

THE MICA GROUP

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INTRODUCTION

After feldspars and quartz, micas are the third most abundant constituent of many pegmatite types. The significance of mica occurrences in pegmatites is manifold. Ferromagnesian micas are commonly the only mafic minerals in the more primitive pegmatite types, and "white" micas are usually the main component responsible for the peraluminous nature of pegmatite bulk compositions. Pegmatitic micas are quite diverse in their chemical composition, typomorphic properties and mineral associations, and they contribute several unique varieties to the structural and crystallochemical spectrum of the mica group. Individual mica types are characteristic of different stages of pegmatite evolution, and their generation, equilibrium and/or reaction relations with other phases aid in delineating pegmatite crystallization paths through PTX space. Micas also serve as indicators of the economic potential of their parent pegmatites or internal pegmatite units. Last but not least, the mica minerals themselves have a variety of industrial uses, from optical, dielectric and construction applications to the extraction of Li, Rb, Cs and Ga.

This paper presents a brief introduction to the mica structure and crystal chemistry, and a review of different paragenetic types of micas in granitic pegmatites. Physical properties are mostly omitted from the following discussion, being a matter of common knowledge for most mica types. Optical properties are highly variable, reflecting mainly the octahedral substitutions and the OH/F ratio. The refractive indices and 2V values span a characteristic range for each substitutional series, and they are convenient as well as relatively accurate for mica determination. Detailed information is available in Winchell (1951), Tröger (1962), Rieder et al. (1971), Rinaldi et al. (1972) and Chaudhry and Howie (1973b).

STRUCTURE

The Ideal Mica

An essential feature of the mica structure is a sheet of corner-linked tetrahedra (Fig. 1a) that extends continuously in two directions and has the general stoichiometry $(T_2O_5)_\infty$. In the idealized form of Figure 1a, this sheet resembles a series of linked hexagonal rings of tetrahedra, in which the apical T-O bond always points in the same direction. Adjacent sheets in which these apical bonds oppose each other are linked by medium-sized (ionic radius 0.53–0.83Å) univalent to tetravalent cations to form a composite layer or sandwich (Fig. 1b). The tetrahedral sheets are staggered such that the apical oxygens assume a pseudo-octahedral arrangement around the linking cations; to complete the cation coordination, monovalent anions lie in the plane of the apical oxygens such that the coordination of the cations is pseudo-octahedral. Thus the composite layer can be considered as a sheet of edge-sharing octahedra sandwiched between two sheets of corner-sharing tetrahedra. These composite layers are linked by large univalent and divalent cations that in the ideal structure are surrounded by twelve anions. The resulting structure has the overall stoichiometry $X_2^{II}Y_4^{VI}T_8O_{20}(OH,F)_4$, is strongly bonded within the composite $[Y_4T_8O_{20}(OH,F)_4]$ layer and relatively weakly bonded between composite layers. In the general formula, if $Y=4$ (that is two of the three possible octahedrally-coordinated positions are occupied) the mica is dioctahedral; if $Y=6$ (all three of the possible octahedrally-coordinated positions are occupied) the mica is trioctahedral.

Polytypism. Polytypism is the ability of a substance to exist in more than one structural form as a result of different stacking sequences of identical layers. Stacking of the composite layers of the mica structure gives rise to different polytypes. In a single layer, a stacking-vector can be drawn from the centre of an hexagonal ring in the lower tetrahedral sheet to the centre of an hexagonal ring in the upper tetrahedral sheet. The back-to-back hexagonal rings of tetrahedra in adjacent layers must superimpose in order to provide the necessary coordination for the large interlayer cation. Thus succeeding layers can each assume six possible configurations, not all of which are unique. The interlayer stacking angle is defined as the angle between the stacking vectors in adjacent sheets, projected into the plane of the layer. Smith and Yoder (1956) have shown that there are only six possible polytypes with periodicities between one and six layers, provided only one interlayer stacking angle (positive or negative) is allowed in a given polytype. The stacking possibilities giving rise to these polytypes are shown in Figure 2, and the crystallographic data for these polytypes is given in Table 1. The polytype symbol indicates the number of layers and the lattice symmetry of the structure, with subscripts to distinguish structures with the same periodicity and lattice symmetry. Table 2 shows the polytypes reported for the dioctahedral and

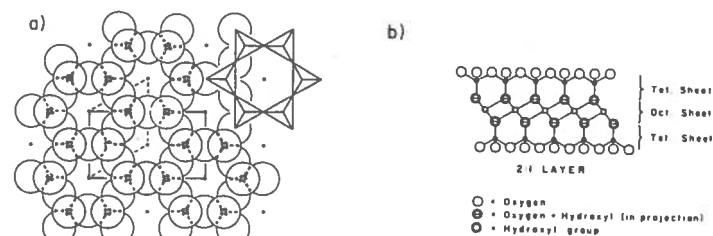


Fig. 1. (a) Plan view of the ideal $[T_2O_5]_\infty$ tetrahedral sheet. (b) Cross section of composite layer $[Y_4-6T_8O_{20}(OH)_4]_\infty$.

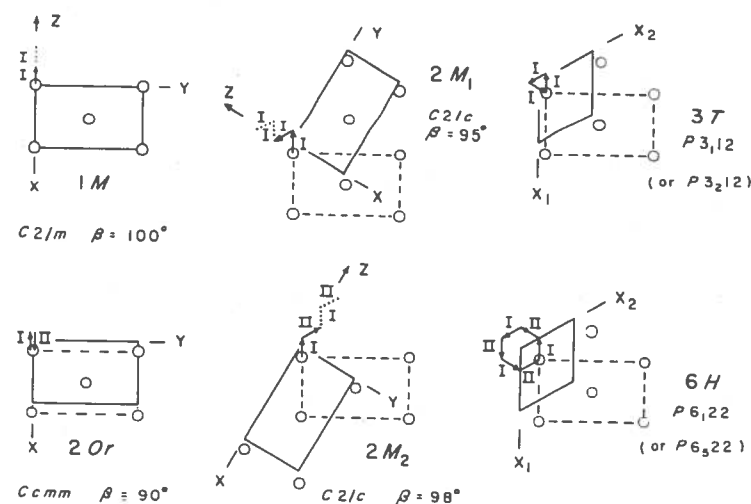


Fig. 2. Derivation of the six ideal mica polytypes. The dashed lines show the unit cell for the first layer, and the full lines show the unit cell of the resultant polytype. Polytype symbol and ideal space group are shown. From Bailey (1980).

TABLE 1. CRYSTALLOGRAPHIC DATA FOR THE IDEAL MICA POLYTYPES

Polytype	$\sim a(\text{\AA})$	$\sim b(\text{\AA})$	$\sim c(\text{\AA})$	$\sim \beta^\circ$	Space Group
1M	5.25	9.10	10.20	100	C2/m
2M ₁	5.25	9.10	20.00	95	C2/c
2M ₂	9.00	5.20	20.00	100	C2/c
20r	5.50	9.50	20.00	90	Ccmm [*]
3T	5.20	5.20	30.00	$\gamma=120$	P3 ₁ 12
6H ⁺	5.20	5.20	60.00	$\gamma=120$	P6 ₁ 22

* only the derivative space group Pnmn has been found in this type

+ this polytype has not yet been found.

trioctahedral micas; there does seem to be some compositional control on polytype occurrence, and these structures are more accurately described as polytypoids. Disordered stacking sequences (termed 1Md) are also observed, and appear to be more common in the trioctahedral micas. More complex polytypes do occur, and have been considered by Ross et al. (1966), Takeda (1971) and Takeda and Ross (1975) among others.

Structural Distortions

In the ideal mica structure described above, the dimensions of the tetrahedral and octahedral sheets are such that they can link together in this ideal configuration. In real structures, the sheets are not of these ideal dimensions and must distort from their ideal configurations if they are to link together into composite layers. There are two principal ways in which this may be done:

(i) tetrahedral sheet rotation: ditrigonal distortion of the ideal hexagonal rings reduces the dimensions of the tetrahedral sheet

(ii) octahedral sheet flattening: relaxation due to cation-cation repulsion across shared octahedron edges leads to a thinning and lateral expansion of the octahedral sheet.

In the great majority of micas, the size of the tetrahedral sheet exceeds that of the octahedral sheet, and thus both mechanisms

TABLE 2. OBSERVED NATURAL IDEAL MICA POLYTYPES

Trioctahedral				Diocahedral			
Species [*]	Abundance			Species [*]	Abundance		
	High	Medium	Low		High	Medium	Low
PHLOGOPITE	1M, 1Md	3T	2M ₁	MUSCOVITE	2M ₁	1M, 1Md	3T
BIOTITE	1M, 1Md	3T	2M ₁	paragonite	2M ₁	1M	
annite	1M			LITHIAN MUSCOVITE	2M ₁		
SIDEROPHYLLITE	1M			Ba MUSCOVITE	1M		
LEPIDOMELANE	1M			MANGANOAN MUSCOVITE	1M		
LEPIDOLITE	1M, 2M ₂	3T	2M ₁	roscoelite	1M		
ZINNWALDITE	1M, 1Md	2M ₁ , 3T		chernykhite	2M ₁		
taeniolite	1M, 3T	2M ₁		glauconite	1M, 1Md		
ephesite	2M ₁			celadonite	1M, 1Md		
hendricksite	1M	2M ₁	3T				
manganoan phlogopite	1M	2M ₁					

* species in upper case are characteristic of pegmatites

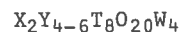
promote inter-sheet linkage. The relative importance of each mechanism is obviously compositionally dependent. Substitution of larger cations into the tetrahedral sheet enlarges its dimensions and requires further tetrahedral rotation. Substitution of larger cations into the octahedral sheet enlarges its dimensions and requires less octahedral sheet flattening. Substitution of higher charge (usually smaller) cations into the octahedral sheet reduces its dimensions but promotes octahedral flattening through increased cation-repulsion forces. Such effects can be described in detail for specific compositional types, and more complex models with additional compensation mechanisms are developed by Franzini (1969), McCauley and Newnham (1971) and Appelo (1978).

Cation Ordering

The ideal polytypic structures of Table 1 can show cation ordering when permitted by the space group symmetry. When cation ordering is energetically favourable but not allowed by the ideal space group, the structure may assume a subgroup symmetry in order to adopt an ordered cation arrangement. Bailey (1975) has suggested that this is more common than was realized, as the derivative structure may show the same diffraction aspect as the ideal structure. This is supported by the work of Guggenheim and Bailey (1975, 1977, 1978), Swanson and Bailey (1981), Bish et al. (1979) and Velde (1978, 1980).

CRYSTAL CHEMISTRY

The general formula of the micas can be written as



..where X = Na, K, Ca, Ba

Y = Mg, Fe²⁺, Mn, Li, Al, Fe³⁺, Ti, Cr

T = Si, Al, Fe³⁺

W = OH, F

They are subdivided into dioctahedral (Y₄) and trioctahedral (Y₆) types, with immiscibility between the two groups. A further subdivision is made on the basis of the interlayer (X) cation; the interlayer cation is univalent (X=Na, K) for the common micas and divalent (X=Ca, Ba) for the brittle micas. Names and chemical formulae are listed in Table 3.

Phlogopite-Biotite Series

Members of this series ideally fall within the compositional limits defined by the end-members phlogopite-eastonite-annite-siderophyllite. However, the synthesis results of Hewitt and Wones (1975) suggest that the more aluminous compositions are not realized (Fig. 3a), except in the (X=)Na analogues. Natural micas in this series frequently contain vacancies at the octahedrally coordinated positions (i.e. show some solid solution towards dioctahedral compositions); this is the result of the substitution mechanisms operative within the composite layer. There are two principal mechanisms whereby trivalent cations can substitute at the octahedral sites in the phlogopite-biotite series:

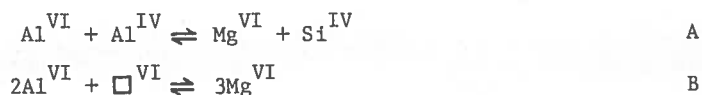


TABLE 3. IDEAL MICA COMPOSITIONS

Dioctahedral	
<u>Common Micas</u>	
MUSCOVITE	K ₂ Al ₄ Si ₆ Al ₂ O ₂₀ (OH, F) ₄
Paragonite	Na ₂ Al ₄ Si ₆ Al ₂ O ₂₀ (OH, F) ₄
Celadonite	K ₂ Mg ₂ Al ₂ Si ₈ O ₂₀ (OH, F) ₄
Glauconite	(K, Na) ₂ (Fe ³⁺ , Al, Mg) ₄ (Si, Al) ₈ O ₂₀ (OH, F) ₄
Roscoelite	K ₂ (V, Al, Mg) ₄ Si ₆ Al ₂ O ₂₀ (OH, F) ₄
<u>Brittle Micas</u>	
Margarite	Ca ₂ Al ₄ Si ₄ Al ₄ O ₂₀ (OH, F) ₄
Chernykhite	(Ba, Na) ₂ (V, Al) ₄ (Si, Al) ₈ O ₂₀ (OH, F) ₄
Trioctahedral	
<u>Common Micas</u>	
PHLOGOPITE	K ₂ Mg ₆ Si ₆ Al ₂ O ₂₀ (OH, F) ₄
Annite	K ₂ Fe ₆ ²⁺ Si ₆ Al ₂ O ₂₀ (OH, F) ₄
'Eastonite'	K ₂ Mg ₄ Al ₂ Si ₄ Al ₄ O ₂₀ (OH, F) ₄
SIDEROPHYLLITE	K ₂ Fe ₄ ²⁺ Al ₂ Si ₄ Al ₄ O ₂₀ (OH, F) ₄
'LEPIDOMELANE'	K ₂ Fe ₄ ²⁺ Fe ₂ ³⁺ Si ₄ Al ₄ O ₂₀ (OH, F) ₄
ZINNWALDITE	K ₂ Fe ₂ ²⁺ Li ₂ Al ₂ Si ₆ Al ₂ O ₂₀ (OH, F) ₄
Polyolithionite	K ₂ Li ₄ Al ₂ Si ₈ O ₂₀ (F, OH) ₄
TRILITHIONITE	K ₂ Li ₃ Al ₃ Si ₆ Al ₂ O ₂₀ (F, OH) ₄
Ephesite	Na ₂ Li ₂ Al ₄ Si ₄ Al ₄ O ₂₀ (OH, F) ₄
Henricksite	K ₂ (Zn, Mn) ₆ Si ₆ Al ₂ O ₂₀ (OH, F) ₄
Taeniolite	K ₂ Mg ₄ Li ₂ Si ₈ O ₂₀ F ₄
LEPIDOLITE	K ₂ (Li, Al) ₆ (Si, Al) ₈ O ₂₀ (F, OH) ₄

Masutomilite	$K_2(Li, Al, Mn)_6(Si, Al)_8O_{20}(F, OH)_4$
BIOTITE	$K_2(Mg, Fe^{2+}, Fe^{3+}, Al)_{6-5}(Si, Al, Fe^{3+})_8O_{20}(OH, F)_4$
Montdorite	$(K, Na)_2(Fe^{2+}, Mn, Mg)_5Si_8O_{20}(OH, F)_4$
<u>Brittle Micas</u>	
Anandite	$(Ba, K)_2(Fe^{2+}, Mg)_6(Si, Al, Fe^{3+})_8O_{20}(OH, F)_4$
Kinoshitalite	$(Ba, K)_2(Mg, Mn, Al)_6Si_4Al_4O_{20}(OH, F)_4$
Clintonite	$Ca_2(Mg, Al)_6Si_6Al_2O_{20}(OH, F)_4$

Pegmatitic micas are in upper case

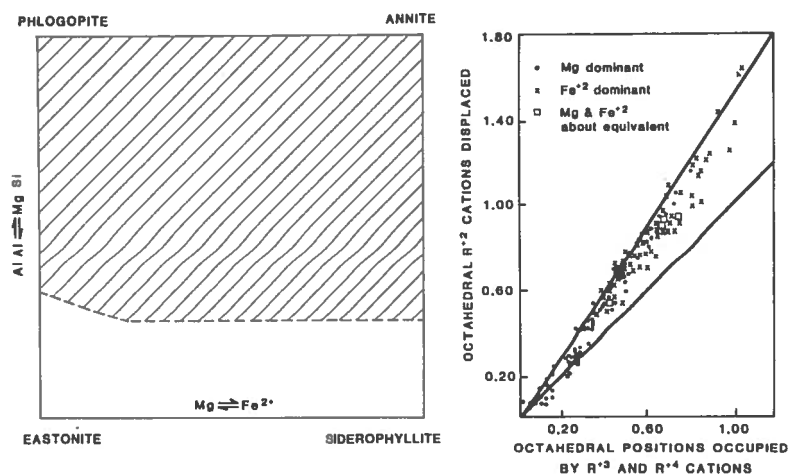
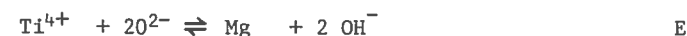
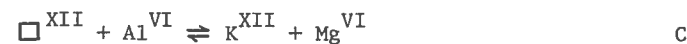


Fig. 3. (a) Compositional stability field of 'biotite' in the system phlogopite-annite-eastonite-siderophyllite at $P_{fluid} = 1\text{ kb}$, $P_{H_2} = 100\text{ bars}$, $T = 800-850^\circ\text{C}$ (Fe-bearing compositions) and $P_{H_2O} = 1-2\text{ kb}$, $T = 800-850^\circ\text{C}$ (Fe-free compositions); modified from Hewitt and Wones (1975). (b) $VI R^{2+}$ displaced in octahedral sheet as a function of $VI(R^{3+}+R^{4+})$ in trioctahedral micas; the lines represent the ideal trends from reactions A and B respectively; from Foster (1960a).

Foster (1960a) has examined the relative importance of these two mechanisms for natural compositions (Fig. 3b). Substitution B is the principal mechanism of trivalent cation accommodation, but is obviously subject to some perturbation as most of the data lie below the ideal trend. At low trivalent cation contents (and low Fe^{2+}), the data correspond to curve A, and Foster (1960a) suggested that mechanism A is operative in phlogopites. Other mechanisms involving polyvalent substitution in the octahedral sheet are



In support of C, micas of this series often have vacancies at the interlayer position (for the data of Foster (1960), $\langle \Sigma X \rangle = 1.94$). In support of D, Forbes (1972) and Levillain (1980) showed that biotites are significantly hydroxyl-deficient, and that this deficiency is related to the net charges of the tetrahedral and octahedral sheets. Bond-valence arguments suggest that E may be preferred to D when sufficient Ti is available.

Figure 3b suggests that $2.0(R^{3+}+R^{4+})^{VI}$ cations p.f.u. is the limit of substitution into the trioctahedral micas. This limit is consistent with an ordered distribution of $R^{3+}+R^{4+}$ in the structure, and stereochemical considerations suggest a lowering of the ideal polytype symmetry.

Variation in $Mg \rightleftharpoons Fe^{2+}$ substitution in this series is shown in Figure 4. In both synthetic compositions (Wones 1963) and natural compositions, there is continuous solid solution across the whole range, with the series divided up into the phlogopites, biotites and lepidomelanes. The compositional limits in Figure 6 are very striking. Guidotti et al. (1975) emphasize that $R^{3+} = Al^{VI} + Ti + Fe^{3+}$ and not just Al^{VI} , and suggest that the trend is caused partly by variations in Ti and Fe^{3+} (for constant Al^{VI} content), and partly by geochemical constraints exerted by total rock chemistry (highly magnesian environments are generally poor in Al, Ti and Fe^{3+}).

Substitution of Na for K at the interlayer position is rare in anything more than minor amounts, but $Na > K$ occurs in sodium phlogopite (Schreyer et al. 1980) and wonesite, a trioctahedral 'layer silicate' intermediate between sodium phlogopite and talc (Spear et al. 1981). Both occurrences coexist with phlogopite, suggesting a miscibility gap between phlogopite and sodium phlogopite and a trioctahedral analogy with the muscovite-paragonite immiscibility. In addition, Veblen (1981) has shown that wonesite shows micro-exsolution to a mixture of sodium phlogopite and talc.

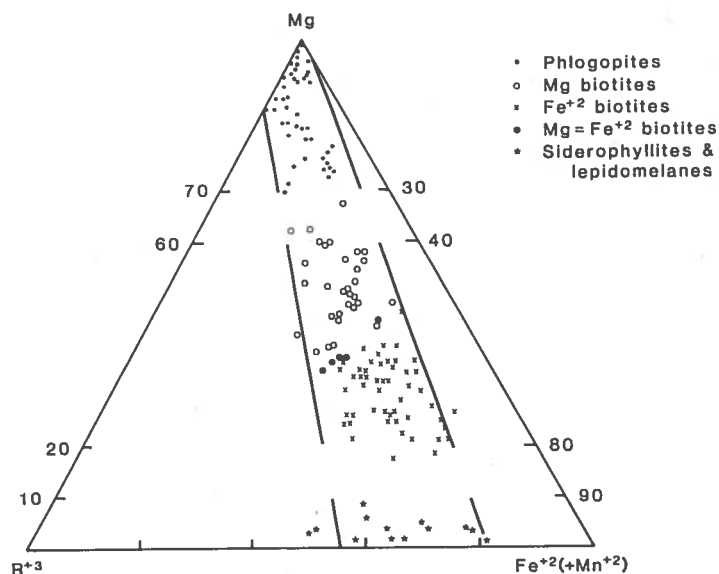
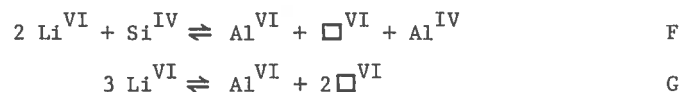


Fig. 4. Relation between Mg, $\text{Fe}^{2+}(+\text{Mn}^{2+})$ and $\text{R}^{3+}(\text{Al}, \text{Fe}^{3+}, \text{Ti})$ in trioctahedral micas; from Foster (1960a).

Muscovite-Lepidolite Series

Members of this series ideally fall within the compositional limits defined by the end members muscovite-trilithionite-polyolithionite. Both natural compositions and synthesis work indicate that micas occur over the whole compositional range, but it is not yet clear whether or not multiphase assemblages are strictly polytypic or polytypoid.

There are two principal mechanisms whereby Li can substitute at the octahedral sites in the dioctahedral muscovite structure:



Foster (1960b) has examined the relative importance of these two mechanisms for natural compositions (Fig. 5). Both are of equal importance, with natural compositions scattered between the two trends. However, the Li substitution is not a simple isomorphous replacement, as muscovite is a dioctahedral mica while the

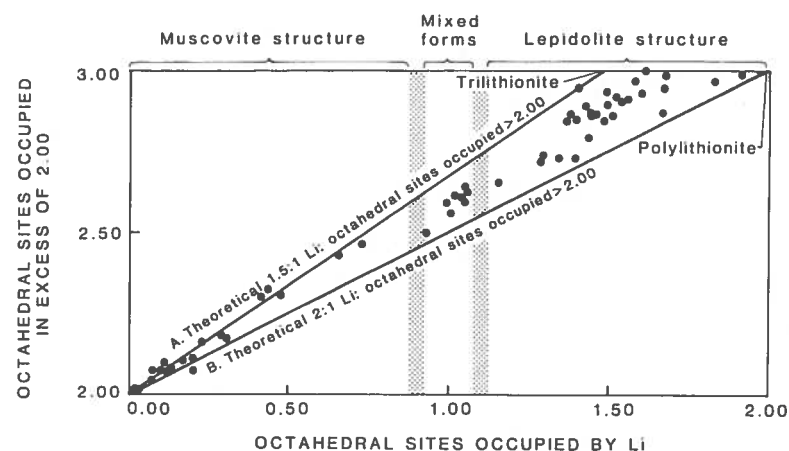


Fig. 5. Relation between Li, octahedral occupancy in excess of 4, and structure type in the muscovite-lepidolite series micas; modified from Foster (1960b).

lepidolites are trioctahedral micas, with a region of 'mixed forms' separating the two types. Series nomenclature is shown in Table 4 and Figure 6, which also shows the limited divalent cation substitution in the dioctahedral members. It is not clear whether or not there is a two-phase region along every pseudo-binary join in this series (Fig. 7). As the same ideal polytypic form may exist on both sides of the 'mixed forms' region, the incomplete miscibility may result from the occurrence of different ordering schemes (in derivative space groups) on either side of the region.

Micas of this series contain stoichiometric $(\text{OH}+\text{F})$ contents, with variation attributable to analytical error (Levillain 1980). Li and F are very strongly correlated in natural compositions (Fig. 8a), and this is borne out by the synthesis work of Munoz (1968).

Zinnwaldite-Lepidomelane Series

Members of this series ideally fall within the compositional limits defined by the end members siderophyllite-polyolithionite. Natural compositions span the complete range (Foster 1960b; Rieder 1970), as shown in Figure 6, but the stability is strongly controlled by the F content. The ideal mechanism whereby Li substitutes at the octahedral sites in the trioctahedral structure is

TABLE 4. NOMENCLATURE OF THE MUSCOVITE-LEPIDOLITE SERIES

	Li p.f.u.	Total Oct. Cations *
Lithium muscovite	0.00-1.80	4.00-4.90
Mixed forms	1.80-2.20	4.90-5.10
Lepidolite	2.20-4.0	5.10-6.00

* these values are only approximate as more than one substitution mechanism is involved

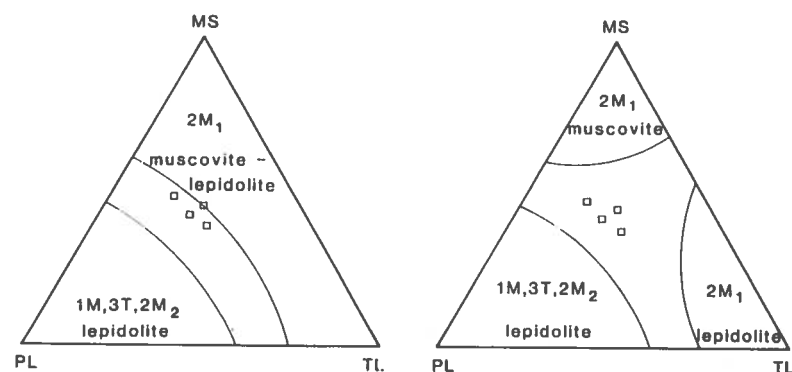
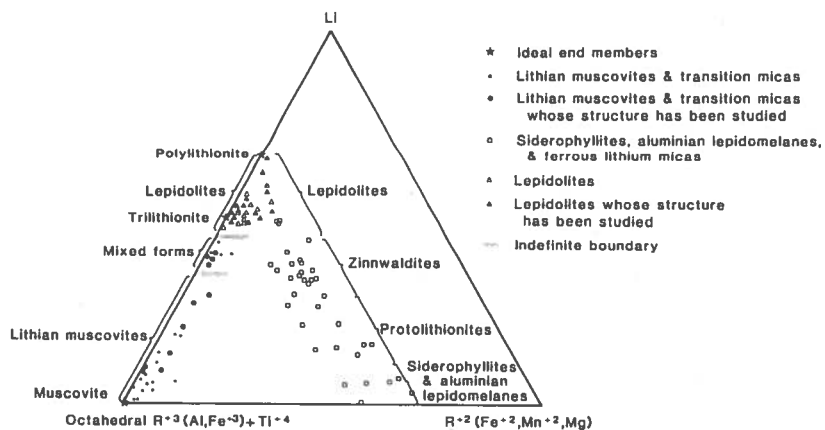


Fig. 7. Two possible interpretations of the distribution of mica polytypes in the muscovite-lepidolite series; from Munoz (1968).

Fig. 6. Relation between Li, $R^{2+}(Fe^{2+}, Mn^{2+}, Mg)$ and $VI R^{3+}(Al, Fe^{3+}, Ti)$ in lithium micas; from Foster (1960b).

H

Data for natural micas (Fig. 8b) lie below the ideal trend for trioctahedral micas with a fully occupied octahedral sheet, indicating the presence of vacancies and small variations in the ideal R^{3+} content (Rieder 1970) of two atoms p.f.u. Deviations from the ideal relationship are similar for the data of Foster (1960b) and Rieder (1970), and seem systematic but there has been no detailed examination of this.

Li and F are not as well correlated for this series as compared with the muscovite-lepidolite series (Fig. 8c), but F is always in excess of Li. Mg is generally negligible. Levillain (1980) has shown that micas of this series have (OH-F) significantly in excess of the ideal amount; moreover, this excess when recalculated as H_3O^+ , is equal to the cation deficiency at the X (interlayer) position.

TABLE 5. PARAGENETIC TYPES OF MICAS IN GRANITIC PEGMATITES

(1) Exomorphic reaction micas:

- (A) Phlogopite at contacts with serpentinite
- (B) Phlogopite at contacts with carbonate rocks
- (C) Rare-alkali biotite at contacts with amphibolites and mica schists
- (D) Other types

(2) Internal pegmatite micas:

- (A) Biotite of primitive pegmatite types
- (B) "Early" muscovite of ceramic to rare-element and miarolitic pegmatites
- (C) Lithium-aluminum micas of rare-element and miarolitic pegmatites
- (D) "Late" muscovite in albitization and greisenization units of rare-element pegmatites
- (E) Late sericitic alteration
- (F) Zinnwaldite

(3) Miscellaneous sheet silicates:

- (A) Cookeite and manandonite
- (B) Beryllian margarite, bityite, bowleyite

feldspathic composition and mechanically undisturbed. The extent of contact reaction increases with the compositional complexity of the pegmatite, progressive compositional difference between the pegmatite and its host rock, and increasing brittle deformation of the host. Thus the most conspicuous reactions are found along the borders of complex pegmatites emplaced into sheared basic and ultra-basic lithologies.

Phlogopite at contacts with serpentinite. Pegmatites intruding peridotites and serpentinites are invariably bordered by layered reaction assemblages carrying phlogopite. This mica is commonly in direct contact with the feldspars (+ quartz) of the pegmatite but separated from the wallrock by a (discontinuous) layer of monoclinic amphibole (usually tremolite-actinolite) and by anthophyllite or talc. The fresh mica is dark brown to brownish-black but the colour lightens considerably with incipient alteration.

The Mg/Fe ratio of this phlogopite is generally high, influenced mainly by the composition of the host rock which is also responsible for the characteristic Cr and Ni contents (Table 6, anal. 3 and 4). In contrast, the rare alkali contents and the OH/F ratio are functions of the pegmatite composition. No single-crystal X-ray diffraction data appear to be available; 1M or 3T polytypes are indicated by powder patterns (Table 6, anal. 3 and 4).

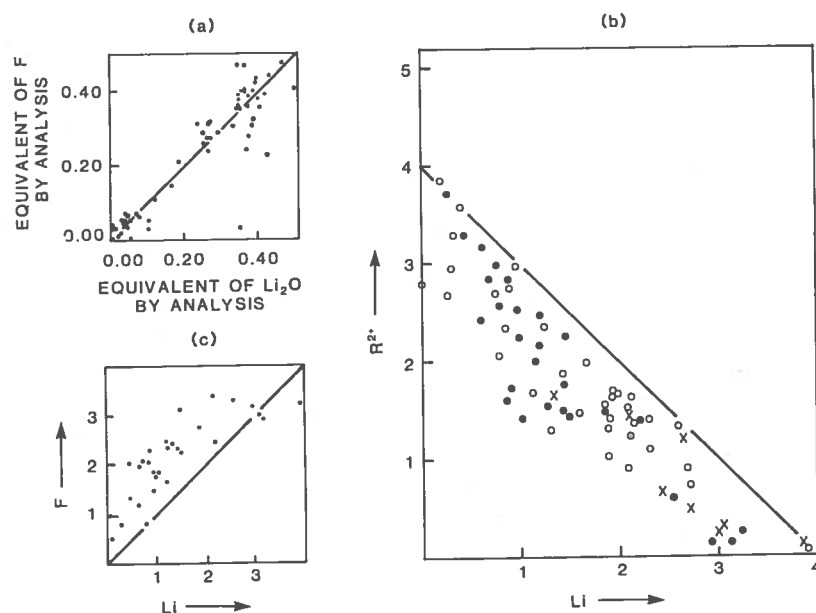


Fig. 8. (a) Li_2O as a function of F in the muscovite-lepidolite series; from Foster (1960b). (b) Li as a function of $\text{VI}_{\text{R}^{2+}}(\text{Mg}+\text{Fe}^{2+}+\text{Mn}^{2+})$ in the zinnwaldite-lepidomelane series; the line represents the ideal trend of reaction H. (c) Li as a function of F in the zinnwaldite-lepidomelane series; after Rieder (1970).

PARAGENETIC TYPES OF PEGMATITE MICAS

Table 5 presents a schematic classification of the principal categories of micas characteristic of different types and crystallization stages of granitic pegmatites. Most of the mica types are rather general, and some species and varieties (e.g. lithian muscovite and lepidolite) are listed under a single heading to simplify their discussion.

Exomorphic Micas

Granitic pegmatites of mineralogically and structurally simple types commonly show negligible reaction with the enclosing rocks, particularly if these are of granitic, gneissic or generally quartzo-

TABLE 6. BIOTITE-PHLOGOPITE MICAS

	1	2	3	4	5	6
SiO ₂	39.97	42.02	38.89	41.87	37.00	37.01
TiO ₂	2.64	1.35	1.70	1.63	1.83	0.02
Al ₂ O ₃	17.51	18.75	16.05	11.28	19.10	15.89
Fe ₂ O ₃	2.26	0.66	3.74	1.96	1.43	tr.
FeO	14.81	8.29	7.01	2.62	18.02	30.18
MnO	0.22	0.27	0.10	0.04	0.39	1.01
MgO	8.45	9.55	18.70	24.25	9.28	0.22
CaO	0.00	0.93	-	0.30	0.43	0.10
SrO	-	-	-	tr.	-	-
BaO	-	-	-	0.01	-	-
Li ₂ O	0.65	1.20	-	0.07	0.03	-
Na ₂ O	0.45	0.73	0.32	0.31	0.13	0.58
K ₂ O	8.48	8.54	8.29	9.52	9.05	9.02
Rb ₂ O	1.48	1.85	-	0.11	0.08	-
Cs ₂ O	1.12	0.47	-	0.005	-	-
H ₂ O-	0.32	0.16	-	0.76	tr.	0.00
H ₂ O+	2.48	2.44	4.68	3.55	3.46	1.92
F ₂	3.17	4.34	0.68	1.75	0.19	3.88
	101.01	101.55	100.29	100.94	100.42	101.37
-O≡F ₂	1.34	1.83	0.29	0.73	0.08	1.68
	99.67	99.72	100.00	100.21	100.34	99.69

- not analyzed; tr. traces

(1) #2 Tin Mountain, South Dakota, biotite in mica schist at pegmatite contact (Hess and Stevens 1937); (2) Kings Mountain, North Carolina, biotite in mica schist at pegmatite contact (Hess and Stevens 1937); (3) #1 Věžná, Czechoslovakia, 1M or 3T ferroan phlogopite at pegmatite contact with serpentinite (with 0.13 Cr₂O₃; corrected for admixed apatite; Černý and Miškovský 1966); (4) Heřmanov, Czechoslovakia, 1M or 3T phlogopite at pegmatite contact with serpentinite (with 0.46 Cr₂O₃, 0.14 NiO, 0.30 P₂O₅; Černý 1972); (5) #656/38 Tedino, Northern Karelia, lath-shaped biotite from a plagioclase-microcline pegmatite (Gordiyenko et al. 1976); (6) Brooks Mountain, Alaska, biotite from pegmatite in granite (Coates and Fahey 1944).

Occurrences of this phlogopite type are widespread; pegmatites of different types commonly crosscut and cement fractured and brecciated ultrabasic rocks in amphibolite grade terrains. Information on this mica, however, is rather scanty (Borodayevskaya 1951; Bogomolova 1962) mainly because it is readily altered into hydrobiotite and vermiculite, or chlorite (e.g., Hadley 1949; Kulp and Brobst 1954; Morel 1955; Omori 1958; Bassett 1959). The cation leaching, exchange and hydration/oxidation of Fe²⁺ could be (locally) hydrothermal but in the majority of cases it is considered to be a hypergene weathering effect (Kulp and Brobst 1954; Bassett 1963; Boettcher 1966).

The contact reaction generating the phlogopite and associated amphibole + talc layers is evidently due to a metasomatic exchange between the silicic pegmatite and the ultrabasic wallrock, facilitated by volatile agents such as water and fluorine (Korzhinskii 1953; Brownlow 1961; cf. also Brady 1977). Development of the reaction rims along pegmatite contacts with anhydrous, non-serpentinized peridotite (P.M. Larrabee, priv. comm. 1970) indicates that the activity of the volatile components in the pegmatite melt alone is sufficient to promote the metasomatism. The above case also indicates that the reaction is a high-temperature process which does not initiate serpentinization. Thus the metasomatic exchange could possibly accompany the initial pegmatite crystallization, and does not need to be a postmagmatic event driven by genetically unrelated fluids (Phillips and Hess 1936; Korzhinskii 1953; Sherstyuk 1965). Serpentinization postdating pegmatite intrusion and reaction results in retrogression of the contact assemblage and even in chloritization of the pegmatite margins (Černý and Povondra 1965; Černý 1968).

Phlogopite at contacts with carbonate rocks. The majority of pegmatites intrusive into marbles belong to mineralogically simple and geochemically primitive types. They seem to be related to local mobilization of granitoid melts in neighbouring metamorphic suites or to igneous differentiation of granitic intrusions. Highly fractionated and mineralized pegmatites crosscutting metamorphosed carbonate sediments are relatively scarce (e.g., Luna 1962; Rossovskii 1963).

Whatever the nature of the pegmatites, they invariably develop reaction envelopes against their carbonate host. The mineral composition and sequence of metasomatic zones are a function of the pegmatite and wallrock composition, the depth of emplacement, and the nature of extraneous fluids which could penetrate along the contacts after the consolidation of the pegmatite. Diopside, vesuvianite, epidote, phlogopite, grossular and wollastonite are the most common constituents of the reaction rims at the amphibolite grade of regional metamorphism, with phlogopite and other ferromagnesian minerals along the pegmatite margins and wollastonite in contact with the host rock. Pegmatites in the Ariege area of France (Lacroix 1898), Žulová in Czechoslovakia, and at Tanneberg and Mansjö Mt. in Sweden (v. Eckermann 1922, 1923) are classic localities of this type. In the upper amphibolite to granulite grade, diopside

and phlogopite are the main components of the reaction envelopes, accompanied by apatite, hornblende, titanite, REE-bearing minerals and other accessory phases, including sulphides and graphite. The Slyudyanka district in the Baikal region, U.S.S.R., the Loolekop phlogopite deposits in Transvaal (Gevers 1948), and a belt of localities in the Grenville Province of Ontario and Quebec (Currie 1951, Hoadley 1960) represent this type.

Phlogopite is usually very close to the ideal end-member composition of $K_2Mg_6Si_6Al_2O_{20}(F,OH)_4$, very poor in Fe and Al^{VI} and with variable F content. No modern chemical analyses including trace elements appear to be available. Older data on phlogopites from Mansjö and Slyudyanka are quoted by Foster (1960a).

The origin of phlogopite is again somewhat controversial. Besides an early reaction of pegmatite melt with carbonate rocks (refuted by Korzhinskii 1953), late solutions penetrating along pegmatite/marble contacts seem to be responsible in some localities (e.g. Kugi-lyal in southwestern Pamir where phlogopite is associated with spinel, forsterite, enstatite, tremolite, anthophyllite and talc; Rossovskii 1963). At least two stages of mineralization, and perhaps two generations of phlogopite, are probable in the Loolekop and Grenvillian localities (Gevers 1948; Currie 1951; Hoadley 1960). The diversified origin of phlogopite in the Slyudyanka district due to pegmatite intrusion, scapolite-bearing silicic veining and metasomatic reaction along marble contacts with gneissic lithologies was recognized by Fersman (1940), and interpreted by Korzhinskii (1947).

Rare-alkali biotite. This exomorphic mica is restricted to amphibolite (gabbro, metabasalt) wallrocks adjacent to highly fractionated lithium-rich pegmatites. Its occurrences in mica schists are very scarce. This mica occurs as seams in immediate contact with the pegmatite, replacing predominantly the mafic minerals of the host rock. It is particularly well developed in tectonically disturbed wallrocks. Fragments of brecciated pegmatite-cemented amphibolite may be completely replaced, and mica schist sequences, warped and "exfoliated" by the pegmatite emplacement, may be pervaded by dispersed secondary micas through sizable volumes. Exomorphic minerals more or less closely associated with this mica include tourmaline, holmquistite, apatite, triphylite, arsenopyrite and pyrrhotite.

The Mg/Fe ratio and aluminum content of this biotite type are highly variable (from about 0.5 to about 2.5 for Mg/Fe). Mg/Fe is evidently controlled by the composition of the wallrock and by the coexisting mafic phases. Complete chemical analyses are available at present for only a small number of samples (Hess and Stevens 1937). On the basis of partial analyses, however, it seems likely that this mica varies from phlogopite to siderophyllite to zinnwaldite.

The feature common to all these micas is the high content of Li, Rb, and Cs (Table 6, anal. 1 and 2). The concentrations of the rare alkalis reach up to 2.33 Li_2O , 1.87 Rb_2O and 1.12 Cs_2O (in wt.%;

Hess and Stevens 1937; Černý et al. 1981).

The rare-alkali biotite originates by the action of Li, Rb, Cs, K- and F-enriched fluids expelled from the pegmatite in the late metasomatic stages of its internal evolution, during which the primary Li, Rb, Cs-bearing minerals (such as spodumene, K-feldspar, polylucite) undergo albitization, greisenization and similar destructive processes mobilizing their rare-alkali contents.

A special type of mica should be mentioned which has no spatial relationship but a definite genetic link with granitic pegmatites. This is the rare-alkali biotite which constitutes (together with chlorite, holmquistite, plagioclase, hornblende and epidote) zoned metasomatic belts in hornblende gabbro-anorthosites of one of the extensive pegmatite provinces of the U.S.S.R. (Gordiyenko et al. 1975). In contrast to the exomorphic biotite discussed above, these metasomatic belts follow tectonic breaks separated from spodumene pegmatites of the region by as much as 300 m of unaltered host rocks. In different zones of the metasomatic belts, the rare alkali contents reach the averages of 0.044 Cs_2O , 1.2 Li_2O , and 0.1 wt.% Rb_2O in whole-rock samples.

Other exomorphic micas. Micas and other layer silicates commonly occur internally and externally along pegmatite contacts with a wide variety of country rocks, even in the absence of other reaction products. Muscovite is, for example, abundant along contacts of pegmatites with low-grade metaturbidites in the Yellowknife and Wekusko pegmatite fields (Kretz 1968; Černý et al. 1981) and in high-grade peraluminous gneisses hosting mica-bearing pegmatites (Gordiyenko and Leonova 1976). Element migration in a predominantly diffusional metasomatism, generating chlorite and biotite along pegmatite contacts with metasediments, was studied by Neiva (1980). All these and analogous micas do not represent any particular crystallochemical or economic interest per se, but they provide clues for understanding the metasomatic exchanges between pegmatites and diverse wallrock compositions, and may serve as exploration guides.

Internal Mica Types

The diversity of micas occurring within granitic pegmatites exceeds that of the exomorphic varieties. The micas are characteristic of different pegmatite types, and of different primary zones and metasomatic units of a single pegmatite type. Some mica species tend to occur in several generations not only in complex rare-element pegmatites but also in simple types.

Biotite. Subordinate to accessory biotite is typical of geochemically primitive pegmatites. It is found mainly in the deep-seated pegmatites of the ceramic and mica-bearing types, and in simple barren to Be, Ti, Nb-Ta, REE(U, Th)-bearing pegmatites. In complex rare-alkali-enriched dikes, biotite is restricted to the outermost units and generally represents a contamination effect rather

than a product of the pristine pegmatite melt.

Biotite of the mica-bearing pegmatites in high-grade metamorphic terrains occurs in three generations (Gordiyenko and Leonova 1976):

- (i) as thin elongate crystals in outer zones of granitic texture, grading into the
- (ii) block biotite of blocky zones, and
- (iii) giant but thin platy biotite in quartz cores.

All generations of this biotite display considerable TiO_2 (1.5-2.75 wt.%), MgO (7-13.5 wt.%) and Al^{VI} contents, high Ba and low Li, Rb and Cs (Table 6, anal. 5). Mg and Ba enrichment is particularly conspicuous when compared to the compositions of biotite of the second type described above. This biotite occurs in coarse flakes and dendritic aggregates of lath-shaped crystals in the granitic and graphic outer zones, and as giant plates and books (some of which may be metasomatic) in blocky zones adjacent to the quartz cores. If not affected by contamination from the wallrock, this mica represents the ultimate product of igneous biotite fractionation, being extremely Fe-rich. The Mg/Fe ratio of the example shown in Table 6, anal. 6 is admittedly unique but many other biotites of this paragenetic type approach this composition. Fe^{3+} is occasionally also high, as shown by Shibata (1952).

Replacement of pegmatitic biotite by muscovite is common. Tourmalinization of biotite is also widespread in B-enriched pegmatites. Biotite is sensitive to surface weathering, with loss of alkalis and oxidation of Fe^{2+} .

"Early" muscovite. This category encompasses primary and secondary muscovites of the main stages of pegmatite consolidation, before the onset of late metasomatic effects. The distinction of primary and metasomatic mica generations in this group may occasionally be difficult.

In mica-bearing pegmatites of high-grade terrains, Gordiyenko and Leonova (1976) distinguish three "early" muscovite types:

- (i) in quartz-muscovite aggregates replacing plagioclase in quasigraphic and blocky zones,
- (ii) euhedral brownish ("ruby") muscovite along the periphery of quartz cores, and
- (iii) muscovite-plagioclase aggregates crosscutting primary zones including the earlier muscovite types.

All generations have relatively high Fe (1.1-3.2 wt.% total Fe as FeO), Fe_2O_3 slightly predominant over FeO, relatively high MgO (0.5-1.45 wt.%), BaO (to 0.61 wt.%), and very low Be and rare-alkali contents (Table 7, anal. 1). The R^{VI} total is commonly somewhat higher than dioctahedral; phengite-type substitution $\text{Al}^{\text{VI}} + \text{Al}^{\text{IV}} \rightleftharpoons \text{R}^{\text{VI}} + \text{Si}^{\text{IV}}$ is negligible. Differences among the three muscovite

TABLE 7. MUSCOVITE AND LITHIAN MUSCOVITE

	1	2	3	4	5	6	7
SiO_2	45.50	44.02	45.02	45.15	46.34	47.80	45.24
TiO_2	0.54	0.06	0.10	0.13	0.06	-	0.01
Al_2O_3	33.90	33.95	35.85	32.91	32.47	28.64	36.85
Fe_2O_3	1.06	3.11	2.44	1.87	0.00	} 0.06	0.09
FeO	0.87	0.79	0.10	3.34	1.06		0.02
MnO	tr.	0.06	0.02	0.35	0.35	0.20	0.12
MgO	0.93	0.37	0.20	0.01	0.00	<0.01	0.08
CaO	-	0.12	0.09	0.005	0.36	-	0.00
SrO	-	-	-	-	-	-	-
BaO	0.61	-	-	-	-	-	-
Li_2O	0.01	0.19	tr.	0.55	2.45	3.12	0.49
Na_2O	0.78	0.84	0.32	0.60	0.5	0.19	0.64
K_2O	10.11	9.44	9.46	9.93	9.46	9.98	10.08
Rb_2O	0.03	0.42	0.84	0.62	1.5	2.91	0.93
Cs_2O	0.0002	0.01	-	-	0.2	0.50	0.20
$\text{H}_2\text{O}-$	0.32	-	-	0.42	0.32	0.48	0.46
$\text{H}_2\text{O}+$	5.32	6.39	5.69	3.17	3.32	5.60	4.12
F_2	-	-	-	2.26	2.82	2.73	0.91
	99.98	99.77	100.13	101.32	101.21	102.22	100.24
$-\text{O} \equiv \text{F}_2$	-	-	-	-0.95	1.19	1.15	0.38
	99.98	99.77	100.13	100.37	100.02	101.07	99.86

- not analyzed; tr. traces

(1) #16/11-II Malinovaya Varaka, Northern Karelia, tabular muscovite from mica pegmatites (Gordiyenko et al. 1976); (2) #16, one of the pegmatite fields in the European part of the U.S.S.R., platy muscovite from a muscovite-feldspar pegmatite of a spodumene-bearing field (Gordiyenko 1970); (3) #12, as in (2), early green flaky muscovite from a spodumene pegmatite (Gordiyenko 1970); (4) #224 Brown Derby No. 1, Colorado, 2M_1 muscovite from wall zone of a lepidolite + topaz + (pollucite) bearing pegmatite; K_2O includes Cs_2O (Heinrich 1967); (5) #6 Varuträsk, Sweden, 2M_1 lithian muscovite from a petalite + pollucite bearing pegmatite (Quensel 1956); (6) #R7 Tanco, Manitoba, 2M_1 lithian muscovite from a petalite + pollucite-rich pegmatite (Rinaldi et al. 1972); (7) Pidlite pegmatite, New Mexico, 2M_1 rose muscovite (Heinrich and Levinson 1953).

generations are small, with a general decrease in R^{2+} , and a closer approach to the ideal muscovite composition from the first to the third type. Gordiyenko and Leonova (1976) also list and discuss variations in 12 trace elements.

"Early" muscovite of intermediate-level rare-element pegmatites occurs in several generations, and is usually confined to outer pegmatite zones. Book muscovite lining the margins of quartz cores occurs only in barren or relatively simple pegmatites with Be, Nb > Ta mineralization, and even in these cases it may be metasomatic, associated with cleavelandite. A good account of muscovite types (and their compositional characteristics) in various pegmatite categories of spodumene-bearing pegmatite fields is given by Gordiyenko (1970). Compared to the muscovites of mica-bearing pegmatites, they have lower ΣFe (1-2 wt.% total Fe as FeO), MgO (0.1-0.4 wt.%), BaO and increased Li, Rb, Cs contents (Table 7, anal. 2, 3, 4). In the low-pressure petalite type of highly fractionated, pollucite bearing lithium pegmatites, Fe drops to 0.2 wt.% (total Fe as FeO) and MgO becomes negligible (0.0 wt.%; Rinaldi et al. 1972; Černý et al. 1981). Except for the substitution of (Rb, Cs) for K and minor Li entering the octahedral sites, the discussed muscovites closely approach the ideal formula (Table 3) as shown in Figure 9. In terms of polytypism, all paragenetic types of pegmatitic muscovite have the $2M_1$ structure.

Lithian muscovite, mixed types, and lepidolite. Lithium-aluminum micas occur with a wide variety of morphologies, generations, polytypes and compositions. In terms of morphology, their aggregates range from dense masses of microscopic flakes to giant flat or curved tabular crystals, and conical columns elongated normal to the cleavage. Zonal overgrowths of Li-Al micas on early muscovite are common, particularly in open vugs (Brock 1974; Foord 1976) and replacement of muscovite and biotite by lepidolite is also widespread (Ginsburg and Berchin 1953; Ginsburg 1957a). In complex pegmatites, several generations are commonly distinguished as typical of specific zones or randomly distributed as a component of pervasive metasomatic units (Thoreau et al. 1956). No general rules apply to the numbers and affiliations of the Li-Al mica generation except in groups of cogenetic pegmatites, in which they may be recognized by persistent colour and/or morphology, aggregation or mineral association. Černý et al. (1981) found a regular succession of lithian muscovite-mixed type-lepidolite in complex petalite-bearing pegmatites of southeastern Manitoba; as an extreme contrast, the Brown Derby, Colorado pegmatite (Heinrich 1967) has no compositional or structural regularity of subsequent mica types.

This variability is understandable in the light of experimental work by Munoz (1971) who demonstrated complex relationships among lepidolite compositions and liquidus to subsolidus stabilities in dependence on silica saturation, $f_{\text{H}_2\text{O}}$ and f_{HF} . His results confirm the observation that natural Li-Al micas can be either secondary metasomatic products generated by the action of late F-bearing aqueous fluids on early lithium aluminosilicates and K-feldspars, or "primary" phases which do not replace any pre-existing minerals (Stewart 1963, 1978).

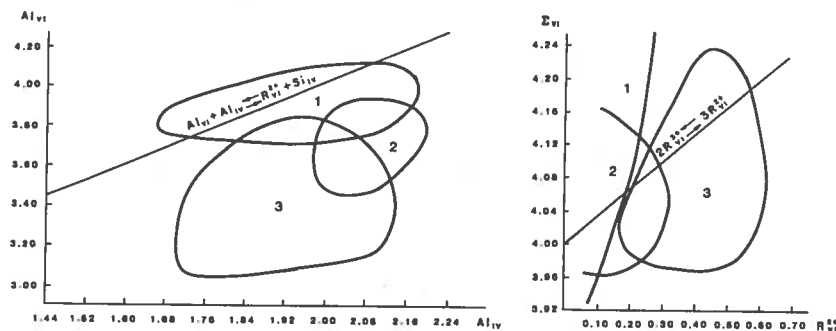


Fig. 9. Octahedral-tetrahedral substitutions in muscovite from rare-element (1), rare-element-muscovite (2), and muscovite-bearing (3) pegmatite types. Phengite substitution $\text{AlVI} + \text{AlIV} = \text{R}_{\text{II}}^{2+} + \text{Si}_{\text{IV}}$ (a), and di-tri-octahedral trend $2\text{R}^{3+} = 3\text{R}^{2+}$ (b); after Salje (1975).

Structural polytype and Li_2O content are adequate for distinguishing the three groups in the Li-Al mica family. Lithian muscovite proper is characterized by the $2M_1$ polytype and Li_2O approximately less than 3.5 wt.% (Table 7, anal. 5 and 6). Heterogeneous muscovite-lepidolite micas (transitional structures of Levinson 1953, mixed forms of Foster 1960, mixed types of Rinaldi et al. 1972) consist of the $2M_1$ muscovite component and one or another of the polytypes characteristic of lepidolite, commonly $2M_2$ or $1M$; their Li_2O contents vary between about 3.5 to approximately 4.5 wt.%. True lepidolites have the $1M$, $2M_2$, $2M_1$, $3T$ or $3M_2$ structure, and Li_2O contents higher than about 4.0 wt.% (Table 8, anal. 1 to 5). The $1M$ and $2M_2$ polytypes are the most abundant and commonly are intergrown on very fine scale (Heinrich et al. 1953; Franzini and Sartori 1969; Černý et al. 1970; Rieder 1970; Taluda et al. 1971). The $3T$ (Yakovleva et al. 1965; Brown 1978) and $3M_2$ (Heinrich et al. 1953; Bailey and Christie 1978) structures are scarce, and only two localities of the Li_2O -rich $2M_1$ lepidolite are known to date (Černý et al. 1970; Swanson and Bailey 1981).

An early attempt to correlate Li_2O percentages with polytypism among the lepidolites proper by Levinson (1953) has not been successful, as pointed out by Heinrich (1967), Rinaldi et al. (1972) and Chaudhry and Howie (1973a). This is not surprising in view of the multiplicity of substitution mechanisms operating in natural Li-Al

TABLE 8. LEPIDOLITE AND ZINNWALDITE

	1	2	3	4	5	6	7
SiO ₂	49.76	49.80	49.78	50.51	52.80	45.6	48.56
TiO ₂	0.22	0.00	0.15	0.14	-	0.23	0.16
Al ₂ O ₃	25.31	25.56	25.13	24.90	19.94	20.0	21.21
Fe ₂ O ₃	0.80	0.08	0.42	0.26	0.38	1.00	0.58
FeO	3.20	0.00	0.44	1.27	0.06	9.61	10.36
MnO	0.42	0.38	0.60	1.45	0.88	0.79	1.73
MgO	0.09	0.22	0.08	0.18	0.63	0.2	0.03
CaO	0.05	0.00	0.06	0.14	0.11	0.02	0.06
SrO	-	-	-	-	-	-	-
BaO	-	-	-	-	-	-	-
Li ₂ O	4.35	5.95	6.00	5.95	5.91	3.96	1.98
Na ₂ O	0.61	0.4	0.40	0.33	0.26	0.4	1.30
K ₂ O	9.20	9.67	9.70	10.09	9.00	10.1	9.60
Rb ₂ O	0.67	1.97	1.27	1.29	1.36	0.01	0.76
Cs ₂ O	0.17	1.2	0.25	0.12	0.75	0.06	0.46
H ₂ O-	0.33	0.38	0.47	0.00	} 2.85	} 2.61	0.10
H ₂ O+	2.81	0.50	1.39	1.24			1.48
F ₂	3.96	6.85	6.30	3.50	7.07	7.7	3.76
	101.95	102.96	102.44	101.37	101.99	102.29	102.13
-O=F ₂	1.67	2.89	2.65	1.47	2.98	3.24	1.88
	100.28	100.07	99.79	99.90	99.01	99.05	100.25

- not analyzed

(1) #MBZ Meldon, Devonshire, 1M lepidolite in sodic aplite (Chaudhry and Howie 1973b); (2) #2 Varuträsk, Sweden, coarse 1M lepidolite (Quensel 1956); (3) #MKC Meldon, Devonshire, 2M₂ lepidolite in pegmatite (Chaudhry and Howie 1973b); (4) Siberia, U.S.S.R., coarse 3T lepidolite (Yakovleva et al. 1965); (5) #B-41 Biskupice, Czechoslovakia, 2M₁ lepidolite (Černý et al. 1970); (6) Wigwam Creek No. 1 Claim, Colorado, zinnwaldite, outer zone (with 0.2 ZnO; E.E. Foord, pers. comm. 1981); (7) Rajagarh, India, zinnwaldite (Babu 1969).

micas. Munoz (1968) attempted to correlate polytypism with composition in a ternary muscovite-polyolithionite-trilithionite system, using data on natural and synthetic micas. No clearcut dependence emerged from his study. Additional data collected since the publication of his paper compounded the uncertainties (Fig. 7), although Rieder et al. (1970) suggested that 2M₂ may be typical of trilithionites. Extreme similarities among the compositions of different lepidolite polytypes are demonstrated in Table 8.

Chaudhry and Howie (1973a) and Swanson and Bailey (1981) noticed striking differences between the lepidolite habits, associated minerals and the general nature of enclosing assemblages for different polytypes within single pegmatite bodies. Swanson and Bailey also quoted experimental evidence for changes in crystal growth mechanism of micas synthesized under different conditions. Both these authors and Sartori (1976) concluded that temperature, pressure, volatiles, degree of saturation and rate of cooling are more important than the composition in determining the stability and the occurrence of lepidolite polytypes.

The Li-Al micas cover most of the muscovite-trilithionite-polyolithionite field (Figs. 5 and 7). It should be noted, however, that compositions close to the polyolithionite end member are typical of silica-undersaturated alkali syenite pegmatites rather than truly granitic types. Other noteworthy compositional features of the Li-Al micas are the positive correlation between Li₂O and F₂ (Fig. 8a) and a reciprocal correlation between F₂ and H₂O. Rb₂O and Cs₂O also tend to increase with Li₂O, but their abundances are affected by geochemical rather than crystallochemical factors. This is demonstrated by the lithian muscovite with the highest Rb₂O content recorded for Li-Al micas, 6.98 wt.% (Khanna 1977) which contains only 1.06 wt.% Li₂O. In contrast, a Rb-rich lepidolite + lithian muscovite intergrowth from northeastern Manitoba (Jambor and Potter 1967) has 4.53 wt.% Rb₂O vs. 3.08 wt.% Li₂O. Compared to biotite and muscovite, the Li-Al micas may also be enriched in Tl and Ga (up to 300 ppm and 900 ppm, respectively) but the increased contents of these elements are again a regional feature rather than a function of normal fractionation.

"Late" muscovite. In the late stages of evolution of complex pegmatites, muscovite may be produced by hydrolysis of potassium feldspars. This hydrolytic breakdown may result in the formation of greisen-like muscovite + quartz assemblages which occasionally carry cassiterite and Nb-Ta-bearing minerals. Also, such a decomposition of K-feldspars may form a muscovite halo advancing ahead of albitization fronts which may be themselves enriched in minerals of Be, Sn, Nb-Ta and other elements. In both cases, muscovite is usually easily distinguished from other mica types of the parent pegmatite, and also spatially separated from Li-rich micas, if present.

The composition of this late muscovite (Table 9, anal. 1 and 2) is typically low in rare alkalis and F₂; the Rb and Cs contents may closely reflect those of the replaced feldspar. The 2M₁ polytype is

TABLE 9. LATE MUSCOVITE AND MISCELLANEOUS LAYER SILICATES

	1	2	3	4	5	6	7
SiO ₂	45.71	44.30	44.67	34.92	25.20	32.22	29.84
TiO ₂	0.19	-	0.12	-	-	-	-
Al ₂ O ₃	35.85	34.36	38.64	46.29	47.80	35.58	47.35
B ₂ O ₃	-	-	-	0.07	9.25	-	-
Fe ₂ O ₃	1.29	-	-	-	-	-	0.81
FeO	0.72	0.32	0.28	-	-	-	-
MnO	0.09	0.07	0.35	0.13	-	-	-
MgO	0.37	<0.01	0.12	0.24	-	-	1.22
CaO	0.00	-	0.45	0.30	-	15.35	11.70
BeO	-	-	-	-	-	8.05	1.18
Li ₂ O	0.008	0.37	0.00	2.67	3.97	2.61	0.47
Na ₂ O	0.59	0.22	0.52	0.14	0.48	0.55	1.93
K ₂ O	10.47	10.12	9.80	-	0.20	0.09	0.55
Rb ₂ O	0.09	1.69	0.58	-	-	-	-
Cs ₂ O	0.055	0.21	0.00	-	-	-	-
H ₂ O-	0.16	0.80	0.34	0.28	}14.10	-	0.12
H ₂ O+	4.50	6.00	3.83	14.12		5.80	4.47
F ₂	-	0.64	0.85	-	-	-	0.70
	100.39	99.11	100.55	99.05	100.22	100.25	100.35
-O≡F ₂	-	0.27	0.37	-	-	-	0.28
	100.39	98.84	100.18	99.05	100.22	100.25	100.07

- not analyzed

(1) #3-3/14,48 Lopatova Guba, Northern Karelia, U.S.S.R., "gilbertite" replacing microcline (Gordiyenko et al. 1976); (2) #R8, Tanco pegmatite, Manitoba, late muscovite associated with Sn,Ta,Be,Zr minerals, replacing microcline (Rinaldi et al. 1972; improved SiO₂ value); (3) #B, Meldon, Devonshire, "gilbertite" from joint faces in sodic aplite (Chaudhry and Howie 1973a); (4) Manono, Katanga, cookeite (with 0.005 P₂O₅; Herman et al. 1961); (5) Sahatany Valley, Madagascar, manandonite replacing rubellite (Lacroix 1922); (6) Londonderry, Western Australia, bowleyite (Rowledge and Hayton 1948); (7) #7 Urals, U.S.S.R., beryllian margarite (Beus 1960).

typical.

A special variety of muscovite is the rose muscovite (Heinrich and Levinson 1953). It is a late muscovite generation resembling lepidolite in pink colour, but corresponding to normal muscovite in composition. The colour results from a combination of low Mn/negligible Fe contents. This mica type emphasizes the fact that even in the Li-Al micas, the colour is independent (contrary to popular belief) of the Li content and varies mainly with the ratios and oxidation states of Mn and Fe. Pale apple-green lepidolites exist as well as rose muscovites, although they are rather exceptional.

Sericitic alteration. The above muscovite type, generated by late rare-element-bearing fluids, should not be confused with the latest mica variety produced by low-temperature alteration, which may be caused even by external factors not connected with the pegmatite evolution. This is a fine-grained yellowish to silvery muscovite penetrating fractures and cleavage planes of K-feldspars, beryl, lithium aluminosilicates and other minerals; it is commonly isolated or accompanied only by quartz or calcite. In older literature the term "gilbertite" was frequently used for this mica. As pointed out by Chaudhry and Howie (1973b), this 2M₁ mica corresponds to the composition of normal muscovite and a special name is not warranted (Table 9, anal. 3).

Zinnwaldite. Several pegmatite micas resemble zinnwaldite in habit, aggregation and colour, and they are frequently designated as such in the field and in hand specimens to distinguish them from other mica varieties. The term zinnwaldite was also applied to several analyzed micas with only minor Li and Fe contents (Heinrich 1967) which plot with lithian muscovites. Actually, ubiquitous as zinnwaldite is in greisen deposits, it is rare in granitic pegmatites. It seems to be typical of Li-poor pegmatites, being the only Li-bearing species in them (Ginsburg and Berchin 1953). It occurs almost exclusively in two pegmatite types:

- (i) those derived from greisen-bearing granites, and
- (ii) the Y, Nb-enriched pegmatites related to anorogenic granitoids.

In the first paragenesis, it commonly constitutes the only mica represented in the pegmatites which carry somewhat unusual, greisen-like associations of W,Sn,Mo,Nb-Ta,P,F-bearing minerals. These pegmatites are usually closely associated with the greisenized granitic cupolas, and they are never abundant. In the second case, zinnwaldite is the typical mica of amazonite-bearing pegmatites (Foord and Martin 1979). Biotite is a minor component, and muscovite is scarce in them. Shibata (1952) analyzed zinnwaldite from pegmatites with a peculiar association of fayalite and scheelite, which could possibly be related to this second type.

Chemical analysis of pegmatitic zinnwaldites are few, particularly as in some crystallochemical studies the provenance of the

analyzed micas is not clearly stated. However, it appears that pegmatitic zinnwaldite does not substantially differ from the zinnwaldites of greisen assemblages. The two analyses shown in Table 8 (anal. 6 and 7) plot well within the crystallochemical trends established for zinnwaldite (Fig. 6).

MISCELLANEOUS SHEET SILICATES

A brief note is desirable on sheet silicates other than the common micas which occur in granitic pegmatites. These are the members of chlorite and brittle mica groups, as described below.

Cookeite and manandonite. Cookeite, ideally a di-, tri-octahedral $\text{Li}_2\text{Al}_8[\text{Al}_2\text{Si}_6\text{O}_{20}(\text{OH})_{16}]$, belongs to the Li, Al-chlorites extending from the non pegmatitic dioctahedral Al-chlorite donbassite $\text{Al}_{8.67}[\text{Al}_2\text{Si}_6\text{O}_{20}(\text{OH})_{16}]$ to the intermediate "2.75"-octahedral occupancy in the Li-richest cookeite from Australia (Brown and Henley 1972). The principal substitution $3\text{Li}^{1+} \rightleftharpoons \text{Al}^{3+}$ is only slightly modified by $3\text{R}^{2+} \rightleftharpoons 2\text{R}^{3+}$. Further details, summary of compositional data and references are given in Černý (1970); an intermediate composition close to the idealized formula is shown in Table 9 (anal. 4).

Boron has been found in subordinate amounts in several cookeites but the only mineral with substantial B is manandonite from Madagascar (Lacroix 1922). This unique Li, Al-boroaluminosilicate owes its origin to a breakdown and replacement of rubellite (Table 9, anal. 5). In contrast, cookeite is a widespread mineral in lithium pegmatites, replacing petalite, spodumene, eucryptite and lithium micas, or lining cavities as a very late mineral on quartz, (corroded) feldspar and mica crystals.

Bowleyite, bityite, beryllian margarite. Crystallochemically, all these minerals are derivatives of the brittle mica margarite or clintonite (Table 3; Ginsburg 1957b; Strunz 1962). They are generated by a combination of two substitutions: $\text{Al}^{\text{VI}} \rightleftharpoons \text{Be}^{2+} + \text{Li}^{1+}$ and $[\text{AlO}_4]^{5-} \rightleftharpoons [\text{BeO}_3(\text{OH})]^{5-}$ (Beus 1960). Bityite was the first one described (Lacroix 1908) and is intermediate in composition; beryllium margarites cover a wide range straddling the bityite chemistry (Vlasov and Kutukova 1959; Beus 1960) whereas bowleyite represents an extreme member of the group with the highest Be content known to date (Rowledge and Hayton 1948). Two chemical analyses illustrating the range of compositional variation are given in Table 9 (anal. 6 and 7).

Paragenetically, these minerals form either by replacement of plagioclase at pegmatite contacts with ultrabasic or carbonate wall-rocks, or by hydrothermal alteration of beryl. In this latter case, association with other secondary beryllium minerals such as bavenite is occasionally encountered.

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ADDENDA

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