Lithium–aluminium micas from the Meldon aplite, Devonshire, England

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SUMMARY. Chemical analyses of sixteen lithium-aluminium micas are presented along with their optical, physical, and X-ray data. Compositional variation, substitution relations in structural positions, and octahedral occupancy are discussed. The $2M_2$ structural types are found to crystallize in volatile-rich low-temperature environments whereas the 1M polytypes occur in comparatively volatile-poor and higher-temperature environments. Variation diagrams have been constructed to show the relationship between octahedral sites occupied by (Fe²⁺+Fe³⁺+ $\frac{1}{3}$ Mn+Ti) and refractive indices and specific gravities.

THE Meldon aplite, Devonshire, is a soda-lithia-rich aplite dyke about 20 to 25 m in thickness, occurring I km north-west of the main Dartmoor granite (Worth, 1920). Albite, quartz, lithium-aluminium micas, and orthoclase are the essential minerals and elbaite, topaz, fluorite, apatite, and petalite occur as accessory minerals of the aplite. Rarely, a pale-green secondary muscovite occurs along joints and fractures. The aplite is divided into three types. The 'blue aplite' is a light blue to azure, marginally chilled facies, which approaches albitite in composition. It consists of albite and quartz with minor amounts of apatite, tourmaline, orthoclase, and colourless lithium-aluminium mica. The 'white aplite' contains albite, quartz, and colourless to very light pink lithium-aluminium mica in order of abundance. The 'brown aplite' is coarse and from moderately to strongly metasomatized (autometasomatism) and contains larger amounts of pink to brown lithium-aluminium mica and quartz than the other two types described earlier. It may also contain notable amounts of elbaite, topaz, fluorite, and apatite. The Meldon aplite is cut by two types of thin (2 to 7 cm) veins; in one type, the 'pegmatite veins' are composed of orthoclase, quartz, lithiumaluminium mica, albite, and elbaite, and in the second type, the 'pegmatitic veins' consist of orthoclase, lithium-aluminium mica, quartz, albite, topaz, and petalite. The veins are richer in volatiles (F_2 , B_2O_3 , and H_2O) than the aplite types.

Chemistry: Chemical analyses of sixteen lepidolites are reported in Table I. SiO_2 was determined on a separate portion after removing fluorine (Groves, 1951) to minimize the loss of this constituent during evaporation following the main fusion. Total R_2O_3 and CaO and MgO were determined gravimetrically. The individual members of the R_2O_3 group were determined by spectrophotometric methods. The alkalis K_2O , Na₂O, and Li₂O were determined flame-photometrically. Rb₂O and Cs₂O

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	MHZ(D)	HM.1	MN.81	MBZ	MN.202	MN.71	MO.I	MN.74
SiO ₂	47.57	4 ^{8·} 47	47.74	49.76	48·18	47.87	48.78	50.32
TiO ₂	0.30	0.60	0.23	0.55	0.09	0.55	0.24	0.94
Al_2O_3	22.67	22.56	25.10	25.31	24.24	27.78	24.25	23.16
Fe ₂ O ₃	0.00	2.59	0.19	0.80	0.82	0.99	0.38	0.62
FeO	4·88	3.30	4.14	3.20	3.16	1.60	1.54	0.40
MnO	1.10	1.35	0.61	0.42	0.38	0.71	1.41	1.50
MgO	0.35	0.26	0.11	0.09	0.05	0.51	0.11	0.19
CaO	0.06	0.00	0.06	0.02	0.02	0.10	0.00	0.51
Li ₂ O	5.30	4.80	4.90	4.35	4.80	3.60	6.20	5.61
Na_2O	0.22	0.41	0.38	0.61	0.43	0.32	0.46	0.44
K ₂ O	9.82	9.74	9.70	9.20	9.80	9.74	9.57	9.49
Rb ₂ O	1.05	0.89	1.18	0.67	0.94	1.18	1.23	1.61
Cs_2O	0.22	0.27	0.14	0.17	0.10	0.33	0.37	0.51
F	8.93	5.27	8.10	3.06	7.93	5.00	5.10	5.92
H_2O^+	0.75	1.41	0.85	2.81	1.50	1.67	1.62	1.61
H ₂ O-	0.22	0.27	0.35	0.33	0.23	0.40	0.41	0.38
-	103.74	102.16	103.78	101.95	103.00	101.84	101.97	102.36
$\mathbf{F} \equiv \mathbf{O}$	3.77	2.22	3.54	1.67	2.92	2.11	2.12	2.40
Total	99.97	99 [.] 94	100.24	100.28	100.17	99 [.] 73	99·82	99·87
α	1.547	1.546	1.542	1.543	1.241	1.540	1.536	1.530
β	1.266	1.262	1.261	1.262	1.559	1.558	1.556	1.221
γ	1.241	1.572	1.564	1.268	1.564	1.263	1.560	1.528
$2V_{\alpha}$	39°	35°	—	34°	41°	37°	42°	
$D (g/cm^3)$	2.97	2.98	2.94	2.95	2.96	2.90	2.88	2.83
b (Å)	9.053	9.078	9.065	9.053	9.045	9.055	9.032	9.028
$c \sin \beta$ (Å)	9.961	9.970	9.973	9.979	9.979	10.026	10.012	10.060
Polymorph	${}_{1}M$	$\mathbf{I}M$	1M	${}_{1}M$	ıМ	ıМ	ıМ	ıМ
$^{*}A'$	0.261	0.811	0.824	0.852	0.864	0.882	o·884	0.935
Numbers of a	ions on the	basis of 2.	4 (O, OH,	F)				
Si	6.498	6.664	6.470	6.641	6.529	6.310	6.617	6.760
Al	1.202	1.336	1.530	1.359	1.471	1.681	1.383	1.240
Ζ	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al	2.148	2.321	2.480	2.623	2.450	2.842	2.494	2.427
Ti	0.031	0.062	0.024	0.023	0.000	0.022	0.025	0.002
Fe ³⁺	0.000	0.268	0.020	o•o8o	0.086	0.098	0.030	0.068
Fe ²⁺	0.222	0.379	0.470	0.322	0.328	0.177	0.175	0.045
Mn	0.127	0.124	0.070	0.047	0.044	0.079	0.162	0.136
Mg	0.065	0.023	0.022	0.018	0.004	0.041	0.022	0.038
Li	2.913	2.292	2.671	2.336	2.616	1.012	3.383	3.026
Y	5.841	5.529	5.757	5.484	5.567	5.171	6.300	5.835
Ca	0.000	0.000	0.000	0.002	0.002	0.017	0.000	0.030
Na	0.146	0.100	0.000	0.122	0.112	0.080	0.121	0.112
K	1.711	1.708	1.678	1.267	1.694	1.640	1.656	1.626
Rb	0.090	0.079	0.103	0.028	0.081	0.100	0.134	0.130
Cs	0.015	0.017	0.008	0.010	0.011	0.010	0.021	0.011
Х	1.971	1.913	1.897	1.200	1.902	1.862	1.932	1.921
он	0.683	1.294	0.760	2.203	1.085	1.471	1.466	1.444
F	3.858	2.292	3.472	1.672	3.300	2.088	2.188	2.516
OH+F	4.241	3.586	4.241	4.175	4.484	3.559	3.654	3.960
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 ${\tt TABLEI}. \ Lithium-aluminium\ micas\ from\ Meldon\ and\ their\ optical\ and\ physical\ properties$

* A' = LR/(LR + Fe'), where LR is the formula coefficient for Li or R_{oct}^{3+} , whichever is smaller, and 'Fe' is the sum of formula coefficients for Fe²⁺ and Mn²⁺.

Т	ABLE	II (cont.)
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	MO.3	M.2	МКС	MN.75	MN.21	M.200	MN.3	MN.7
SiO ₂	49.67	49.71	49.78	50.07	49.49	49.88	51.14	50.58
TiO ₂	0.18	0.15	0.12	0.15	0.18	0.51	0.15	0.55
Al ₂ O ₃	24.32	24.65	25.13	25.46	25.35	26.23	24.80	24.09
Fe ₂ O ₃	1.14	0.03	0.42	0.19	0.23	0.12	0.34	0.19
FeO	0.70	0.55	0.44	0.10	0.26	0.12	0.23	0.06
MnO	0.91	1.12	0.60	0.72	o·36	0.32	0.30	0.42
MgO	0.13	0.06	0.08	0.14	0.06	0.08	0.06	0.13
CaO	0.09	0.09	0.06	0.06	0.02	0.10	0.02	0.08
Li₂O	5.50	6.40	6.00	5.60	6.00	4.20	5.55	5.80
Na ₂ O	0.20	0.40	0.40	0.42	0.42	0.63	0.23	0.40
K ₂ O	9.74	9.80	9.70	9.80	9.90	9.70	9.81	10.20
Rb ₂ O	1.36	1.53	I.27	1.75	1.23	1.53	1.07	1.76
Cs ₂ O	0.26	0.37	0.25	0.21	0.47	0.46	0.36	0.45
F	6.73	7.20	6.30	5.84	6.40	8.10	6.70	6.93
H_2O^+	1.41	0.93	1.39	1.64	1.37	0.95	1.18	1.05
H ₂ O-	0.24	0.21	0.47	0.30	0.38	0.39	0.32	0.33
-	102.88	103.17	102.44	102.45	102.48	103.19	102.61	102.72
$\mathbf{F} = \mathbf{O}$	2.84	3.04	2.65	2.47	2.70	3.40	2.82	2.92
Total	100.04	100.13	99 [.] 79	99.98	99 [.] 78	99.79	99.79	<u>99</u> .80
α	1.237	1.528	1.231	1.529	1.530	1.241	1.533	1.231
β	1.555	1.550	1.550	1.548	1.550	1.562	1.553	1.221
γ	1.569	1.553	1.556	1.221	1.555	1.566	1.556	1.224
$2V_{\alpha}$	38	23°	40°	39°	30°	35	3°	34°
$D(g/cm^3)$	2.86	2.85	2.83	2.81	2.82	2.84	2.82	2.81
b (Å)	9.024	9.023	5.335	9.024	5.283	5.283	5.283	5.283
$c \sin \beta$ (Å)	10.190	10.060	19.912	10.071	19.766	19.968	19.875	19.962
Polymorph	ıМ	ıМ	$2M_2$	ıМ	$2M_3$	$2M_{2}$	$2M_2$	$2M_2$
*A'	0.932	0.943	0.928	0.967	o·974	0.976	0.979	0.979
Numbers of	ions on the	basis of 2	4 (O, OH,	F)				
Si	6.654	6-661	6.657	6.681	6.632	6.681	6.816	6.793
Al	1.346	1.339	1.343	1.310	1.368	1.319	1.184	1.207
Z	8.000	8.000	8.000	8.000	8.000	8.000	8.000	8.000
Al	2.495	2.555	2.619	2.685	2.636	2.823	2.712	2.608
Ti	0.010	0.012	0.012	0.012	0.010	0.021	0.012	0.023
Fe ³⁺	0.114	0.003	0.042	0.010	0.023	0.012	0.034	0.010
Fe ²⁺	0.028	0.025	0.040	0.011	0.020	0.014	0.026	0.001
Mn	0.103	0.130	0.068	0.082	0.041	0.042	0.034	0.051
Mg	0.026	0.012	0.016	0.028	0.012	0.016	0.012	0.026
Li	2.964	3.440	3.227	3.002	3.234	2.264	2.949	3.133
Y	5.799	6.186	6.036	5.842	5.994	5.195	5.779	5.867
Ca	0.013	0.013	0.000	0.000	0.007	0.027	0.010	0.011
Na	0.130	0.102	0.104	0.117	0.118	0.164	0.138	0.102
K	1.665	1.675	1.655	1.668	1.693	1.458	1.667	1.748
Rb	0.118	0.132	0.100	0.121	0.132	0.132	0.001	0 1 5 2
Cs	0.014	0.016	0.011	0.011	0.027	0.026	0.021	0.026
X	1.040	1.041	1.801	1.026	1.977	1.807	1.927	2.042
OH	1.201	0.831	I·2/1	I.7220	1.224	0.848	1.040	0.941
F	2.852	3.041	2.665	2.486	2.712	3.432	2.824	2.944
OH+F	4.113	3.882	3.906	3.945	3.936	4.280	3.873	3.885

* A' = LR/(LR+Fe'), where LR is the formula coefficient for Li or R_{oct}^{3+} , whichever is smaller, and 'Fe' is the sum of formula coefficients for Fe²⁺ and Mn²⁺.

were determined by X-ray fluorescence. The positive bias in the flame-photometric determinations of K_2O due to the interference of Rb_2O was corrected by preparing the interference curves. Fluorine was determined by the method of Grimaldi, Ingram, and Cuttitta (1955).

To investigate the systematic variations in the composition of these micas, a correlation matrix of their analytical data was obtained. The following pairs showed significant negative correlations: Li_2O and Al_2O_3 , SiO_2 and FeO, H_2O and F, K_2O and H_2O , Rb_2O and FeO. The inverse relations in the first three pairs are a common feature of the lithium-aluminium micas. The inverse relation between K_2O and H_2O is most probably due to $(H_3O)^+$ replacing K⁺. This relation has not been reported from the lithiumaluminium micas, but has been reported from the potassium-deficient dioctahedral micas (Radoslovich, 1963). There is a tendency for Rb and Cs to increase with increasing Li, as noted by Rinaldi *et al.* (1972), but for the Meldon lepidolites the Li/(Rb+Cs)ratios are considerably higher than those for the Li-Rb-Cs micas of the Tanco pegmatite reported by these authors. The inverse relation between Rb_2O and FeO is not a replacement relation, but due to petrogenetic factors. Geochemical studies of the rock samples from which these micas have been collected show that rocks richer in Fe are poorer in Rb (except in some late-stage veins).

Interpretation of lithium-aluminium micas in terms of end-members

The analyses of lithium–aluminium micas have been variously interpreted in terms of end-members by Hallimond (1925), Stevens (1938), and Winchell (1942). Hallimond's additional series of lepidolite–protolithionite consider the lithium content as fixed, with changes taking place by introduced *RO*. Stevens interpreted the lithium–aluminium micas in terms of four end-members, namely muscovite $K_4Al_8Al_4Si_{12}O_{40}(OH)_8$, polylithionite $K_4Li_8Al_4Si_{16}O_{40}(F_8)$, biotite $K_4Mg_{12}Al_4Si_{12}O_{40}(F,OH)_8$, and lithium– muscovite $K_4Li_6Al_6Al_4Si_{12}O_{40}(F,OH)_8$. Following Stevens (1938), lithium–aluminium micas have been interpreted by Berggren (1940, 1941) in terms of the lithium–muscovite series (with lithium–muscovite molecule more than 50 %), the muscovite series (with muscovite molecule more than 50 %), and the lepidolite series (with none of the molecules more than 50 %). Winchell (1942) used three end-members, namely polylithionite $K_2Li_4Al_2(OH,F)_4Si_8O_{20}$, paucilithionite $K_2Li_3Al_3(OH,F)_4Si_6Al_2O_{20}$, and protolithionite $K_2LiFe_4Al(OH,F)_4Si_6Al_2O_{20}$.

These methods of expressing lithium-aluminium micas in terms of end-members and end-member series, however, have been found to be unjustified on both theoretical and practical grounds.

Cationic substitutions in structural positions and octahedral occupancy

A detailed study of these relations led Foster (1960) instead to interpret the lithiumaluminium micas as if derived by isomorphous replacements from muscovite. According to Foster 'starting with muscovite, analyses and formulae of aluminum-lithium micas, arranged in order of increasing lithium content, are characterized by decrease

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in octahedral aluminum and in tetrahedral aluminum and by increase in silicon and octahedral occupancy. These changes can be interpreted as a result of progressive replacement of octahedral aluminum by lithium in ratios varying between 2 and 3 lithium for one octahedral aluminum.' The concept of this supposed replacement series (discontinuous) is very useful for studying substitutions in a given set of analyses. The lithium content and total octahedral occupancy have been used by Foster to classify the lithium-aluminium micas (fig. 1), as follows:

The lithian muscovite series starts with muscovite and extends to 0.85lithium occupancy and 2.50 total octahedral occupancy.

The lepidolite series starts from a lithium occupancy of 1.10 and a total octahedral occupany of 2.65 and extends to a lithium occupancy of 1.80 and more and a total octahedral occupancy of 3.00.

Mixed forms. Between an octahedral occupancy of 2.50 and 2.60 and lithium occupancy of 0.95 to 1.05 lie mixed structural forms.

For the purpose of studying cationic substitutions and the octahedral occu-



FIG. I. Nomenclature and relation between Li, R^{2+} (Fe²⁺, Mn²⁺, Mg), and octahedral R^{3+} (Al, Fe³⁺) +Ti⁴⁺ in lithium micas (after Foster, 1960).

pancy Foster's (1960) modification of Marshall's (1949) method of formula calculation was used. The results are presented diagrammatically in fig. 2. The following four relations may be observed:

For every octahedral site vacated by Al³⁺, 2·30 to 3.00 sites are occupied by Li⁺. This is illustrated in fig. 2a.

For each octahedral site occupied in excess of 2.00, the total sites occupied by Li+ vary from 1.50 to 1.70 (fig. 2b).

Li:Si (Si in excess of 3) varies from 1:2.80 to 5:1 (fig. 2c).

The relation between the formula sites occupied by Al^{3+} (+Fe³⁺) and Li⁺ is given in fig. 2d.

According to Foster's classification all but one of the present micas belong to the lepidolite series containing a variable amount of iron. For zinnwaldite, Hey (1955) gives the formula 2[K₂(Li, Fe²⁺, Al)₆ (Si, Al)₈O₂₀(F,OH)₄], noting that there is often considerable deficiency in the (Li, Fe²⁺, Al) group and that typically Li \approx 2, $Fe^{2+} \approx I_{\frac{1}{2}}$, and $Si \approx 6\frac{1}{2}$. Foster (1960) thus defined zinnwaldite as having a Li content of 2.00 ± 0.50 octahedral sites per unit cell and commented that the median value for Fe^{2+} in zinnwaldites is 1.10, i.e. nearer to 1.00 than to 1.50 octahedral sites. Rieder et al. (1970) also define zinnwaldite as having $Fe^{2+} > 1.00$. On this basis none of the Meldon micas can be classed as zinnwaldite, although many of the more iron-rich lepidolites are distinctly brown in hand specimen. This classification of Foster is 386C73 т



FIG. 2. (a) Relation between the octahedral sites occupied by Li and those vacated by Al. (b) Relation between the octahedral sites occupied by Li and those occupied in excess of 2.00. (c) Relation between the octahedral sites occupied by (Li R^{2+}) and the tetrahedral sites occupied by Si. (d) Relation between the formula sites occupied by Li and those occupied by Al + Fe³⁺.

superior to that of Winchell (1942) and of Stevens (1938) because it is based on lithium (and iron) content and octahedral occupancy.

These two series in the Li–Al micas are natural rather than arbitrary, and possesss characteristic physical or structural properties.

More recently the graphical system of Foster (1960) has been extended by Rieder *et al.* (1970), using a classification based largely on the ratio A' = LR/(LR+'Fe') where LR is the formula coefficient for Li or R_{oct}^{3+} , whichever is smaller, and 'Fe' is the sum of formula coefficients for Fe²⁺ and Mn²⁺. In Table I the analyses are arranged in order of increasing values of A'.

Polymorphism: X-ray diffraction patterns of the micas were obtained on a diffractometer using Cu- K_{α} radiation to determine their structural type. The samples M.2, MHZ(D), MBZ, Mo.3, MN.71, MN.75, M.21, MN.81, MN.74, MN.202, HM.1, and MO.1 were found to belong to the 1*M* polytype. The samples MN.7, MN.3, M.21, and MKC (all from the pegmatite and pegmatitic veins) belong to the $2M_2$ polytype. The sample M.200 is a mixed type, containing the $2M_2$ polytype together with some of the $2M_1$ type.

Studies of the structural types of lepidolites have been carried out by Hendricks and Jefferson (1939), Levinson (1953), and Smith and Yoder (1956). According to Levinson the variation in structural type can be correlated with the chemical composition, particularly the lithium content.

The normal muscovites may have as much as $3\cdot3 \frac{0}{10}$ Li₂O. The micas that contain $3\cdot4 \frac{0}{10}$ to $4\cdot0 \frac{0}{10}$ Li₂O show anomalous optics and transitional forms. Six-layer mono-

clinic lepidolites $2M_2$ contain 4.0% to 5.1% Li₂O. Those lepidolites which contain more than 5.1% Li₂O belong to the one-layer (1M) monoclinic polytype.

In the lithium-aluminium micas from the Meldon aplite, however, the samples HM.1, MN.202, MBZ, and MN.81 contain between $4 \cdot 0 \%$ to $5 \cdot 1 \%$ Li₂O, but do not belong to the $2M_2$ structural type. The samples MO.1, MN.74, MO.3, M₂, and MHZ(D) contain more than $5 \cdot 1 \%$ Li₂O and belong to the 1M polytype. The present work thus does not lend any support to Levinson's scheme, deviations from which have indeed been noted in several other studies, e.g. Heinrich (1967) and Rinaldi *et al.* (1972). It



FIGs. 3 and 4: FIG. 3 (left). The relationship between refractive indices and specific gravity and the octahedral site occupancy by $(Fe^{2+}+Fe^{3+}+Ti^{4+}+\frac{1}{3}Mn^{2+})$ calculated on the basis of 24 (O,OH,F). FIG. 4 (right). The relationship between the *b* cell parameter of 1*M* polymorphs and the octahedral sites occupied by $(Fe^{2+}+Fe^{3+})$.

can be seen from Table I, however, that when the analyses of the Meldon lepidolites are arranged in order of the increasing value of A' (Rieder *et al.*, 1970), with the single exception of specimen Mn.75, all the lepidolites with an A' value of > 0.943 have crystallized in the $2M_2$ structural type. It is important to note that all these $2M_2$ micas have been found to occur in the volatile-rich lower-temperature pegmatites and pegmatitic veins in the Meldon aplite. Following Smith and Yoder (1956), it is suggested that these two structural types formed in two distinct temperature, pressure, and compositional environments such that each environment favoured one polytype. In the Meldon aplite the $2M_2$ crystallized in volatile-rich (H₂O, F, B₂O₃) and low-temperature environments whereas the 1*M* polymorph crystallized in relatively volatile-poor and higher-temperature environments.

Optical and physical properties. The Li content of the lithium-aluminium micas does not influence the optics or the specific gravity appreciably; the earlier efforts of Winchell (1925, 1927, and 1942) to correlate optical properties in terms of

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end-members were of limited value as only his siderophyllite end-member molecule, $K_2Fe^{2+}(Al,Fe^{3+})_2Al_4Si_4O_{20}(OH,F)_4$, had any effect. It is fully appreciated, therefore, that the optical and physical methods can only predict the effect of cations such as Fe^{2+} , Fe^{3+} , Mn^{2+} , and Ti^{4+} , and diagrams have been constructed to present the relationship between these cations and the optical and physical properties. The refractive indices (determined by the single variation method with an accuracy of ± 0.001) and the specific gravities increase with the increase in the $(Fe^{2+}+Fe^{3+}+\frac{1}{3}Mn^{2+}+Ti)$, (fig. 3). The *b* cell dimensions of the 1*M* lepidolites have been plotted against the octahedral sites occupied by $(Fe^{2+}+Fe^{3+})$ in fig. 4, and are seen to increase with increase in $(Fe^{2+}+Fe^{3+})$. These results are in general agreement with those of Rieder *et al.* (1971).

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