been mentioned above. In addition, variations in optics and size and shape of the zonal pattern are found.

### Zoning in Muscovite

Color zoning is a rather common phenomenon in muscovites of pegmatitic origin. Zoning of different types has been reported in muscovites from the following localities:

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 run-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 megastructural 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 zoning was selected from our mica collection for optical study. Indices of refraction were measured on an Abbe refractometer and  $2E$  was determined by the Mallard equation. The optical data are listed below.

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No. 5. Gregory Mine, Jackson County, North Carolina

green core	clear margin
$\alpha = 1.562$	$\alpha = 1.565$
$\beta = 1.598$	$\beta = 1.602$
$\gamma = 1.603$	$\gamma = 1.612$
$2E = 72^\circ$	$2E = 67^\circ 20'$

No. 7. Jasper Mica Mine, Jackson County, North Carolina

dark-brown core	medium-brown intermediate zone	light-brown margin
$\alpha = 1.565$	$\alpha = 1.564$	$\alpha = 1.566$
$\beta = 1.602$	$\beta = 1.601$	$\beta = 1.601$
$\gamma = 1.606$	$\gamma = 1.605$	$\gamma = 1.605$
$2E = 67^\circ 25'$	$2E = 65^\circ 15'$	$2E = 62^\circ 40'$

No. 9. Franklin-Sylva District, North Carolina

clear core	light-brown margin
$\alpha = 1.569$	$\alpha = 1.572$
$\beta = 1.607$	$\beta = 1.612$
$\gamma = 1.613$	$\gamma = 1.615$
$2E = 54^\circ 10'$	$2E = 59^\circ 15'$

No. 15. Franklin-Sylva District, North Carolina

stained core	clear margin
$\alpha = 1.566$	$\alpha = 1.567$
$\beta = 1.602$	$\beta = 1.604$
$\gamma = 1.608$	$\gamma = 1.608$
$2E = 61^\circ 40'$	$2E = 60^\circ 10'$

No. 46. Franklin-Sylva District, North Carolina

stained core	clear margin
$\alpha = 1.562$	$\alpha = 1.565$
$\beta = 1.598$	$\beta = 1.601$
$\gamma = 1.607$	$\gamma = 1.607$
$2E = 62^\circ 30'$	$2E = 63^\circ 30'$

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No. 57. Franklin-Sylva District, North Carolina

medium-brown core	clear margin
$\alpha = 1.560$	$\alpha = 1.560$
$\beta = 1.595$	$\beta = 1.593$
$\gamma = 1.604$	$\gamma = 1.607$
$2E = 70^{\circ}00'$	$2E = 69^{\circ}45'$

No. 64. Franklin-Sylva District, North Carolina

clear core	spotted margin
$\alpha = 1.564$	$\alpha = 1.559$
$\beta = 1.601$	$\beta = 1.596$
$\gamma = 1.607$	$\gamma = 1.606$
$2E = 65^{\circ}30'$	$2E = 63^{\circ}$

No. 75. Franklin-Sylva District, North Carolina

light-brown core	medium-brown margin
$\alpha = 1.562$	$\alpha = 1.566$
$\beta = 1.599$	$\beta = 1.601$
$\gamma = 1.606$	$\gamma = 1.606$
$2E = 65^{\circ}30'$	$2E = 65^{\circ}40'$

No. 82. Franklin-Sylva District, North Carolina

light-brown core	medium-brown margin
$\alpha = 1.565$	$\alpha = 1.566$
$\beta = 1.599$	$\beta = 1.603$
$\gamma = 1.605$	$\gamma = 1.606$
$2E = 64^{\circ}50'$	$2E = 64^{\circ}50'$

No. 124. Spruce Pine District, North Carolina

medium-green core	light-green margin
$\alpha = 1.570$	$\alpha = 1.569$
$\beta = 1.610$	$\beta = 1.603$
$\gamma = 1.615$	$\gamma = 1.609$
$2E = 64^{\circ}15'$	$2E = 67^{\circ}10'$

No. 128. Spruce Pine District, North Carolina

dark-green core	light-green margin
$\alpha = 1.570$	$\alpha = 1.569$
$\beta = 1.610$	$\beta = 1.606$
$\gamma = 1.616$	$\gamma = 1.616$
$2E = 70^{\circ}00'$	$2E = 67^{\circ}30'$

No. 190. Muscovite Claim, Latah County, Idaho

light-brown portion	clear portion
$\alpha = 1.562$	$\alpha = 1.562$
$\beta = 1.598$	$\beta = 1.599$
$\gamma = 1.602$	$\gamma = 1.604$
$2E = 63^{\circ}15'$	$2E = 63^{\circ}$

No. 374. Hebron, Maine

medium-tan core	light-tan intermediate zone	clear margin
$\alpha = -$	$\alpha = 1.563$	$\alpha = 1.560$
$\beta = 1.594$	$\beta = 1.598$	$\beta = 1.591$
$\gamma = 1.603$	$\gamma = 1.601$	$\gamma = 1.598$
$2E = 67^{\circ}10'$	$2E = 65^{\circ}15'$	$2E = 61^{\circ}50'$

The above data indicate that slight differences in index of refraction and size of the optic angle accompany variation in color. The darker-colored zones and those that are stained or contain inclusions tend to have higher indices and a larger optic angle. However, no general tendency is apparent in the order of zoning from this limited number of specimens. A zoned muscovite may have either a lighter or a darker core than the surrounding border. No quantitative correlation has yet been attempted between chemical composition and color in the muscovites. Olson (1942) states that green muscovite probably contains more iron than ruby. Heinrich and Levinson (1953) correlated the pink color in muscovite with an absence of  $Fe^{2+}$  and an equality or predominance of  $Mn^{3+}$  to  $Fe^{3+}$ . The chromophores in muscovite are  $Fe^{2+}$ ,  $Fe^{3+}$ ,  $Mn^{3+}$ , Cr, and, rarely, Ti.

### Zoning in Lepidolites

Polymorphic zoning with associated optical properties is the only type recorded in lepidolites. The following specimens, described in Quarterly Report No. 2, show optical and polymorphic variation:

	biaxial portion	uniaxial portion
No. 539	1-layer	3-layer rhombohedral
No. 679	2-layer lithian muscovite	3-layer rhombohedral
No. 476	1-layer	3-layer monoclinic

The zoning is irregularly distributed as spots or patches on the basal cleavage. Only specimen No. 539 examined showed any systematic shape or distribution of zoning. It has been shown that the variation in polymorphic forms is related to a slight variation in the  $\text{Li}_2\text{O}$  content.

### Zoning in Biotite

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 and usually has weak pleochroism.

Johannsen (1948) notes the occurrence of color-zoned biotite in minette, a lamprophyre of the syenite series composed of biotite 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 zoned structure.

Over 100 specimens of biotite in the mica collections were examined for zoning. None of the biotite of pegmatitic origin showed zoning of any type. One thin section of minette from Freiberg, Saxony, contained phenocrysts of zoned biotite as described by Johannsen. Apparently zoning in biotite is very uncommon and is restricted to the phaneritic and porphyritic igneous rocks.

### Zoning in Siderophyllite

None of the specimens of siderophyllite in the mica collection displays zoning. However, two occurrences have been described in the literature.

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Clarke (1887) describes a bronzy black mica, from Pikes Peak, Colorado, "... resembling phlogopite externally, ... but made up of a core composed of a soft rotten material, evidently 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 <sub>2</sub> O	4.54	7.28
SiO <sub>2</sub>	34.21	34.63
Al <sub>2</sub> O <sub>3</sub>	16.53	17.95
Fe <sub>2</sub> O <sub>3</sub>	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 <sub>2</sub> O	1.43	0.89
K <sub>2</sub> O	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  $2V = 6^{\circ}-8^{\circ}$  surrounded by a discontinuous rim of pale-blue mica having a double refraction of 0.028 and  $2V = 37^{\circ}$ .

Zoning in Phlogopite

Orcel (1924) 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	Project M978
$n_p = 1.5529$	$\alpha = 1.552$
$n_g = 1.5910$	$\beta = 1.592$
	$\gamma = 1.593$
uniaxial	$2V = 2-3^{\circ}$

Both color zones have essentially the same optical properties.

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Larsen (1941) reports color-zoned phlogopite of hydrothermal origin associated with aegirine and sodic amphibole in contact-metamorphosed limestone. He states that the micas vary moderately in indices of refraction. 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:

Specimen IH - 2

core:  $\alpha = 1.576$      $\beta = 1.613$      $\gamma = 1.613$   
 pleochroism  
 $\alpha$ , nearly colorless     $\beta$  and  $\gamma$ , faint brown

intermediate zone:  $\alpha = 1.576$      $\beta = 1.613$      $\gamma = 1.614$   
 pleochroism  
 $\alpha$ , deep chestnut brown     $\beta$  and  $\gamma$ , nearly colorless

outer zone:  
 pleochroism  
 $\alpha$ , deep chestnut brown     $\beta$  and  $\gamma$ , 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.

Specimen 1369 from Thorne Township, Quebec, Canada, (No. 462A-11) on loan from Cranbrook Institute) 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.

<u>Zone</u>	<u><math>\alpha</math></u>	<u><math>\beta</math></u>	<u><math>\gamma</math></u>	<u>2V</u>	<u>Color</u>
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.

Specimen 1370, an irregularly shaped specimen from Canada, has parallel arrangement of light-brown inclusion-free zones alternating with darker-brown zones containing numerous inclusions of rutile. Both types are essentially uniaxial. The inclusion-free zones show no asterism and yield a sharp



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optic axis figure whereas the zones bearing inclusions show strong asterism and yield an extremely diffuse figure. Indices of refraction for each zone are:

	<u><math>\alpha</math></u>	<u><math>\beta</math> and <math>\gamma</math></u>
Clear zones	1.541	1.570
Zones bearing inclusions	1.544	1.578

Numerous specimens of Canadian phlogopite in the mica collection commonly display concentric color zones. Generally, the optical properties for 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.

Correlation between color and chemical composition has been attempted by Hall (1941) and Kennard and Howell (1941). The relative amounts of  $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$  and Ti are responsible for the different colors in biotite. Ferrous iron gives the green colors and titania produces red and brown colors, but magnesia tends to mask the colors produced by titania according to Hall (1941), however, it seems unlikely that a nonchromophore such as Mg could seriously influence the pigmenting power of such elements as  $\text{Fe}^{2+}$  and Ti. Ferric iron imparts shades of yellow and red; whereas a combination of  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  causes shades of blue. Small amounts of  $\text{Fe}^{3+}$  and  $\text{Ti}^{4+}$  have a greater effect on indices of refraction than a comparable amount of  $\text{Fe}^{2+}$ .

As Hall points out, color does not give a good indication of the chemical composition. However, a darker-colored zone showing higher indices of refraction indicates an increase in  $\text{Fe}_2\text{O}_3$  or  $\text{TiO}_2$ .

TABLE 5

## TYPES AND RELATIVE ABUNDANCE OF ZONING IN MICAS

Mineral	Type of Zoning	Frequency	Occurrence
Muscovite	color-indices	common	pegmatite
	inclusion-indices	common	pegmatite
	megastructural	very common	pegmatite
Lepidolite	polymorphic	not uncommon	pegmatite
Zinnwaldite	color	rare	pegmatite
	optical	rare	pegmatite
	megastructural	rare	pegmatite
	polymorphic	rare	pegmatite
Phlogopite	color	common	pegmatite
	inclusions	common	pegmatite
	optical	common	pegmatite
Biotite	color	uncommon	igneous rocks
	optical	uncommon	igneous rocks
	polymorphic	uncommon	igneous rocks

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