# Lithium-aluminium micas from the Meldon aplite, Devonshire, England 

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#### Abstract

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 $2 \mathrm{M}_{2}$ structural types are found to crystallize in volatile-rich low-temperature environments whereas the 1 M polytypes occur in comparatively volatilepoor and higher-temperature environments. Variation diagrams have been constructed to show the relationship between octahedral sites occupied by $\left(\mathrm{Fe}^{2+}+\mathrm{Fe}^{3+}+\frac{1}{3} \mathrm{Mn}+\mathrm{Ti}\right)$ 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, I920). 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 $\left(\mathrm{F}_{2}, \mathrm{~B}_{2} \mathrm{O}_{3}\right.$, and $\left.\mathrm{H}_{2} \mathrm{O}\right)$ than the aplite types.

Chemistry: Chemical analyses of sixteen lepidolites are reported in Table I. $\mathrm{SiO}_{2}$ 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 $R_{2} \mathrm{O}_{3}$ and CaO and MgO were determined gravimetrically. The individual members of the $R_{2} \mathrm{O}_{3}$ group were determined by spectrophotometric methods. The alkalis $\mathrm{K}_{2} \mathrm{O}, \mathrm{Na}_{2} \mathrm{O}$, and $\mathrm{Li}_{2} \mathrm{O}$ were determined flame-photometrically. $\mathrm{Rb}_{2} \mathrm{O}$ and $\mathrm{Cs}_{2} \mathrm{O}$

[^0]TABLE I. Lithium-aluminium micas from Meldon and their optical and physical properties

|  | MHZ(D) | HM. I | MN.8I | MBZ | MN. 202 | MN. 71 | MO.I | MN. 74 |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $\mathrm{SiO}_{2}$ | 47.57 | 48.47 | 47.74 | $49 \cdot 76$ | 48.18 | $47 \cdot 87$ | $48 \cdot 78$ | $50 \cdot 32$ |
| $\mathrm{TiO}_{2}$ | 0.30 | $0 \cdot 60$ | 0.23 | $0 \cdot 22$ | 0.09 | 0.22 | 0.24 | 0.94 |
| $\mathrm{Al}_{2} \mathrm{O}_{3}$ | 22.67 | 22.56 | $25 \cdot 10$ | 25.31 | 24.54 | 27.78 | 24.25 | $23 \cdot 16$ |
| $\mathrm{Fe}_{2} \mathrm{O}_{3}$ | 0.00 | $2 \cdot 59$ | -.19 | 0.80 | $0 \cdot 85$ | 0.99 | $0 \cdot 38$ | 0.67 |
| FeO | $4 \cdot 88$ | 3.30 | 4.14 | $3 \cdot 20$ | $3 \cdot 16$ | 1.60 | I. 54 | $0 \cdot 40$ |
| MnO | I. 10 | $1 \cdot 32$ | 0.61 | 0.42 | 0.38 | 0.71 | I.4I | 1.20 |
| MgO | $0 \cdot 32$ | $0 \cdot 26$ | $0 \cdot 11$ | 0.09 | 0.02 | 0.21 | O.II | $0 \cdot 19$ |
| CaO | 0.06 | 0.00 | 0.06 | 0.05 | 0.05 | $0 \cdot 10$ | 0.00 | $0 \cdot 21$ |
| $\mathrm{Li}_{2} \mathrm{O}$ | $5 \cdot 30$ | $4 \cdot 80$ | $4 \cdot 90$ | $4 \cdot 35$ | 4.80 | $3 \cdot 60$ | $6 \cdot 20$ | $5 \cdot 61$ |
| $\mathrm{Na}_{2} \mathrm{O}$ | 0.55 | 0.41 | 0.38 | 0.61 | 0.43 | 0.35 | $0 \cdot 46$ | 0.44 |
| $\mathrm{K}_{2} \mathrm{O}$ | 9.82 | 9.74 | 9.70 | $9 \cdot 20$ | 9.80 | 9.74 | $9 \cdot 57$ | $9 \cdot 49$ |
| $\mathrm{Rb}_{2} \mathrm{O}$ | 1.02 | 0.89 | 1-18 | 0.67 | 0.94 | $1 \cdot 18$ | 1.53 | I.6I |
| $\mathrm{Cs}_{2} \mathrm{O}$ | $0 \cdot 25$ | 0.27 | 0.14 | 0.17 | -19 | - 33 | $0 \cdot 37$ | 0.21 |
| F | 8.93 | $5 \cdot 27$ | $8 \cdot 10$ | 3.96 | 7.93 | $5 \cdot 00$ | $5 \cdot 10$ | $5 \cdot 92$ |
| $\mathrm{H}_{2} \mathrm{O}^{+}$ | 0.75 | 1.41 | 0.85 | $2 \cdot 81$ | 1.20 | I. 67 | I. 62 | $1 \cdot 61$ |
| $\mathrm{H}_{2} \mathrm{O}^{-}$ | $0 \cdot 22$ | 0.27 | 0.35 | 0.33 | 0.53 | 0.49 | 0.41 | $0 \cdot 38$ |
|  | 103.74 | 102.16 | 103.78 | 101.95 | 103.09 | 101.84 | IOI•97 | 102.36 |
| $\mathrm{F} \equiv \mathrm{O}$ | $3 \cdot 77$ | $2 \cdot 22$ | $3 \cdot 54$ | 1.67 | 2.92 | $2 \cdot 11$ | $2 \cdot 15$ | 2.49 |
| Total | 99.97 | 99.94 | $100 \cdot 24$ | $100 \cdot 28$ | $100 \cdot 17$ | 99.73 | 99.82 | 99.87 |
| $\alpha$ | I. 547 | I. 546 | 1.542 | 1.543 | I.54I | $1 \cdot 540$ | 1.536 | I.530 |
| $\beta$ | 1.566 | r. 565 | $1 \cdot 561$ | 1.562 | 1.559 | 1-558 | 1.556 | 1.551 |
| $\gamma$ | I.571 | 1.572 | I 564 | 1.568 | $1 \cdot 564$ | 1.563 | 1.560 | 1-558 |
| ${ }_{2} \mathrm{~V}_{\alpha}$ | $39^{\circ}$ | $35^{\circ}$ | , | $34^{\circ}$ | $41^{\circ}$ | $37^{\circ}$ | $42^{\circ}$ | 5s |
| D (g/cm ${ }^{3}$ ) | 2.97 | $2 \cdot 98$ | $2 \cdot 94$ | $2 \cdot 95$ | $2 \cdot 96$ | $2 \cdot 90$ | $2 \cdot 88$ | $2 \cdot 83$ |
| $b$ ( $\AA$ ) | 9.053 | 9.078 | 9.065 | 9.053 | 9.045 | $9 \cdot 055$ | 9.032 | 9.028 |
| $c \sin \beta(\AA)$ | 9.961 | 9.970 | 9.973 | 9.979 | 9.979 | 10.026 | 10.017 | 10.060 |
| Polymorph | IM | IM | I $M$ | I $M$ | IM | I $M$ | I $M$ | I $M$ |
| ${ }^{*} A^{\prime}$ | $0 \cdot 76 \mathrm{I}$ | 0.811 | 0.824 | 0.852 | 0.864 | 0.882 | 0.884 | 0.935 |
| Numbers of ions on the basis of $24(\mathrm{O}, \mathrm{OH}, \mathrm{F})$ |  |  |  |  |  |  |  |  |
| Si | 6.498 | 6.664 | 6.470 | $6 \cdot 641$ | $6 \cdot 529$ | $6 \cdot 319$ | $6 \cdot 617$ | $6 \cdot 760$ |
| Al | 1.502 | 1.336 | I $\cdot 530$ | 1.359 | I-47I | I.68I | $1 \cdot 383$ | I. 240 |
| $Z$ | 8.000 | 8.000 | 8.000 | 8.000 | $8 \cdot 000$ | 8.000 | 8.000 | 8.000 |
| AI | 2.148 | $2 \cdot 32 \mathrm{I}$ | $2 \cdot 480$ | 2.623 | $2 \cdot 450$ | $2 \cdot 842$ | 2.494 | $2 \cdot 427$ |
| Ti | 0.031 | 0.062 | 0.024 | 0.023 | 0.009 | 0.022 | 0.025 | 0.095 |
| $\mathrm{Fe}^{3+}$ | 0.000 | 0.268 | 0.020 | 0.080 | 0.086 | 0.098 | 0.039 | 0.068 |
| $\mathrm{Fe}^{2+}$ | 0.557 | 0.379 | 0.470 | 0.357 | 0.358 | 0.177 | -.175 | 0.045 |
| Mn | $0 \cdot 127$ | -.154 | 0.070 | 0.047 | 0.044 | 0.079 | -. 162 | 0.136 |
| Mg | 0.065 | 0.053 | 0.022 | 0.018 | 0.004 | 0.041 | 0.022 | 0.038 |
| Li | 2.913 | $2 \cdot 292$ | $2 \cdot 671$ | $2 \cdot 336$ | 2.616 | 1.912 | $3 \cdot 383$ | 3.026 |
| $Y$ | $5 \cdot 84 \mathrm{I}$ | $5 \cdot 529$ | $5 \cdot 757$ | $5 \cdot 484$ | 5.567 | $5 \cdot 171$ | $6 \cdot 300$ | $5 \cdot 835$ |
| Ca | 0.009 | 0.000 | 0.009 | 0.007 | 0.007 | 0.014 | 0.000 | 0.030 |
| Na | $0 \cdot 146$ | - 109 | 0.099 | 0.157 | $0 \cdot 112$ | 0.089 | $0 \cdot 121$ | 0.115 |
| K | 1.711 | $1 \cdot 708$ | 1.678 | 1.567 | 1.694 | 1.640 | I. 656 | 1.626 |
| Rb | 0.090 | 0.079 | $\bigcirc \cdot 103$ | 0.058 | 0.081 | $0 \cdot 100$ | -.134 | -139 |
| Cs | 0.015 | 0.017 | 0.008 | 0.010 | 0.011 | 0.019 | 0.021 | $0 \cdot 011$ |
| $X$ | I.97I | 1.913 | r.897 | 1.799 | 1.905 | I. 862 | 1.932 | 1.92I |
| OH | 0.683 | I-294 | 0.769 | 2.503 | 1.085 | 1.471 | 1.466 | I 444 |
| F | 3.858 | $2 \cdot 292$ | $3 \cdot 472$ | 1. 672 | 3.399 | 2.088 | 2.188 | $2 \cdot 516$ |
| $\mathrm{OH}+\mathrm{F}$ | 4.541 | 3.586 | $4 \cdot 241$ | 4.175 | 4.484 | 3.559 | 3.654 | 3.960 |

${ }^{*} A^{\prime}=L R /\left(L R+{ }^{`} \mathrm{Fe}\right)$, where $L R$ is the formula coefficient for Li or $R_{\text {oct }}^{8+}$, whichever is smaller, and ' Fe ' is the sum of formula coefficients for $\mathrm{Fe}^{2+}$ and $\mathrm{Mn}^{2+}$.

Table II (cont.)

|  | MO. 3 | M. 2 | MKC | MN. 75 | MN. 21 | M. 200 | MN. 3 | MN. 7 |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $\mathrm{SiO}_{2}$ | $49 \cdot 67$ | $49 \cdot 71$ | $49 \cdot 78$ | 50.07 | 49'49 | $49 \cdot 88$ | 5I•14 | $50 \cdot 58$ |
| $\mathrm{TiO}_{2}$ | 0.18 | O.12 | 0.15 | $0 \cdot 12$ | 0.18 | $0 \cdot 21$ | 0.12 | $0 \cdot 22$ |
| $\mathrm{Al}_{2} \mathrm{O}_{3}$ | 24.32 | 24.65 | 25.13 | $25 \cdot 46$ | $25 \cdot 35$ | $26 \cdot 23$ | 24.80 | 24.09 |
| $\mathrm{Fe}_{2} \mathrm{O}_{3}$ | I-14 | 0.03 | 0.42 | 0.19 | 0.23 | $0 \cdot 15$ | 0.34 | $0 \cdot 19$ |
| Fc | 0.70 | 0.22 | 0.44 | $0 \cdot 10$ | 0.26 | 0.12 | 0.23 | 0.06 |
| MnO | 0.91 | 1.15 | 0.60 | 0.72 | 0.36 | 0.37 | 0.30 | 0.45 |
| MgO | $0 \cdot 13$ | 0.06 | 0.08 | $0 \cdot 14$ | $0 \cdot 06$ | $0 \cdot 08$ | 0.06 | 0.13 |
| CaO | 0.09 | 0.09 | 0.06 | 0.06 | 0.05 | $0 \cdot 19$ | 0.07 | 0.08 |
| $\mathrm{Li}_{2} \mathrm{O}$ | 5.50 | 6.40 | 6.00 | $5 \cdot 60$ | 6.00 | $4 \cdot 20$ | 5.55 | $5 \cdot 80$ |
| $\mathrm{Na}_{2} \mathrm{O}$ | 0.50 | 0.40 | $0 \cdot 40$ | 0.45 | 0.45 | 0.63 | 0.53 | 0.40 |
| $\mathrm{K}_{2} \mathrm{O}$ | $9 \cdot 74$ | 9.80 | $9 \cdot 70$ | $9 \cdot 80$ | $9 \cdot 90$ | $9 \cdot 70$ | $9 \cdot 81$ | $10 \cdot 20$ |
| $\mathrm{Rb}_{2} \mathrm{O}$ | $1 \cdot 36$ | 1.53 | 1.27 | I.75 | 1.53 | 1.53 | I.07 | I•76 |
| $\mathrm{Cs}_{2} \mathrm{O}$ | 0.26 | 0.37 | 0.25 | 0.21 | 0.47 | $0 \cdot 46$ | $0 \cdot 36$ | 0.45 |
| F | $6 \cdot 73$ | $7 \cdot 20$ | $6 \cdot 30$ | $5 \cdot 84$ | 6.40 | 8.10 | $6 \cdot 70$ | 6.93 |
| $\mathrm{H}_{2} \mathrm{O}^{+}$ | 1-41 | 0.93 | $1 \cdot 39$ | 1. 64 | 1.37 | 0.95 | I•18 | 1.05 |
| $\mathrm{H}_{2} \mathrm{O}^{-}$ | 0.24 | 0.51 | 0.47 | 0.30 | $0 \cdot 38$ | 0.39 | 0.35 | 0.33 |
|  | 102.88 | 103.17 | 102.44 | 102.45 | 102.48 | 103.19 | 102.6I | 102.72 |
| $\mathbf{F}=\mathbf{O}$ | $2 \cdot 84$ | $3 \cdot 04$ | $2 \cdot 65$ | $2 \cdot 47$ | $2 \cdot 70$ | 3.40 | 2.82 | $2 \cdot 92$ |
| Total | 100.04 | $100 \cdot 13$ | 99•79 | $99 \cdot 98$ | 99•78 | 99•79 | $99 \cdot 79$ | 99.80 |
| $\alpha$ | I. 537 | 1.528 | I.53I | I. 529 | 1.530 | 1-541 | I. 533 | I'531 |
| $\beta$ | 1. 555 | 1.550 | 1.550 | I 548 | 1.550 | I $\cdot 562$ | I-553 | I.55I |
| $\gamma$ | 1.569 | 1.553 | I $\cdot 556$ | 1.551 | I. 555 | 1.566 | I-556 | I 5557 |
| ${ }_{2} \mathrm{~V}_{x}$ | 38 | $23^{\circ}$ | $40^{\circ}$ | $39^{\circ}$ | $30^{\circ}$ | 35 | $3^{\circ}$ | $34^{\circ}$ |
| D (g/cm ${ }^{3}$ ) | 2.86 | 2.85 | 2.83 | 2.8 I | 2.82 | $2 \cdot 84$ | $2 \cdot 82$ | 2.81 |
| $b(\AA)$ | 9.024 | $9 \cdot 023$ | $5 \cdot 335$ | 9.024 | $5 \cdot 283$ | $5 \cdot 283$ | $5 \cdot 283$ | $5 \cdot 283$ |
| $c \sin \beta(\AA)$ | 10.190 | 10.060 | 19.912 | 10.071 | 19.766 | 19.968 | 19.875 | I9.962 |
| Polymorph | $1 M$ | $\mathrm{I} M$ | $2 M_{2}$ | $\mathrm{I} M$ | $2 M_{3}$ | $2 M_{2}$ | $2 M_{2}$ | $2 M_{2}$ |
| $* A^{\prime}$ | 0.935 | 0.943 | 0.958 | $0.967$ | 0.974 | 0.976 | 0.979 | 0.979 |
| Numbers of ions on the basis of $24(\mathrm{O}, \mathrm{OH}, \mathrm{F})$ |  |  |  |  |  |  |  |  |
| Si | 6.654 | $6 \cdot 661$ | 6.657 | $6 \cdot 681$ | 6.632 | $6 \cdot 68 \mathrm{I}$ | 6.8I6 | 6.793 |
| Al | I. 346 | I $\cdot 339$ | I. 343 | I. 319 | I. 368 | I-319 | I•184 | I-207 |
| $Z$ | $8 \cdot 000$ | 8.000 | 8.000 | $8 \cdot 000$ | $8 \cdot 000$ | 8.000 | $8 \cdot 000$ | $8 \cdot 000$ |
| Al | $2 \cdot 495$ | 2.555 | $2 \cdot 619$ | 2.685 | 2.636 | $2 \cdot 823$ | $2 \cdot 712$ | $2 \cdot 608$ |
| Ti | 0.019 | 0.012 | 0.015 | 0.012 | 0.019 | 0.021 | 0.012 | 0.023 |
| $\mathrm{Fe}^{3+}$ | 0.114 | 0.003 | $0 \cdot 042$ | -0.019 | 0.023 | $0 \cdot 015$ | 0.034 | 0.019 |
| $\mathrm{Fe}^{+}$ | 0.078 | 0.025 | $0 \cdot 049$ | 0.011 | 0.029 | 0.014 | 0.026 | 0.007 |
| Mn | $0 \cdot 103$ | 0.130 | 0.068 | 0.082 | 0.041 | 0.042 | 0.034 | 0.051 |
| Mg | $0 \cdot 026$ | 0.012 | 0.016 | 0.028 | 0.012 | 0.016 | 0.012 | 0.026 |
| Li | $2 \cdot 964$ | 3.449 | 3.227 | 3.005 | $3 \cdot 234$ | $2 \cdot 264$ | 2.949 | $3 \cdot 133$ |
| $Y$ | $5 \cdot 799$ | $6 \cdot 186$ | 6.036 | $5 \cdot 842$ | 5.994 | 5•195 | $5 \cdot 779$ | 5.867 |
| Ca | 0.013 | 0.013 | 0.009 | 0.009 | 0.007 | 0.027 | 0.010 | 0.01 I |
| Na | 0.130 | $0 \cdot 105$ | $0 \cdot 104$ | 0.117 | O.II8 | $0 \cdot 164$ | 0.138 | $0 \cdot 105$ |
| K | 1. 665 | I. 675 | 1. 655 | I. 668 | I. 693 | 1.458 | I. 667 | 1.748 |
| Rb | $0 \cdot 118$ | 0.132 | 0.109 | 0.151 | 0.132 | O.132 | 0.091 | 0.152 |
| Cs | 0.014 | 0.016 | 0.014 | 0.011 | 0.027 | 0.026 | 0.021 | 0.026 |
| $X$ | I 940 | I.94I | I-89I | I•956 | I. 977 | I. 807 | I.927 | 2.042 |
| OH | I $\cdot 261$ | 0.83 I | I $\cdot 24 \mathrm{I}$ | I-459 | I $\cdot 224$ | 0.848 | I-049 | 0.941 |
| F | $2 \cdot 852$ | 3.051 | $2 \cdot 665$ | $2 \cdot 486$ | $2 \cdot 712$ | $3 \cdot 432$ | $2 \cdot 824$ | 2.944 |
| $\mathrm{OH}-\mathrm{F}$ | 4.113 | $3 \cdot 882$ | 3.906 | $3 \cdot 945$ | 3.936 | $4 \cdot 280$ | $3 \cdot 873$ | 3.885 |

* $A^{\prime}:-L R /\left(L R+{ }^{\prime} \mathrm{Fe}\right.$ '), where $L R$ is the formula coefficient for Li or $R_{o c t}^{3+}$, whichever is smaller, and ' Fe ' is the sum of formula coefficients for $\mathrm{Fe}^{2+}$ and $\mathrm{Mn}^{2+}$.
were determined by X-ray fluorescence. The positive bias in the flame-photometric determinations of $\mathrm{K}_{2} \mathrm{O}$ due to the interference of $\mathrm{Rb}_{2} \mathrm{O}$ 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: $\mathrm{Li}_{2} \mathrm{O}$ and $\mathrm{Al}_{2} \mathrm{O}_{3}, \mathrm{SiO}_{2}$ and $\mathrm{FeO}, \mathrm{H}_{2} \mathrm{O}$ and $\mathrm{F}, \mathrm{K}_{2} \mathrm{O}$ and $\mathrm{H}_{2} \mathrm{O}$, $\mathrm{Rb}_{2} \mathrm{O}$ and FeO . The inverse relations in the first three pairs are a common feature of the lithium-aluminium micas. The inverse relation between $\mathrm{K}_{2} \mathrm{O}$ and $\mathrm{H}_{2} \mathrm{O}$ is most probably due to $\left(\mathrm{H}_{3} \mathrm{O}\right)^{+}$replacing $\mathrm{K}^{+}$. 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 $\mathrm{Li} /(\mathrm{Rb}+\mathrm{Cs})$ ratios are considerably higher than those for the $\mathrm{Li}-\mathrm{Rb}-\mathrm{Cs}$ micas of the Tanco pegmatite reported by these authors. The inverse relation between $\mathrm{Rb}_{2} \mathrm{O}$ 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 $R O$. Stevens interpreted the lithium-aluminium micas in terms of four end-members, namely muscovite $\mathrm{K}_{4} \mathrm{Al}_{8} \mathrm{Al}_{4} \mathrm{Si}_{12} \mathrm{O}_{40}(\mathrm{OH})_{8}$, polylithionite $\mathrm{K}_{4} \mathrm{Li}_{8} \mathrm{Al}_{4} \mathrm{Si}_{16} \mathrm{O}_{40}\left(\mathrm{~F}_{8}\right)$, biotite $\mathrm{K}_{4} \mathrm{Mg}_{12} \mathrm{Al}_{4} \mathrm{Si}_{12} \mathrm{O}_{40}(\mathrm{~F}, \mathrm{OH})_{8}$, and lithiummuscovite $\mathrm{K}_{4} \mathrm{Li}_{6} \mathrm{Al}_{6} \mathrm{Al}_{4} \mathrm{Si}_{12} \mathrm{O}_{40}(\mathrm{~F}, \mathrm{OH})_{8}$. Following Stevens (I938), 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 $\mathrm{K}_{2} \mathrm{Li}_{4} \mathrm{Al}_{2}\left(\mathrm{OH}, \mathrm{F}_{4} \mathrm{Si}_{8} \mathrm{O}_{20}\right.$, paucilithionite $\mathrm{K}_{2} \mathrm{Li}_{3} \mathrm{Al}_{3}\left(\mathrm{OH}, \mathrm{F}_{4} \mathrm{Si}_{6} \mathrm{Al}_{2} \mathrm{O}_{20}\right.$, and protolithionite $\mathrm{K}_{2} \mathrm{LiFe}_{4} \mathrm{Al}\left(\mathrm{OH}, \mathrm{F}_{4} \mathrm{Si}_{6} \mathrm{Al}_{2} \mathrm{O}_{20}\right.$.

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
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. r), as follows:

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

The lepidolite series starts from a lithium occupancy of $1 \cdot 10$ and a total octahedral occupany of 2.65 and extends to a lithium occupancy of $1 \cdot 80$ 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


Fig. I. Nomenclature and relation between $\mathrm{Li}, \mathrm{R}^{2+}$ $\left(\mathrm{Fe}^{2+}, \mathrm{Mn}^{2+}, \mathrm{Mg}\right)$, and octahedral $\mathrm{R}^{3+}\left(\mathrm{Al}, \mathrm{Fe}^{3+}\right)$ $+\mathrm{Ti}^{4+}$ in lithium micas (after Foster, 1960). substitutions and the octahedral occupancy 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 $\mathrm{Al}^{3+}, 2 \cdot 30$ to 3.00 sites are occupied by $\mathrm{Li}^{+}$. This is illustrated in fig. $2 a$.

For each octahedral site occupied in excess of $2 \cdot 00$, the total sites occupied by $\mathrm{Li}^{+}$ vary from I. 50 to I 70 (fig. $2 b$ ).
$\mathrm{Li}: \mathrm{Si}$ ( Si in excess of 3 ) varies from $\mathrm{I}: 2 \cdot 80$ to $5: 1$ (fig. $2 c$ ).
The relation between the formula sites occupied by $\mathrm{Al}^{3+}\left(+\mathrm{Fe}^{3+}\right)$ and $\mathrm{Li}^{+}$is given in fig. $2 d$.

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 $2\left[\mathrm{~K}_{2}\left(\mathrm{Li}, \mathrm{Fe}^{2+}, \mathrm{Al}_{6}(\mathrm{Si}, \mathrm{Al})_{8} \mathrm{O}_{20}(\mathrm{~F}, \mathrm{OH})_{4}\right]\right.$, noting that there is often considerable deficiency in the ( $\mathrm{Li}, \mathrm{Fe}^{2+}, \mathrm{Al}$ ) group and that typically $\mathrm{Li} \approx 2$, $\mathrm{Fe}^{2+} \approx \mathrm{I} \frac{1}{2}$, and $\mathrm{Si} \approx 6 \frac{1}{2}$. Foster ( 1960 ) thus defined zinnwaldite as having a Li content of $2.00 \pm 0.50$ octahedral sites per unit cell and commented that the median value for $\mathrm{Fe}^{2+}$ in zinnwaldites is $\mathrm{I} \cdot 10$, i.e. nearer to $\mathrm{I} \cdot 00$ than to $\mathrm{I} \cdot 50$ octahedral sites. Rieder et al. (1970) also define zinnwaldite as having $\mathrm{Fe}^{2+}>\mathrm{I} \cdot 00$. 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


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 $R^{2 \dagger}$ ) and the tetrahedral sites occupied by Si. (d) Relation between the formula sites occupied by I.i and those occupied by $\mathrm{Al}+\mathrm{Fe}^{3}$.
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 $\mathrm{Li}-\mathrm{Al}$ 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 $A^{\prime}:--L R(L R+' F e ')$ where $L R$ is the formula coefficient for Li or $R_{\mathrm{sct}}^{3-}$, whichever is smaller, and ' Fe ' is the sum of formula coefficients for $\mathrm{Fe}^{2+}$ and $\mathrm{Mn}^{2+}$. In Table I the analyses are arranged in order of increasing values of $A^{\prime}$.

Polymorphism: X-ray diffraction patterns of the micas were obtained on a diffractometer using $\mathrm{Cu}-K_{\alpha}$ radiation to determine their structural type. The samples M.2, MHZ(D), MBZ, Mo.3, MN.7I, MN.75, M.21, MN.8I, MN.74, MN.202, HM.i, and MO.i were found to belong to the I $M$ polytype. The samples MN.7, MN.3, M.2I, and MKC (all from the pegmatite and pegmatitic veins) belong to the $2 M_{2}$ polytype. The sample M. 200 is a mixed type, containing the $2 M_{2}$ polytype together with some of the $2 M_{1}$ 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 $3.3 \% \mathrm{Li}_{2} \mathrm{O}$. The micas that contain $3.4 \%$ to $4.0 \% \mathrm{Li}_{2} \mathrm{O}$ show anomalous optics and transitional forms. Six-layer mono-
clinic lepidolites $2 \mathrm{M}_{2}$ contain $4.0 \%$ to $5 \cdot \mathrm{I} \% \mathrm{Li}_{2} \mathrm{O}$. Those lepidolites which contain more than $5 \cdot 1 \% \mathrm{Li}_{2} \mathrm{O}$ belong to the one-layer ( $\mathrm{I} M$ ) monoclinic polytype.

In the lithium-aluminium micas from the Meldon aplite, however, the samples HM.I, MN.202, MBZ, and MN.8I contain between $4.0 \%$ to $5 . \mathrm{I} \% \mathrm{Li}_{2} \mathrm{O}$, but do not belong to the $2 M_{2}$ structural type. The samples MO.I, MN. 74, MO.3, M ${ }_{2}$, and MHZ(D) contain more than $5 \cdot 1 \% \mathrm{Li}_{2} \mathrm{O}$ and belong to the $\mathrm{I} M$ 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 ( $\mathrm{Fe}^{2+}+\mathrm{Fe}^{3+}+\mathrm{Ti}^{4+}+\frac{1}{3} \mathrm{Mn}^{2+}$ ) calculated on the basis of 24 ( $\mathrm{O}, \mathrm{OH}, \mathrm{F}$ ). Fig. 4 (right). The relationship between the $b$ cell parameter of i $M$ polymorphs and the octahedral sites occupied by $\left(\mathrm{Fe}^{2+}+\mathrm{Fe}^{3+}\right)$.
can be seen from Table I, however, that when the analyses of the Meldon lepidolites are arranged in order of the increasing value of $A^{\prime}$ (Rieder et al., 1970), with the single exception of specimen Mn.75, all the lepidolites with an $A^{\prime}$ value of $>0.943$ have crystallized in the $2 M_{2}$ structural type. It is important to note that all these $2 M_{2}$ 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 $2 M_{2}$ crystallized in volatile-rich $\left(\mathrm{H}_{2} \mathrm{O}, \mathrm{F}, \mathrm{B}_{2} \mathrm{O}_{3}\right)$ and low-temperature environments whereas the $\mathrm{I} M$ 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
end-members were of limited value as only his siderophyllite end-member molecule, $\mathrm{K}_{2} \mathrm{Fe}^{2+}\left(\mathrm{Al}, \mathrm{Fe}^{3+}\right)_{2} \mathrm{Al}_{4} \mathrm{Si}_{4} \mathrm{O}_{20}(\mathrm{OH}, \mathrm{F})_{4}$, had any effect. It is fully appreciated, therefore, that the optical and physical methods can only predict the effect of cations such as $\mathrm{Fe}^{2+}$, $\mathrm{Fe}^{3+}, \mathrm{Mn}^{2+}$, and $\mathrm{Ti}^{4+}$, 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 $\pm 0.00 \mathrm{I}$ ) and the specific gravities increase with the increase in the ( $\mathrm{Fe}^{2+}+\mathrm{Fe}^{3+}+\frac{1}{3} \mathrm{Mn}^{2+}+\mathrm{Ti}$ ), (fig. 3). The $b$ cell dimensions of the $I M$ lepidolites have been plotted against the octahedral sites occupied by $\left(\mathrm{Fe}^{2+}+\mathrm{Fe}^{3+}\right)$ in fig. 4, and are seen to increase with increase in $\left(\mathrm{Fe}^{2+}+\mathrm{Fe}^{3+}\right)$. These results are in general agreement with those of Rieder et al. (1971).

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