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# The correlation between lithium and magnesium in trioctahedral micas: Improved equations for $\text{Li}_2\text{O}$ estimation from $\text{MgO}$ data

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## ABSTRACT

A major disadvantage of the electron microprobe is its inability to determine Li, which may make up an essential component in micaceous minerals. Correct classification of micas and proper calculation of their formulae require alternative methods. One of these is the indirect estimation of the lithium concentration by empirical approaches based on element correlations. Relationships between  $\text{Li}_2\text{O}$  and  $\text{SiO}_2$  have long been used for this purpose; however, they are valid only for a limited range of mica compositions (i.e. Li-rich, Mg-poor varieties).

In this paper we report the results from a renewed study of the correlation between  $\text{Li}_2\text{O}$  and  $\text{MgO}$  contents in trioctahedral micas. It is demonstrated that the relations between both oxides are strongly dependent upon the geological environment in which the mica was formed. It is necessary to distinguish a 'normal group', which comprises micas from 'common' S- and I-type magmatic rocks and most metamorphic rocks, a 'low Li-Mg group', to which belong Fe-rich micas from non-alkaline rocks of A-type affinity, and a 'high Li-Mg group' which includes micas from either peraluminous or peralkaline igneous rocks and Mg-enriched wall-rocks. These wall rocks supplied additional magnesium to the mica-forming environment. We offer empirical expressions relating  $\text{Li}_2\text{O}$  and  $\text{MgO}$  contents for these three types which, in many cases, allow a sufficiently good estimation of the lithium content in trioctahedral micas of widely varying compositions.

**KEYWORDS:** mica, lithium, magnesium, correlation, classification.

## Introduction

KNOWLEDGE of the Li contents in micas is important for petrogenetic or classification purposes. Bulk analysis of mineral separates is a poor substitute for *in situ* microanalysis because zoning and heterogeneities on a grain-size scale are common. Lithium contents cannot be obtained by electron microprobe, and other methods of microanalysis capable of determining this element (secondary ion-mass spectrometry and laser ablation-inductively coupled plasma mass spectrometry) are still out of reach for most researchers.

As demonstrated by Tindle and Webb (1990), Li in trioctahedral micas can be estimated with

good accuracy from conventional electron microprobe analysis by using the equation  $\text{Li}_2\text{O} = (0.287 \times \text{SiO}_2) - 9.552$ . These authors limited the applicability of this equation to micas with less than 8 wt.%  $\text{MgO}$ . In a companion paper (Tischendorf *et al.*, 1997), the present authors re-examined the relationship between  $\text{Li}_2\text{O}$  and  $\text{SiO}_2$  and, using a different set of compositional data, came up virtually with the same result,  $\text{Li}_2\text{O} = (0.289 \times \text{SiO}_2) - 9.658$ . To minimize error, these authors recommended this equation only be used for micas with  $\text{MgO} < 6$  wt.% and  $\text{SiO}_2 > 34$  wt.%.

Using all the data on micas available to us, we have tested the correlation between  $\text{Li}_2\text{O}$  and  $\text{SiO}_2$

for certain MgO intervals, and established that estimating the Li<sub>2</sub>O content from SiO<sub>2</sub> content is only sensible for samples with a maximum of 3 wt.% MgO. As shown in Table 1, beyond this point the correlation coefficients quickly decline. This proves that the Li<sub>2</sub>O-SiO<sub>2</sub> correlation is strictly applicable only to micas containing <3 wt.% MgO. Consequently, this equation is not applicable to 'common' micas from magmatic and metamorphic rocks, which typically have MgO contents greater than that critical value. To look for an alternative for this particular population of micas, a new Li<sub>2</sub>O/MgO-correlation would appear necessary (Tischendorf *et al.*, 1997).

For the Li<sub>2</sub>O-MgO relationship in trioctahedral micas, Tischendorf *et al.* (1997) proposed the equation  $Li_2O = [2.7/(0.35 + MgO)] - 0.13$  (goodness of fit ( $R^2$ ) = 0.88; number of mica analyses (n) = 434). However, a number of exceptions were noted which seriously limited its applicability. In this paper we present a solution to this problem. A major advantage of the Li<sub>2</sub>O-MgO correlation is that the entire variation of the trioctahedral micas can be covered by one expression because of the completely antipathetic behaviour of both oxides. Also, both oxides are components of the octahedral layer of the micas and therefore knowledge of the Li<sub>2</sub>O contents is essential for their classification (Foster, 1960*a,b*; Tischendorf *et al.*, 1997).

This renewed study of the relationship between Li<sub>2</sub>O and MgO in trioctahedral mica compositional data is based upon a compilation of more

than 1200 mica analyses, either from sources in the literature or from unpublished data sets of the authors. Generally, published analyses of even the most Li-rich micas (lepidolite) include MgO whereas Li<sub>2</sub>O is rarely analysed in Mg-rich micas (phlogopite). This is not a major problem because the Li<sub>2</sub>O contents in Mg-rich micas are relatively low anyway, and not so important for classification and petrological interpretation. On the other hand, accurate determination of the MgO contents in Li-rich micas is essential, because small amounts of MgO (0.5–0.01 wt.%) correspond to greater contents of Li<sub>2</sub>O (2–6 wt.%).

Our evaluation of this data set assumes that the Li<sub>2</sub>O and MgO values are analytically correct. To minimize the effect of alteration we did not include micas with K<sub>2</sub>O (+ Na<sub>2</sub>O + Rb<sub>2</sub>O + Cs<sub>2</sub>O + CaO) <8 wt.%.

## Results

Analysing the relationship between Li<sub>2</sub>O and MgO in trioctahedral micas in more detail, we are now able to improve the information contained in Tischendorf *et al.* (1997, Fig. 4*f* and equation tri 2). This improvement is mainly due to grouping of the micas into four populations which are genetically well defined. These are:

(1) Most of the available analyses (n = 870; 72%) belong to the 'normal group' and more than 98% of them are from igneous rocks and their derivatives (pegmatites, aplites) as well as related orthomagmatic ore deposits (tin-tungsten). In detail, granites and granodiorites largely predominate over more mafic rocks (diorites, gabbros and ultramafic rocks). The remaining analyses (2%) are from granulites and paragneisses. Data sources for Li<sub>2</sub>O and MgO analyses of the normal group micas are: Cundy *et al.* (1960), Müller (1966), Heinrich (1967), Franzini and Sartori (1969), Němec (1969), Rimsaite (1970), Hall (1971), Rimsaite (1971), Rub *et al.* (1971), Vejnar (1971), Yakovleva (1972), Koval' *et al.* (1972*a*), Koval' *et al.* (1972*b*), de Albuquerque (1973), Chaudhry and Howie (1973), Hazen and Burnham (1973), Hejtman (1975), Bokonbaev (1976), Fiala *et al.* (1976), Harada *et al.* (1976), Kramer (1976), Neiva (1976), Al-Saleh *et al.* (1977), Kabesh *et al.* (1977), Lapidés *et al.* (1977), Pomârleanu and Movileanu (1977–1978), Kozlov *et al.* (1978), Barrière and Cotton (1979), Kozlov *et al.* (1979), Bea (1980), Neiva (1980), Rub and Rub (1980), Volkov and Gorbacheva (1980), Luecke (1981),

TABLE 1. Goodness of fit ( $R^2$ ) for the correlation between Li<sub>2</sub>O and SiO<sub>2</sub> in trioctahedral micas in dependence on MgO

MgO interval (wt.%)	n	$R^2$
< 0.1	103	0.76
0.1–0.5	144	0.86
0.5–1.0	88	0.88
1.0–2.0	90	0.90
2.0–3.0	57	0.90
3.0–4.0	42	0.48
4.0–5.0	59	0.37
5.0–6.0	50	0.17
6.0–7.0	64	0.11
7.0–8.0	70	0.27

Němec (1981), Radvanec and Radvanová (1981), Smith *et al.* (1982), Borodanov (1983), Gavrikova (1983), Němec (1983), Zaritskiy *et al.* (1983), Bagdasarov *et al.* (1984), Minařík *et al.* (1984), Sun Shihua (1984), Bagdasarov *et al.* (1985), Edmunds *et al.* (1985), Nedosekin (1985), Orlov (1985), Shearer *et al.* (1986), Fonteilles (1987), Jolliff *et al.* (1987), Korenbaum (1987), Monier *et al.* (1987), Neiva *et al.* (1987), Neves and Godinho (1987), Savage *et al.* (1987), Stone *et al.* (1988), Trunilina and Royev (1988), Malyshonok (1989), Grew *et al.* (1990), Němec (1990a), Silva and Neiva (1990), Neiva and Gomes (1991), Cuney *et al.* (1992), Ilton *et al.* (1992), Pechar and Rykl (1992), Taylor (1992), Uhlíř (1992), Hecht (1993), Neiva (1993), Němec and Povondra (1993), Bea *et al.* (1994), Bigi and Brigatti (1994), Breiter (1995), Černý *et al.* (1995), Grew *et al.* (1995), Rieder *et al.* (1995), Roda *et al.* (1995), Rub *et al.* (1996), Breiter *et al.* (1997), as well 178 published (Tischendorf *et al.*, 1969; Gottesmann *et al.*, 1994; Tischendorf *et al.*, 1997; Table 3) and unpublished analyses of the authors.

The Li<sub>2</sub>O/MgO distribution in this normal group (as well as for the micas of the other groups) is represented by a hyperbola with values clustered subparallel with the axes. Taking the logarithm of the values results in an improved reliability for both low Li<sub>2</sub>O and MgO contents (Fig. 1).

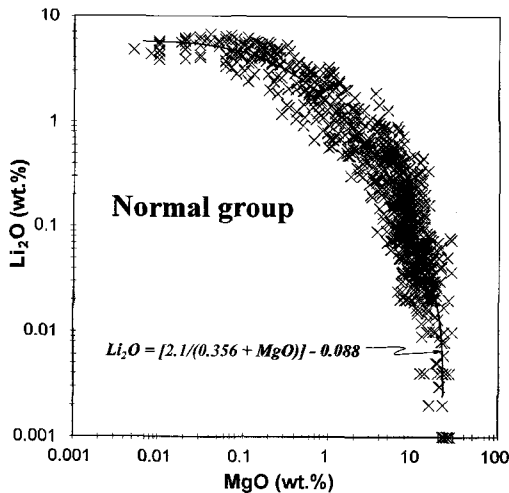


FIG. 1. Variation of Li<sub>2</sub>O with MgO in trioctahedral micas for the 'normal group' with the regression equation.

Curve-fitting leads to the following expression:

$$\text{Li}_2\text{O} = [2.1/(0.356 + \text{MgO})] - 0.088 \quad (n = 870) \\ (R^2 = 0.907) \quad [1]$$

(Range of validity: MgO: 0–23.5 wt.%)

Tischendorf *et al.* (1997) proposed a new graphic presentation of tri- and dioctahedral micas using a diagram with the abscissa (Mg-Li) (*mgli*) and the ordinate (Fe<sub>tot</sub> + Mn + Ti - Al<sup>VI</sup>) (*feal*). Figure 2 shows the positions of the 'normal group' samples in that diagram and demonstrates that for each of the different trioctahedral mica varieties a sufficiently large number of analyses was available for estimating the Li<sub>2</sub>O content by correlation. The *mgli-feal* diagram also indicates the existence of a continuous solid solution between phlogopite and lepidolite. Fe biotite, zinnwaldite and lepidolite show maximum variation in the Fe<sub>tot</sub>/Al<sup>VI</sup> relationship.

(2) The second group of micas (n = 232; 19% of the data) is distinguished by plotting on the concave side of the 'normal' distribution curve (Fig. 3). These micas contain less Li<sub>2</sub>O at a given MgO content (or less MgO at a given Li<sub>2</sub>O

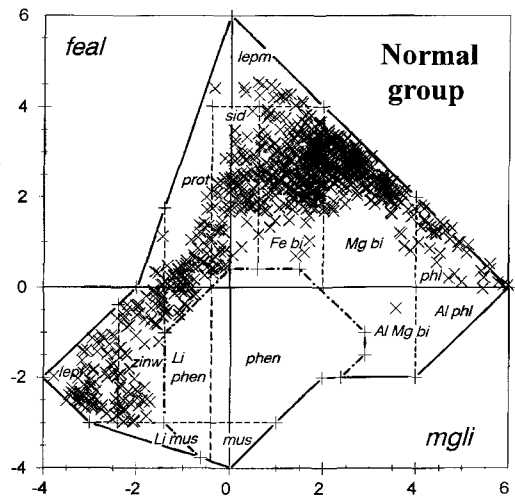


FIG. 2. Position of the 'normal group' micas in the *mgli* vs. *feal* diagram [= Mg - Li vs. Fe<sub>tot</sub> + Mn + Ti - Al<sup>VI</sup> diagram] of Tischendorf *et al.* (1997). Al phl = aluminophlogopite, Al Mg bi = Al-Mg biotite, Fe bi = Fe biotite, Li mus = Li muscovite, Li phen = Li phengite, lepm = lepidomelane, lepl = lepidolite, Mg bi = Mg biotite, mus = muscovite, phen = phengite, phl = phlogopite, prot = protolithionite, sid = siderophyllite, zinw = zinnwaldite.

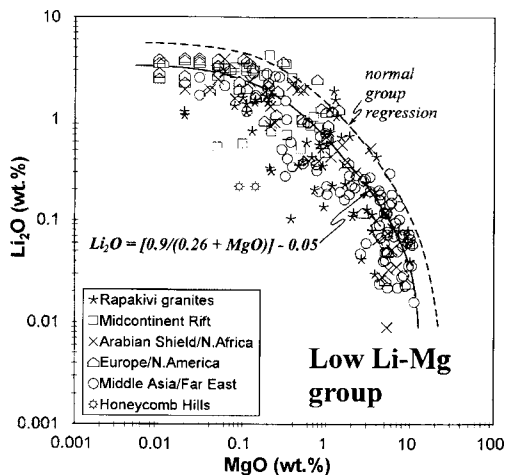


FIG. 3. Variation of  $\text{Li}_2\text{O}$  with  $\text{MgO}$  in trioctahedral micas for the 'low Li-Mg group' with regression equation. The 'normal group' regression is shown for comparison.

content) than the normal group and, therefore, we refer to them as the 'low Mg-Li group'. Because the sum of cations in the octahedral layer is nearly constant in the interval from 5.4 to 6.0, it follows that the lack of Mg and/or Li in this group must be balanced by Fe (with Mn and Ti) and/or  $\text{Al}^{\text{VI}}$ . Micas from the 'low Mg-Li group' occur in the following rock types:

(a) Proterozoic rapakivi granites and related pegmatites and metasomatic rocks from the Fennoscandian and Ukrainian shields (Bur'yanova, 1940; Pavlishin *et al.*, 1968; Rub *et al.*, 1974; Haapala, 1977; Rub and Rub, 1980; Hawthorne and Černý, 1982; Haapala, 1988; Buchinskaya and Nechayev, 1990; Velikoslavinsky, 1994; Rieder *et al.*, 1996);

(b) Mid-Proterozoic granites and related pegmatites from the Midcontinent Rift System of North America (Hawley and Wobus, 1977; Hawthorne and Černý, 1982; Foord *et al.* 1995);

(c) Late Proterozoic (Pan-African) granites of the Arabian Shield and northern Africa (Kabesh *et al.*, 1977, du Bray *et al.*, 1988; El Shestawi *et al.*, 1993; Abdalla *et al.*, 1994; du Bray, 1994; Ahmed-Said *et al.*, 1995);

(d) Paleozoic granites, pegmatites, dolerites, and metasomatic rocks associated with Caledonian to Hercynian orogenic belts of Europe and North America (Rieder, 1970; Hall and Walsh, 1972; Kramer, 1976; Rub *et al.*, 1983;

Pechar and Rykl, 1992; Uhlig, 1992; Taylor, 1992; present authors;

(e) Mesozoic granodiorites, granites, pegmatites, and metasomatic rocks from the territory of the former Soviet Union [Altai, Sayan, Zabaikal, Far East] (Sobachenko *et al.*, 1979; Antipin *et al.*, 1980; Rub and Rub, 1980; Gryazev *et al.*, 1985; Rub and Rub, 1986; Trunilina and Kulagina, 1986; Nedosekin, 1988; Sobachenko *et al.*, 1989);

(f) Cenozoic rhyolites (Honeycomb Hills) from the Basin and Range Province, SW USA (Nash, 1993).

For these micas, the functional relationship between  $\text{Li}_2\text{O}$  and  $\text{MgO}$  is

$$\text{Li}_2\text{O} = [0.9/(0.26 + \text{MgO})] - 0.05 \quad (n = 232) \\ (R^2 = 0.63) [2]$$

(Range of validity:  $\text{MgO}$ : 0–17.7 wt.%)

(3) A third group of analyses ( $n = 104$ ; 9% of the included samples) is characterized by greater  $\text{Li}_2\text{O}$  values at a given  $\text{MgO}$  or greater  $\text{MgO}$  values at a given  $\text{Li}_2\text{O}$  and these, therefore, plot on the convex side of the normal distribution curve (Fig. 4). As a rule, the samples of this 'high Mg-Li group' are characterized by a Fe (+ Mn + Ti) and/or Al-deficiency in comparison to the normal group.

The micas in this group can be assigned to two sub-groups according to aluminosity of their host

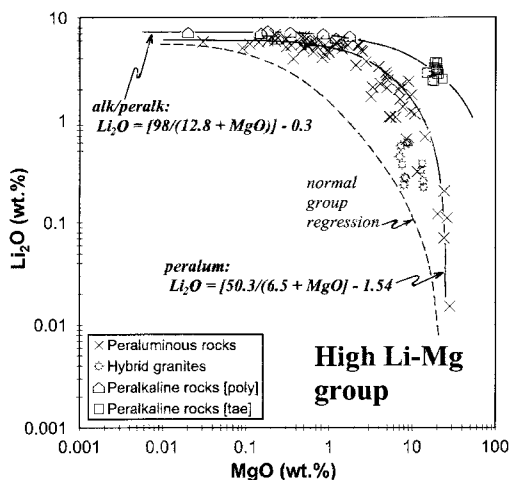


FIG. 4. Variation of  $\text{Li}_2\text{O}$  with  $\text{MgO}$  in trioctahedral micas for the 'high Li-Mg group' with regression equations for the peraluminous and the alkaline-peralkaline sub-groups. The 'normal group' regression is shown for comparison.

rock, namely those from peraluminous rocks and those from rocks of alkaline to peralkaline affiliation:

(a) Peraluminous I- and S-type granites, external (rare-element) pegmatites and their alteration envelopes (Stevens, 1938; Shibata, 1952; Ukai *et al.*, 1956; Němec, 1969; Černý *et al.*, 1970; Černý, 1972; Kovač *et al.*, 1972a; Rinaldi *et al.*, 1972; Khvostova *et al.*, 1973; Moloshag and Teremetskaya, 1975; Lapidés *et al.*, 1977; Neiva, 1981a,b; Hawthorne and Černý, 1982; Kuznetsova and Zagorskiy, 1984; Pomârleanu *et al.*, 1986; Semenov and Shmakín, 1988; Němec, 1990b; Icenhower and London, 1995; Lagache and Quéménéur, 1997; present authors)

(b) Alkaline magmatic and metasomatic rocks which contain 'polyolithionite' [in the sense of Černý and Trueman, 1985] (Stevens, 1938; Vlasov *et al.*, 1959; Vlasov (ed.), 1964; Gamaleya, 1968; Semenov *et al.*, 1969; Raade and Larsen, 1980; Černý and Trueman, 1985) or taeniolite (Vlasov *et al.*, 1959; Vlasov, 1964; Ganzeeva, 1973; Rumyantseva *et al.*, 1984; Cooper *et al.*, 1995).

The relationship between Li<sub>2</sub>O and MgO can be formulated for the micas of the 'peraluminous sub-group' as follows:

$$\text{Li}_2\text{O} = [50.3/(6.5 + \text{MgO})] - 1.54 \quad (n = 84) \\ (R^2 = 0.895) \quad [3]$$

(Range of validity: MgO: 0–26.2 wt.%)

Micas from the 'hybrid granites' in Portugal (n = 12) are, with regard to their Li<sub>2</sub>O/MgO-distribution, transitional between the normal and the high Li-Mg group (see Fig. 4). Thus, they were not used for the calculation of Eqn. 3.

'Polyolithionite' and taeniolite from alkaline-peralkaline rocks are distinguished from micas in peraluminous rocks by still higher Li<sub>2</sub>O contents at a given MgO (see Fig. 4). This 'alkaline-peralkaline subgroup' is characterized by only very few analyses, but can be well described by the following equation:

$$\text{Li}_2\text{O} = [98/(12.8 + \text{MgO})] - 0.3 \quad (n = 22) \\ (R^2 = 0.944) \quad [4]$$

(Range of validity: MgO: 0–29.0 wt.%)

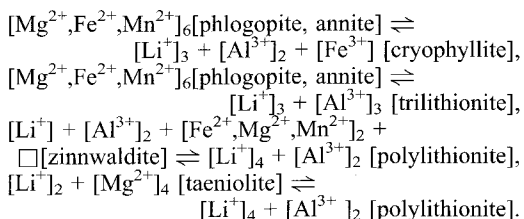
The mica varieties of the low Li-Mg group and of the high Li-Mg group are shown in Fig. 5. Within the latter group the micas of the peraluminous, the peraluminous-hybrid and the alkaline-peralkaline subgroup are designated. In the low Li-Mg group, Fe-rich members predominate in nearly all mica varieties. The lepidomelane

field is intensely occupied. Within the high Li-Mg group, Al-rich mica varieties as well as taeniolites predominate.

## Discussion

### General aspects

The assumption that a fixed relationship exists between Li<sub>2</sub>O and MgO in trioctahedral micas, which can be used for determination of Li<sub>2</sub>O from an MgO analysis, depends upon the reciprocal substitution of Mg and Li. Of course, there are a great number of substitutions among the octahedral cations in mica, so the relation between Mg and Li is coupled. Concerning our objective the following substitutions are essential (see also Černý and Burt, 1984):



In micas from the normal group, substitutions involving phlogopite-trilithionite and zinnwaldite-

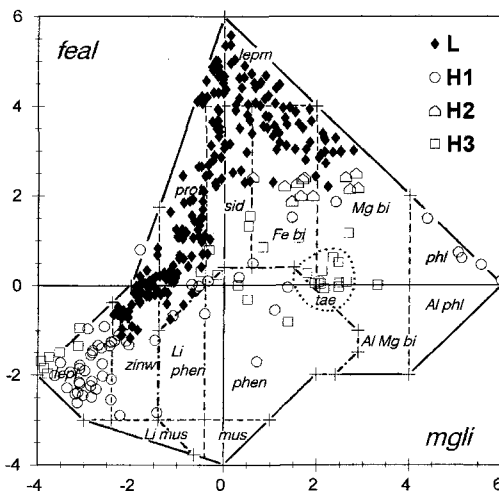


FIG. 5 Position of the 'low Li-Mg' (L) and the 'high Li-Mg group' micas in the *mgli* vs. *feal* diagram. Micas belonging to the latter group are plotted separately for the peraluminous (H1), peraluminous-hybrid (H2) and alkaline-peralkaline sub-groups (H3). See Fig. 2 for explanation of the abbreviations.

polyolithionite are of particular importance. Micas belonging to the annite-cryophyllite join are characteristic of the low Li-Mg group. Those forming the taeniolite-polyolithionite join, however, are characteristic of the high Li-Mg group.

Estimation of the  $\text{Li}_2\text{O}$  contents of very Mg-rich micas using Eqns 1–4 is not a problem because the Li-values are very low and not essential for classification or for petrogenetic or geochemical discussions. In Table 2 we compile  $\text{Li}_2\text{O}$  contents of very Mg-rich micas found in the literature and include two samples from our data bank (see Table 4). Most of the values approach the theoretically expected ones of the order of 0.001 wt.%. However, some  $\text{Li}_2\text{O}$  values are relatively high — this may be due to analytical difficulties or to a geochemically anomalous situation (see below).

The accurate determination of very small MgO contents is more important for the evaluation of  $\text{Li}_2\text{O}$  by correlation than vice versa. The position of some samples with very low MgO and very high  $\text{Li}_2\text{O}$  contents in Fig. 1 suggests that, with decreasing MgO contents (below 0.05 wt.%), the  $\text{Li}_2\text{O}$  contents may also decrease. This could either be connected with a slight increase of Mn in Li-rich micas, (see Tischendorf *et al.*, 1997, Fig. 4c) or inaccurate determination of MgO

when contents are close to the microprobe detection limit. Table 3 lists the MgO contents of the Li-richest micas that have been published, and three analyses of lepidolites from the authors' data bank (see Table 4). Their MgO contents were determined by atomic absorption spectrometry, a method with higher sensitivity than EPMA, but with the disadvantage that bulk separates, and not single grains were used. There are also analyses published for which the value of MgO is set to 0.00 wt.% [i.e. Perrault (1966) for one sample with 7.5 wt.%  $\text{Li}_2\text{O}$ ; Skosyreva and Vlasova (1983) for two samples with 7.37 and 7.67 wt.%  $\text{Li}_2\text{O}$ ; Černý *et al.* (1995) for 14 samples with an interval from 3.94–6.45 wt.%  $\text{Li}_2\text{O}$ ; Novák and Povondra (1995) for two samples with 5.60 and 6.06 wt.%  $\text{Li}_2\text{O}$ ].

#### *Li<sub>2</sub>O-MgO relations between trioctahedral micas and their host rocks*

In our suite of samples, normal-group micas occur in common I-type or S-type rocks (or mixed types), which are derived by melting processes in connection with subduction and collision processes. Micas from other rock-types, such as regional metamorphic rocks, are subordinate in our database because of the scarcity of published Li-values.

TABLE 2.  $\text{Li}_2\text{O}$  contents in very Mg-rich trioctahedral micas (in order of decreasing MgO). Stars indicate micas belonging to the 'high Li-Mg group'

Author	MgO (wt.%)	$\text{Li}_2\text{O}$ (wt.%)
Phlogopite, theoretical	28.97	0.000
Hazen and Burnham (1973), Table 2, Franklin	28.60	0.01
Pomârleanu <i>et al.</i> (1986), no.1 ★	28.46	0.015
Malyshonok (1989), Table 1, no. 17	27.90	0.075
Malyshonok (1989), Table 1, no. 18	27.52	0.037
Bagdasarov <i>et al.</i> (1985), Table 1, no. 9	27.49	0.001
Bagdasarov <i>et al.</i> (1985), Table 1, no. 2	27.22	0.001
Bagdasarov <i>et al.</i> (1985), Table 1, no. 12	26.68	0.004
Malyshonok (1989), Table 1, no. 15	26.66	0.078
This work, Table 4, no. SK1 ★	26.30	0.11
Rimsaite (1970), Table, no. 5c	25.69	0.02
Bagdasarov <i>et al.</i> (1985), Table 1, no. 25	25.61	0.001
Malyshonok (1989), Table 1, no. 13	25.15	0.001
Bagdasarov <i>et al.</i> (1985), Table 1, no. 21	24.81	0.051
Bagdasarov <i>et al.</i> (1985), Table 1, no. 11	24.41	0.004
This work, Table 4, no. 189 ★	24.40	0.20
Černý (1972), no. 1 ★	24.25	0.07
Grew <i>et al.</i> (1990), Table 8, no. 10	23.44	0.001

TABLE 3. MgO contents in very Li-rich trioctahedral micas (in order of decreasing Li<sub>2</sub>O). Stars indicate micas belonging to the high Li-Mg group. P = polyolithionite, sum alk > sum Al; L = lepidolite, sum alk < sum Al

Author	Li <sub>2</sub> O (wt.%)	MgO (wt.%)
Polyolithionite, theoretical	7.28–7.65	0.000
Gamaleya (1968), no. 1 ★ [P]	7.45	0.18
Stevens (1938), Table 1, no. 17 ★ [P]	7.26	0.34
Semenov <i>et al.</i> (1969) [P]	7.18	0.02
Raade and Larsen (1980), no. 1 ★ [P]	7.07	0.15
Černý and Trueman (1985), no. T ★ [P]	6.97	0.84
Stevens (1938), Table 1, no. 16 ★ [L]	6.84	0.22
Uhlig (1992), Table 5.1, No. Z 11 [L]	6.69	0.04
Černý and Trueman (1985), no. S ★ [P]	6.62	1.75
Ukai <i>et al.</i> (1956), Table 3, no. 7 ★ [L]	6.55	0.46
Lapides <i>et al.</i> (1977), Table 2, No. 62 ★ [L]	6.54	0.50
Franzini and Sartori (1969), Table [L]	6.51	0.07
Chaudhry and Howie (1973), Table 1, no. M.2 [L]	6.40	0.06
Monier <i>et al.</i> (1987), Table 1, no. 2 [L]	6.33	0.03
Chaudhry and Howie (1973), Table 1, no. MO.1 [L]	6.20	0.11
Stevens (1938), Table 1, no. 15 ★ [L]	6.18	0.30
This work, Table 4, no.1001 [L]	6.15	0.020
This work, Table 4, no.1022 [L]	6.15	0.029
Chaudhry and Howie (1973), Table 1, no. MN.21 [L]	6.00	0.06
Chaudhry and Howie (1973), Table 1, no. MKC [L]	6.00	0.08
Hawthorne and Černý (1982), Table 8, no. 2 ★ [L]	5.95	0.22
Hawthorne and Černý (1982), Table 8, no. 4 ★ [L]	5.95	0.18
Němec (1990a), Table 3, no. P IV [L]	5.92	0.03
Černý <i>et al.</i> (1970), no. 41 ★ [L]	5.91	0.63
This work, Table 4, no.1004 [L]	5.90	0.039

The low Li-Mg group is primarily composed of trioctahedral micas that originate from metaluminous to mildly peraluminous granitoid rocks and their derivatives (pegmatites, aplites) which show A-type affiliation. They are either truly anorogenic or post-orogenic. These micas show a remarkable scatter in Li<sub>2</sub>O-MgO space with some, particularly those from post-collisional A-type granites, approaching the relations observed in the normal group. Several factors, such as source rock composition (a F-rich and water-poor protolith) as well as *PTX*-conditions of anatectic melting and crystallization (low oxygen fugacity), have been invoked to explain the typically strongly ferruginous composition of these micas which belong to the annite-siderophyllite series (e.g. Kinnaird *et al.*, 1992; Abdel-Rahman, 1994; Shannon *et al.*, 1997; Smith *et al.*, 1997).

However, neither enhanced fluorine (which is equally high in evolved S-type granites) nor low oxygen fugacities (the A-type rocks considered here cover a wide range in  $f_{O_2}$ , from below QFM

to above HM; e.g. Anderson and Bender, 1989; Rieder *et al.*, 1996) appear crucial in shifting the mica composition in terms of Li<sub>2</sub>O vs. MgO away from the curve which characterizes the normal group. The extraordinary position of the Honeycomb Hills annitic biotites (see Fig. 3), even with respect to other A-type micas, may be the result of sub-solidus re-equilibration at high  $f_{O_2}$  as discussed by Icenhower and London (1997).

Micas from the high Li-Mg group (Fig. 4) come from rocks of very different genesis and chemical composition. Host rocks with peraluminous affinity are characterized by extensive interaction between the granitic melt or external pegmatitic liquids with the country rock. The Li and Mg in these micas may have originated from different sources, the lithium coming from granitic-pegmatitic melt (mother-rock component), and Mg from the rocks adjacent to the place of formation (wall-rock component). The shift towards higher MgO (at a given Li<sub>2</sub>O) is greatest for micas in contact with the Mg-richest wall



TABLE 4. Chemical analyses of mica separates

Region Locality Rock type Sample	Erzgebirge Pöhla skarn SK 1	Erzgebirge Zöblitz serpentinite 189	Granulitgebirge Wolkenstein pegmatite 1004	Granulitgebirge Wolkenstein pegmatite 1022	Mocambique Alto Ligonha pegmatite 1001
SiO <sub>2</sub> (wt.%)	43.3	43.3	50.7	51.6	51.1
TiO <sub>2</sub>	0.12	0.26	0.03	0.01	0.01
Al <sub>2</sub> O <sub>3</sub>	11.2	12.3	23.2	23.5	22.6
Fe <sub>2</sub> O <sub>3</sub>	0.33	1.00	0.08	0.29	0.17
FeO	3.46	5.38	0.05	0.17	0.07
MnO	0.06	0.08	0.22	0.08	1.23
MgO	26.3	24.4	0.039	0.029	0.020
CaO	0.16	0.02	0.03	0.01	0.05
Li <sub>2</sub> O	0.11	0.20	5.90	6.15	6.15
Na <sub>2</sub> O	0.07	0.27	0.29	0.27	0.25
K <sub>2</sub> O	8.3	8.3	9.95	10.3	9.6
Rb <sub>2</sub> O	0.044	0.110	1.461	1.016	1.156
Cs <sub>2</sub> O	0.118	0.024	1.105	1.083	0.245
F	2.3	0.44	5.6	5.5	6.2
Cations normalized to 22 oxygens					
Si	6.149	6.078	6.883	6.876	6.909
Al(IV)	1.851	1.922	1.117	1.124	1.091
Ti	0.013	0.027	0.003	0.001	0.001
Al(VI)	0.029	0.107	2.596	2.567	2.512
Fe <sup>3+</sup>	0.035	0.106	0.008	0.029	0.017
Fe <sup>2+</sup>	0.411	0.631	0.006	0.019	0.008
Mn	0.007	0.010	0.025	0.009	0.141
Mg	5.565	5.103	0.006	0.004	0.004
Li	0.063	0.113	3.221	3.296	3.344
Sum [Y]	6.122	6.096	5.865	5.925	6.027
Ca	0.024	0.002	0.004	0.002	0.007
Na	0.018	0.072	0.075	0.070	0.066
K	1.503	1.486	1.723	1.751	1.656
Rb	0.004	0.010	0.128	0.087	0.100
Cs	0.007	0.001	0.064	0.062	0.014
Sum [X]	1.557	1.572	1.994	1.971	1.843
F	1.033	0.195	2.404	2.318	2.651
<i>mgli</i>	5.502	4.990	-3.215	-3.291	-3.340
<i>feal</i>	0.437	0.667	-2.554	-2.510	-2.344
Variety	phlogopite	phlogopite	lepidolite	lepidolite	lepidolite

*mgli* = Mg - Li (octahedral), *feal* = Fe(tot) + Mn + Ti - Al(VI)(octahedral)

Analytical conditions are given in Tischendorf *et al.* (1969). All samples were recently re-analysed for Si, Al, Mg, Ca, Na, Rb and Cs by XRF, AAS, ICP-AES and INAA, and modified accordingly.

rock. In our suite of samples this comprises serpentinites, amphibolites, keratophyres, gabbros. As a result, we observe Li-poor as well as Li-rich phlogopites (e.g. Pomârleanu *et al.*, 1986), abnormally Mg-rich zinnwaldites (e.g.

Semenov and Shmakin, 1988; Lagache and Quéméneur, 1997), and even the extraordinary micas (high-Li-Rb-Cs-Mg 'siderophyllites') formed at the exocontact of the Tanco pegmatite (e.g. Icenhower and London, 1995).

We interpret the unique position of the micas from the Portuguese hybrid granites (Neiva, 1981*a,b*) at the concave side of the 'normal group' regression hyperbola (Fig. 4) as a consequence of crystallization under disequilibrium conditions. Obviously, these micas are characterized by their bigenetic origin which assumes derivation of lithium from the granites and mobilization of magnesium from the wall-rock.

Concerning the group of Li-richest micas, polyolithionite (in the sense of Černý and Trueman, 1985) is the characteristic mica in 'peralkaline igneous rocks'. The highly variable  $\text{Al}_2\text{O}_3$  content in lepidolites (10–32 wt.%) can be regarded as sensitive to the activity of alumina in the parent medium (i.e. Němec and Povondra, 1993). Thus, micas with a polyolithionite composition reflect the peralkaline environment and trillithionitic micas are typical of peraluminous host rocks. According to their composition and environment of formation, the taeniolites also belong to the group with peralkaline affinity (e.g. Fig. 4). This has called attention to the possible existence of a polyolithionite-taeniolite solid solution series with intermediate compositions (Černý and Trueman, 1985). The Li and Mg in these micas can be mono- as well as bigenetic. It is, however, likely that a major part of the Mg is derived from the wall rock.

#### The problem of trioctahedral micas low in $R^{\text{VI}}$

The  $\text{Li}_2\text{O}/\text{MgO}$  correlation changes with decreasing  $R^{\text{VI}}$  from trioctahedral lepidolite, zinnwaldite and protolithionite to the dioctahedral Li muscovite/Li phengite. The  $\text{Li}_2\text{O}/\text{MgO}$  relations in both the transitional and dioctahedral micas resemble those characteristic of the low Li-Mg group. To avoid large errors in estimating  $\text{Li}_2\text{O}$  by correlation with MgO, we recommend including only micas with  $R^{\text{VI}} > 5.0$ . In order to give an impression about the  $\text{Li}_2\text{O}/\text{MgO}$  distribution in dioctahedral micas, we refer the reader to Fig. 6.

#### Calculation of errors

In Table 5 we present an evaluation of the average error when calculating  $\text{Li}_2\text{O}$  as a function of MgO separately for the four mica groups discussed and for individual MgO intervals. Note that the calculated  $\text{Li}_2\text{O}$  mean corresponds very well with the  $\text{Li}_2\text{O}$  mean from the analysed samples. With this in mind, the standard deviation

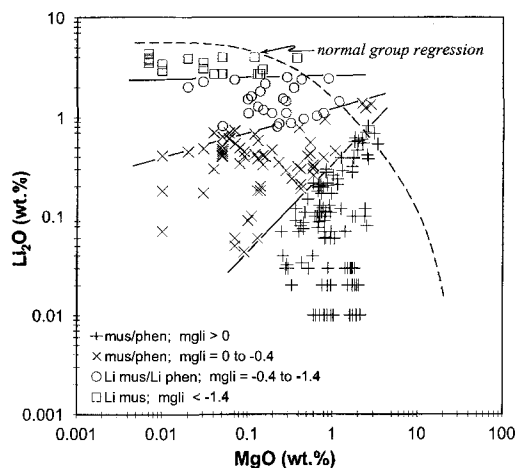


Fig. 6. Variation of  $\text{Li}_2\text{O}$  with MgO in different varieties of dioctahedral micas. For database used see Tischendorf *et al.* (1997, Appendix). See Fig. 2 for explanation of the abbreviations.

from the analysed samples can be used as the average error for the calculated  $\text{Li}_2\text{O}$  value. As expected, relative errors from the estimation at lower  $\text{Li}_2\text{O}$  contents are larger (35–92%) than for the higher ones (7–42%). Due to the large scatter of values for micas in the low Li-Mg group, the error here is greatest. This evidently reflects greater genetic variety among the host rocks. To minimize the errors, workers who want to get an impression of the probable  $\text{Li}_2\text{O}$  contents in very annite-rich micas of the low Li-Mg group ( $\text{Fe}_{\text{tot}} > 4$ ) should prefer the equation  $\text{Li}_2\text{O} = [0.25/(0.25 + \text{MgO})] - 0.03$ . The error in the other groups has the same order of magnitude as for most of the direct analytical methods for  $\text{Li}_2\text{O}$ .

#### Conclusions

Advantages in estimating  $\text{Li}_2\text{O}$  using the relationship with MgO include: (a) the applicability to the whole existing compositional range of Li and Mg (and not, as in the case of correlation with  $\text{SiO}_2$ , for only a part of the range), and (b) the reversed behaviour of both the elements across the entire compositional range. We point out that, due to analytical reasons and the existing chemical variability, especially in isomorphous mineral groups, estimation of  $\text{Li}_2\text{O}$  in micas by direct correlation with any element, including MgO, is only an approximation. Nevertheless, evaluation of the absolute and relative errors in  $\text{Li}_2\text{O}$

TABLE 5. Average error from the estimation of Li<sub>2</sub>O as function of MgO in trioctahedral micas

Group	MgO interval (wt.%) of analysed samples	n	Li <sub>2</sub> O interval and mean (wt.%)	Li <sub>2</sub> O mean (wt.%) of analysed samples	Li <sub>2</sub> O standard deviation of analysed samples	Relative error (%)
normal	0.03-0.1	39	5.35-4.52/4.90	4.92	1.02	21
	0.3-0.8	50	3.11-1.73/2.23	2.30	0.97	42
	1.0-3.0	96	1.46-0.54/0.80	0.99	0.63	63
	8.0-10.0	127	0.16-0.11/0.14	0.13	0.084	65
	16.0-22.0	35	0.04-0.006/0.02	0.025	0.020	80
low Li-Mg	0.03-0.1	28	3.05-2.45/2.72	2.63	0.95	36
	0.3-0.8	25	1.56-0.80/1.06	1.05	0.97	92
	1.0-3.0	33	0.66-0.23/0.35	0.54	0.46	85
	6.0-10.0	10	0.094-0.038/0.059	0.061	0.020	33
	0.03-0.6	16	6.16-5.54/5.87	5.68	1.10	19
high Li-Mg peralum	1.0-3.0	16	5.17-3.75/4.38	4.80	0.66	14
	7.0-10.0	5	2.19-1.51/1.81	1.47	0.52	35
high Li-Mg peralk	20.0-26.0	4	0.36-0.01/0.17	0.12	0.047	39
	0.2-2.0	8	7.24-6.32/6.85	6.85	0.48	7
	14.0-24.0	14	3.36-2.36/2.78	3.03	0.40	13

TABLE 6. Genetic grouping of trioctahedral micas according to their Li-Mg distribution

Group	Characteristic mica varieties	Host-rock, environment and conditions of mica formation	Frequency of analysed samples (in %)	Li <sub>2</sub> O estimated by
Normal	Phlogopite-biotite-siderophyllite-protolithionite-zinnwaldite-lepidolite series	I- and S-type, calc-alkaline to peraluminous magmatic rocks	72	Eqn. 1
Low Li-Mg	Fe-rich varieties, esp. annite and lepidomelane	A-type, metaluminous to weakly peraluminous magmatic rocks	19	Eqn. 2
High Li-Mg peralum	Al-rich varieties; for Li-rich conditions esp. 'lepidolite' ( $\Sigma Al > K + Na + Rb + Cs$ )	I-S-type peraluminous magmatic rocks and Mg-enriched environment of crystallization	7	Eqn. 3
High Li-Mg peralk	Relative Al-poor varieties; for Li-rich conditions esp. 'polyolithionite' ( $K + Na + Rb + Cs > \Sigma Al$ ); taeniolite	Alkaline to peralkaline magmatic rocks and Li-Mg-enriched environment of crystallization	2	Eqn. 4

estimation for different MgO intervals shows that Li<sub>2</sub>O amounts can be determined quite satisfactorily. One important prerequisite for the use of this relationship between Li<sub>2</sub>O and MgO is taking into account the genetic group to which the micas belong. This, however, should be resolved readily using further geological, geochemical, mineralogical and isotopic criteria which properly characterize the origin of the host-rocks of the mica and the environment and conditions of its formation. It may happen that transitions exist from one group to the other, as in the example of the micas from the Portuguese hybrid granites. Without doubt, this limits the application of the Eqns 1–4. This limitation is acceptable because there is no better alternative for evaluation of Li<sub>2</sub>O in Mg-rich, Li-poor micas. The Mg/Li relations in micas with R<sup>VI</sup> between 5.0 and 4.4 are similar to those of dioctahedral micas and, therefore, cannot be described reliably by any of the equations proposed in this paper.

The use of the Eqns 1–4 is sufficiently accurate for felsic to intermediate magmatic rocks and their high-temperature derivatives, taking into consideration the huge number of analyses included in this evaluation. A summary of the results for the distribution of Li<sub>2</sub>O as a function of MgO content in trioctahedral micas with respect to their geological position and the environment in which they formed is presented in Table 6. It remains to be seen whether or not these relationships are also valid for rocks from other geological environments.

For the determination of Li<sub>2</sub>O in Li-rich, Mg-poor trioctahedral micas, equations with Li<sub>2</sub>O as a function of SiO<sub>2</sub>, as suggested by Tindle and Webb (1990) and Tischendorf *et al.* (1997), remain reasonable options, and it is up to the reader to use one or the other equation. However, one important result of this work is that the calculation of Li<sub>2</sub>O as a function of SiO<sub>2</sub> should be restricted to micas with <3 wt.% MgO and this equation is only viable for micas with >34 wt.% SiO<sub>2</sub>. For all other trioctahedral micas we recommend Eqns 1–4 in this paper. From a practical point of view, it seems reasonable to use Eqns 1–3 for the estimation of Li<sub>2</sub>O in Mg-Fe micas (phlogopite, Mg biotite, Fe biotite, siderophyllite, lepidomelane) and possibly associated Li-Fe micas (Li-bearing siderophyllite and lepidomelane, protolithionite). Equations with Li<sub>2</sub>O plotted as a function of SiO<sub>2</sub> can be used for Li-Al micas (lepidolite, zinnwaldite) and their associated Li-Fe micas (protolithionite, Li-

bearing siderophyllite). Equations 3 and 4 converge for Li<sub>2</sub>O at MgO contents below 0.5 wt.%. Li<sub>2</sub>O contents of micas chemically similar to taeniolite can only be estimated using Eqn. 4.

Finally, we would discourage the use of different regression equations when studying the compositional evolution of micas in multi-stage cogenetic plutons or intrusive series. This could result in artificial breaks in the Li<sub>2</sub>O variations. In such cases, the most primitive trioctahedral mica (in terms of Mg concentration) determines which equation should be used to estimate Li<sub>2</sub>O for the entire series.

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