

Li, Be, B, and Sr in margarite and paragonite from Antarctica

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ABSTRACT

Peraluminous margarite and paragonite of retrograde origin in a staurolite- and corundum-bearing talc-tourmaline-phlogopite-chlorite schist from Mount Bernstein, northern Victoria Land, Antarctica, contain 0.17 and 0.18 wt% Li_2O , 0.05 and 0.004% BeO , 0.02 and 0.31% BaO , and 0.26 and 1.3% SrO , respectively, by ion-microprobe analysis (estimated accuracy: $\pm 20\%$). In addition, only 0.01% F and 0.02–0.03% B_2O_3 and no Rb were found with the ion microprobe. This is the first report of significant amounts of Li and Sr in paragonite. The unusual compositions of the paragonite and margarite are attributed to their ability to incorporate Li released from breakdown of lithian staurolite and Sr made available during a retrograde metamorphic stage in a silica-undersaturated rock. Their low B_2O_3 contents are attributed to the inability of the mica structure to accommodate the small ^{10}B ion in the tetrahedral sheet even in a B-enriched host rock.

INTRODUCTION

Since the advent of the electron microprobe, the light-element contents ($Z < 9$) of rock-forming minerals have been largely neglected in metamorphic petrology. For the dioctahedral micas, this could be a serious oversight, because Li can play an important role in these micas. Muscovite and margarite are known to incorporate significant amounts of Li in the appropriate environment, as reviewed by Černý and Burt (1984) and Guggenheim (1984). In this communication, we report electron- and ion-microprobe analyses of a margarite and a paragonite containing significant Li, Sr, and Be. As far as we are aware, this is the first report of paragonite containing more than traces of Li and one of the few reports of a mica containing more than traces of Sr. We also discuss possible constraints on the incorporation of B in micas.

We studied lithian and strontian paragonite and margarite occurring as a few flakes in a staurolite- and corundum-bearing talc + phlogopite + chlorite + tourmaline schist from Mount Bernstein ($71^{\circ}37'\text{S}$, $163^{\circ}07'\text{E}$) in northern Victoria Land, Antarctica (Grew and Sandiford, 1984). Margarite forms flakes 0.05–0.25 mm across in a sericitic aggregate surrounded by tourmaline. Pumpellyite, chlorite, and relict staurolite are also present in the aggregate. Paragonite occurs as two contiguous flakes 0.2 mm across surrounded by tourmaline and 0.1 mm distant from corundum, which is partially enveloped in a sericitic aggregate. The analyzed paragonite and margarite are only 2 mm apart, but are separated by tourmaline and thus may not have equilibrated with one another. The margarite and paragonite formed from the breakdown of staurolite

and corundum at 300–370°C and 3–5 kbar during the late (retrograde) stage in the metamorphic cycle (Grew and Sandiford, 1984, 1985).

MICROPROBE ANALYSIS

Electron-microprobe analyses of 10 major elements in margarite (several flakes, analysis A in Table 2 of Grew and Sandiford, 1984), paragonite (two flakes), and spodumene were obtained at the Ruhr-Universität Bochum, and of 13 major elements in biotite at the University of Melbourne and University of California, Los Angeles (Table 1; details of the analytical methods given in Grew and Sandiford, 1984, and Grew, 1980).

Secondary ion spectra in the mass range 1 to 150 (H through Nd) from one flake each of the margarite and paragonite as well as from the four standards listed in Table 2, were scanned and recorded with an ARL ion-microprobe mass analyzer (IMMA) at the Aerospace Corporation in Los Angeles (method of Grew and Hinthorne, 1983). After each mass scan, quantitative data were collected by ion counting. Each standard was analyzed quantitatively one time during the course of the day-long session.

Raw IMMA count ratios, except for Sr, were corrected with mineral standards of known composition (Table 2): spodumene for $^7\text{Li}/^{28}\text{Si}$, biotite for $^{138}\text{Ba}/^{28}\text{Si}$ and $^{19}\text{F}/^{28}\text{Si}$, grandidierite for $^{11}\text{B}/^{28}\text{Si}$, and surinamite for $^9\text{Be}/^{28}\text{Si}$ (working-curve approach of Grew and Hinthorne, 1983). Positively charged secondary ions were measured in all cases. In applying standards of differing crystal structure and composition from the unknowns, we have assumed that the matrix has a relatively minor effect on the count ratios; several figures in Andersen and Hinthorne (1973) show that this assumption is justifiable in a wide variety of silicates for data from the IMMA. The grandidierite B/Si and surinamite Be/Si atom ratios were assumed to be 1 and 0.309, respectively, on the basis of crystal-structure refinements (Stephenson and Moore, 1968; Moore and Araki, 1983). The Li/Si ratio in the

Table 1. Compositions of spodumene and biotite standards and of paragonite and margarite in sample no. 4017E

	Spodumene MS2975A	Biotite* 2045G	Paragonite 4017E	Margarite** 4017E
Electron-microprobe analysis (weight percent)				
SiO ₂	64.17	38.34	41.73	30.12
TiO ₂	<0.01	4.86	0.02	0.24
Al ₂ O ₃	27.32	13.25	41.24	49.46
Cr ₂ O ₃	≤0.02	0.12	≤0.02	0.10
FeO	0.02	6.14	0.27	0.33
MnO	0.07	0.03	<0.01	<0.01
MgO	<0.01	20.96	0.27	0.69
CaO	≤0.01	b.d.	1.00	10.95
Na ₂ O	0.15	0.12	6.66	1.66
K ₂ O	≤0.01	10.24	0.62	0.03
BaO	—	0.91	—	—
F	—	3.41	—	—
Cl	—	0.02	—	—
Ion-microprobe analyses (weight percent)				
Li ₂ O	(7.89)	0.03	0.18	0.17
BeO	b.d.	b.d.	0.007	0.05
B ₂ O ₃	b.d.	0.01	0.03†	0.02
SrO	b.d.	trace	1.3	0.26
BaO	b.d.	(0.91)‡	0.31	0.02
F	b.d.	(3.41)‡	0.01	0.01
Total	99.62	98.44	93.65	94.11
—O=F,Cl		1.63		
Total		96.81		
Cations				
Si	1.998	5.584	5.521	4.053
Al	1.003	2.274	2.470	3.927
B	0.000	0.004	0.007	0.004
Be	0.000	0.000	0.002	0.016
Total	3.001	7.862	8.000	8.000
Al	—	0.000	3.961	3.917
Ti	0.000	0.532	0.002	0.024
Cr	0.000	0.014	0.000	0.011
Fe	0.001	0.748	0.030	0.037
Mn	0.002	0.004	0.000	0.000
Mg	0.000	4.550	0.053	0.138
Li	0.989	0.018	0.094	0.092
Total	—	5.866	4.140	4.219
Ca	0.000	0.000	0.142	1.579
Sr	0.000	—	0.099	0.021
Ba	0.000	0.052	0.016	0.001
Na	0.009	0.034	1.709	0.433
K	0.000	1.902	0.105	0.005
Total	1.001	1.988	2.071	2.039
Total cations	4.002	15.716	14.211	14.258
F	0.000	1.573	0.004	0.003
Cl	—	0.004	—	—
Oxygens	5.5§	22	22	22

Note: All Fe as FeO. Dash—not analyzed or not calculated. In parentheses—calculated or assumed values in the standards. b.d.—below detection.

* Electron-microprobe analysis done at the University of Melbourne and UCLA, remainder at the Ruhr-Universität Bochum.

** Electron-microprobe data from Grew and Sandiford (1984, Table 2, margarite A).

† Possible contribution from tourmaline (see text).

‡ Not included in the analytical total.

§ For Al and Si only.

Table 2. IMMA count ratios for mineral standards

Isotope ratio	Standard mineral	IMMA Count ratio	Ideal or analyzed ratio	Measured sensitivity ratio	Predicted sensitivity ratio
¹¹ B/ ²⁸ Si	Grandidierite*	0.344	1.00	0.344	0.355
⁹ Be/ ²⁸ Si	Surinamite**	0.286	0.309	0.926	1.36
⁷ Li/ ²⁸ Si	Spodumene†	1.339	0.495	2.704	2.98
¹³⁸ Ba/ ²⁸ Si	Biotite‡	0.0295	0.00928	3.179	3.36
¹⁹ F/ ²⁸ Si	Biotite‡	0.0146	0.282	0.0518	0.017§

* As inclusions in kornerupine from Port Shepstone, South Africa. Sample BM1940,39, British Museum (Natural History). See de Villiers (1940).

** From pegmatite, Enderby Land, Antarctica. Sample 2292B, Grew (1981).

† From pegmatite, Tin Mountain mine, near Custer, South Dakota. Sample MS2975A of the museum in the Department of Earth and Space Sciences, University of California, Los Angeles.

‡ From a sapphire-garnet rock, Sample 2045G, Gage Ridge, Enderby Land, Antarctica (see Grew, 1983).

§ Measured value (see text).

For Sr, the measured ⁸⁸Sr/²⁸Si ion ratio was divided by the relative sensitivity factor of 3.7 (computed from the local thermal equilibrium (LTE) model of Andersen and Hinthorne, 1973). This correction appears reasonable, given the close correspondence of the measured and theoretical Ba sensitivity factors (Table 2), and the nearly identical behavior of Ba and Sr during ion bombardment.

Of the elements sought in this study, namely Li, Be, B, F, Sr, and Ba, only Be analysis is complicated by interference at the concentrations that we found of these elements. Triply charged ²⁷Al interferes with the ⁹Be signal at mass 9. In order to correct for the contribution of triply charged ²⁷Al, we estimated the ratio of triply charged ²⁷Al at mass 9 to singly charged ²⁷Al at mass 27 to be 5×10^{-5} from counts obtained on minerals containing negligible Be, such as sillimanite and andalusite in Be-poor rocks and grandidierite. We calculated the contribution of ⁹Be to the counts at mass 9 in Be-bearing minerals by deducting 5×10^{-5} times the counts at mass 27.

In contrast to most positively charged secondary ions, F has such a high first ionization potential that only a small fraction of F ions are emitted as "thermal" ions, and thus, F sensitivity cannot be predicted by the LTE model. The relative sensitivity (F/Si) value of 0.017 listed in Table 2 for F was determined from a working curve relating F/Si ratios obtained by classical chemical techniques with those measured on the IMMA (Hinthorne and Andersen, 1975).

In sum, the weight percent of a given oxide was calculated by the following equation, for example, for Li₂O:

$$2\text{Li}_2\text{O} = \text{SiO}_2 \times \frac{29.8814}{60.0843} \times \frac{\text{Li}}{\text{Si}} \times \frac{1}{2.704}$$

and weight percent F, by

$$\text{F} = \text{SiO}_2 \times \frac{18.9984}{60.0843} \times \frac{\text{F}}{\text{Si}} \times \frac{1}{0.0518},$$

spodumene standard was calculated from the electron-microprobe analysis assuming a formula (Li,Na,Mn,Fe)AlSi₂O₆ (Table 1). The Ba/Si and F/Si ratios in the biotite standard were calculated directly from the electron-microprobe analysis (Table 1).

where SiO₂ = weight percent from electron-microprobe analysis, Li/Si and F/Si = count ratios corrected for background, and the remaining numbers are the atomic weights and the measured sensitivity ratios from Table 2. For margarite, the following values were measured (in counts per second corrected for a background of 3 c.p.s.): Li/Si = 12 438/201 601 and 12 352/200 538

(repeated measurements) and $F/Si = 9/201\ 601$, which were entered into the above equations to obtain the values listed in Table 1.

Estimated precision of the IMMA data (based on our experience using the IMMA for similar analyses on several occasions) is about $\pm 20\%$ of the given value for Ba (in paragonite), Li, and Sr, for which the observed intensity exceeded 1000 counts (counting time = 10 s). Precision is poorer than $\pm 20\%$ for intensities below this, as for Ba (in margarite), Be, and B. For example, Li_2O values on biotite 2045G from two different sessions on the IMMA differed by less than 3%, whereas B_2O_3 values differed by 30%. Values given in Table 1 for biotite 2045G are a weighted average of two IMMA sessions for Li and three sessions for B; weighting was based on the observed ion intensity for each session, assuming that the higher count rates provided better results. For comparison, we note that Grew and Hinthorne (1983) reported spreads of 15 to 20% of the measured values for B_2O_3 in two sillimanites (count rates of at least 100 c.p.s.), and 5% for B_2O_3 in two kornorupines (count rates of at least 1000 c.p.s.).

The good agreement between the measured and predicted B/Si sensitivity ratios (Table 2) confirms Hinthorne and Ribbe's (1974) use of the predicted ratios to measure the B contents of chondrodite. We note that Guggenheim et al. (1983, p. 253) also used spodumene as a standard for measuring Li in the Li-Be mica bityite and reported that their ion-microprobe data "confirm" earlier wet-chemical and spectroscopic data.

ION-MICROPROBE RESULTS

Both the margarite and paragonite contain significant Li and Sr and measurable amounts of Be (Table 1). A small amount of Ba is present in the paragonite, but no Rb was detected in the scans of either mica (detectability limit for Rb is < 50 ppm in the qualitative mass scans). The margarite and paragonite in 4017E do not contain significant F or B (Table 1). The low F contents are not unexpected. F was not detected in tourmaline, which was analyzed at one spot in this sample with the IMMA during a previous session (F detection limit is about 0.05 wt%). On the other hand, the low B contents of the margarite and paragonite suggest that they incorporate little B in a B-rich environment (cf. Harder, 1959, who reported 0.03–0.07 wt% B_2O_3 in paragonite). The 0.03% B_2O_3 reported for paragonite in Table 1 may in part be due to a hidden tourmaline impurity or to interference from the surrounding tourmaline, for no B was detected in the element scan.

DISCUSSION

A discussion of the light-element and Sr contents of micas involves two considerations: (1) the effect of these elements on the major-element compositions of the micas and (2) the crystallographic and environmental constraints on incorporation of these elements into the mica structure.

Li, Be, and Sr in margarite and paragonite

A characteristic feature of margarite compositions is an excess of ^{IV}Al relative to an ideal stoichiometric mixture of the major endmember compositions of margarite, paragonite, and muscovite (Frey et al., 1982, Fig. 6). In his review of paragonite compositions, Guidotti (1984, Fig.

4) found that ^{IV}Al contents rarely exceeded the ideal value of 2 atoms per ($20O + 4OH$). The paragonite in sample 4017E is unique in its substantial excess of ^{IV}Al relative to the amount of Ca.

In order to clarify possible substitutions leading to the peraluminous margarite and paragonite compositions, we have compiled in Table 3 a set of mica endmember components that appear most appropriate for this task. In addition, we give in Table 3 the exchange operators that will produce these endmember components when applied to a starting composition of, in this case, paragonite (method of Thompson, 1982). The only component not involving Li, Be, Sr, and Ba is a hypothetical endmember $Na_2MgAl_4(Si_4Al_4)O_{20}(OH)_4$ in which five of the six octahedral sites per formula unit are occupied. Kienast and Velde (1968) and Chopin (1977) proposed this component to explain compositional variations in paragonite and margarite, respectively. However, not all the excess ^{IV}Al in our micas can be explained through this substitution. If Li, Be, Sr, and Ba are assumed to be incorporated as ephesite, bityite, and the hypothetical Sr-Al and Ba-Al endmember micas, all but about 0.1 and 0.2 atoms per formula unit of the excess Al are accounted for in paragonite and margarite, respectively (Table 3). Further, the calculated octahedral occupancies of 4.140 and 4.219 for paragonite and margarite exceed the ideal value of 4 for dioctahedral mica by the amount of Li and Mg present (Table 1). We have selected bityite, $Ca_2Li_2Al_4(Al_2Be_2Si_4)O_{20}(OH)_4$, and ephesite, $Na_2Li_2Al_4(Al_4Si_4)O_{20}(OH)_4$, as endmembers, because available data suggest some degree of solid solution between these Li-bearing micas and the Li-free margarite and paragonite. For example, the chemical data of Gallagher and Hawkes (1966) and the crystal-structure refinement of Lin and Guggenheim (1983) imply that solid solution between bityite and margarite may be close to complete. The extent of ephesite solid solution in margarite is less well known. Margarites containing up to 0.29 wt% Li_2O but less than 0.01% BeO (Schaller et al., 1967) may have some 7–8 mol% ephesite. Ephesite solid solution with paragonite may be more extensive, for Drits and Semenov (1975) reported a mica containing 67 mol% ephesite, 25% paragonite, 6% margarite, and 2% muscovite. We selected the Sr and Ba dioctahedral endmembers because the most appropriate exchange location for Sr and Ba substitution is the interlayer site. Sr commonly substitutes for Ca and K. In calculating a formula for "sodian margarite" containing 0.62 wt% Sr reported by Afanasyev and Aydinyan (1952), Schaller et al. (1967) assumed that the resulting 0.06 Sr per formula unit substituted for Ca. Ba substitutes for K in muscovites (e.g., 17% in Dunn, 1984; 22% in Dymek et al., 1983).

The unusual composition of the Antarctic margarite and paragonite can be attributed not only to their ability to incorporate Li, Be, and Sr, but also to local enrichment of Li, Be, and Sr during the retrograde metamorphic stage. Li probably originated from breakdown of nearby staurolite (grain 2, Table 1, Grew and Sandiford, 1984), which contains about 0.2 wt% Li_2O (IMMA analysis, this study).

Table 3. Hypothetical components and substitution operations in margarite and paragonite (in terms of cations per 200 + 4OH)

Mineral	Component Cation composition*	Operation	Percentage in	
			Margarite	Paragonite
Paragonite	Na ₂ Al ₄ (Al ₂ Si ₆)	—	3	77
Margarite	Ca ₂ Al ₄ (Al ₄ Si ₄)	CaAlNa ₋₁ Si ₋₁	77	7
Ephesite	Na ₂ Li ₂ Al ₄ (Al ₄ Si ₄)	LiAlSi ₋₁	4	5
—	Na ₂ MgAl ₄ (Al ₄ Si ₄)	MgAl ₂ Si ₋₂	14	5
Bityite	Ca ₂ Li ₂ Al ₄ (Al ₂ Be ₂ Si ₄)	CaLiBeNa ₋₁ Si ₋₁	1	0
—	Sr ₂ Al ₄ (Al ₄ Si ₄)	SrAlNa ₋₁ Si ₋₁	1	5
—	Ba ₂ Al ₄ (Al ₄ Si ₄)	BaAlNa ₋₁ Si ₋₁	0	1

* K is included with Na, and Fe with octahedral Al.

The source of Be is problematic, as no Be was detected with the *IMMA* in staurolite or tourmaline. The source of Sr may be the same as that of Ca. Grew and Sandiford (1984) suggested the following possible sources for Ca: (1) calcic plagioclase that formed early in the metamorphic history and subsequently was replaced by albite, (2) solutions percolating through the rock during the late stage, or (3) tourmaline. An analysis at one spot in the rim of a tourmaline prism in sample 4017E (electron-microprobe analysis given in Table 2, Grew and Sandiford, 1984) yields roughly 0.05 wt% SrO (estimated by applying the sensitivity factor given in Table 2 to *IMMA* data from a previous session).

Another factor leading to Li and Sr enrichment in the margarite and paragonite may be the silica-undersaturated environment of sample 4017E. Li incorporation as an ephesitic component in paragonite and margarite may be enhanced in quartz-free rocks because ephesite is not stable in the presence of quartz (Chatterjee and Warhus, 1984). The Antarctic sample illustrates that more extensive compositional variation in micas, particularly substitutions involving Li and Sr, is possible in the appropriate chemical environment. Staurolite in which Li is sought analytically generally is found to contain Li (see references cited in Grew and Sandiford, 1984; also Dutrow et al., 1984; and unpub. data reported by Hinthorne, 1975), and consequently, secondary mica forming from staurolite may incorporate Li. Moreover, limited Li incorporated in margarite as ephesite in conjunction with increasing octahedral occupancy through the substitution MgAl₂Si₋₂ (Table 3) may explain the margarite compositions with excess ¹⁹Al discussed by Frey et al. (1982).

B in micas

The limited extent of Li, Be, and Sr substitution in natural paragonite and margarite may be controlled largely by the availability of these elements, which are present in low concentrations in most rocks (e.g., Sr in Guggenheim, 1984, p. 61). Furthermore, there are no obvious crystallochemical constraints on substitutions involving Li, Be, and Sr. However, micas rarely incorporate much B, even in B-rich environments. Harder (1959) and Ofstedal (1964) reported that a few muscovites incorporate

as much as 0.1–0.6 wt% B₂O₃, whereas margarite, paragonite, biotite, lepidolite, and most muscovites contain 0.1% or less. More recent data on micas associated with borosilicate minerals are further evidence that micas in general incorporate little B, even when B is available. One example is biotite formed at temperatures near 700°C in association with B-bearing kornepupine, an Al-rich Mg-Fe silicate. Of six such biotites we analyzed, five contain less B₂O₃ than the biotite in sample 2045G (0.01% B₂O₃, Table 1), a rock lacking borosilicates, and the sixth contains roughly the same amount as biotite 2045G. Another example is polyolithionite [K₂Al₂Li₄(Si₈O₂₀)F₄], associated with reedmergnerite (NaBSi₃O₈) and other borosilicates in alkali pegmatites from the southern Tian-Shan Mountains (USSR). Dusmatov et al. (1972) reported B₂O₃ contents of 0.06–0.10% (average 0.08), and W. Schreyer and G. Werdinger obtained 0.13% B₂O₃ (unpub. data) on polyolithionite in a specimen from the Fersman Museum of reedmergnerite-quartz rock from the same alkali intrusive complex. The low B contents of paragonite and margarite in 4017E imply that these two dioctahedral micas do not incorporate B in a B-rich environment.

The mechanism limiting B substitution in micas remains poorly understood. Hazen and Wones (1972) reported synthesizing at 720°C a mica of composition K₂Mg₆(B₂Si₆O₂₀)(OH)₄. However, according to their equations for a geometric fit of the tetrahedral and octahedral sheets (see Hazen and Wones, 1978, p. 886), such a composition is impossible. The BSi₃ tetrahedral sheet is too small to match the Mg octahedral sheet. Given a mean tetrahedral metal–oxygen distance (*d_t*) of 1.58 Å (for BSi₃) and a mean octahedral metal–oxygen distance (*d_o*) of 2.06 Å (for Mg), the ratio *d_t*/*d_o* of 1.30 falls outside the range of 1.235–1.275 that Hazen and Wones (1978) specified for trioctahedral-mica stability. Robert (1981, p. 46), who reached the same conclusion, proposed that incorporation of ¹⁹Mg will enlarge the tetrahedral sheet sufficiently to match the octahedral sheet. Robert (1981) reported infrared evidence for ¹⁹Mg in a synthetic B-bearing “micaceous phase” that he obtained during his unsuccessful attempts to synthesize a mica of composition K₂Mg₆(B₂Si₆O₂₀)(OH)₄. However, the mechanism proposed by Robert (1981) may not be operative in meta-

morphic trioctahedral micas, as these micas generally contain enough Al to fill the tetrahedral sites. An exception is the biotite 2045G, in which $Si + Al + B = 7.862$. Because the ideal value is 8, addition of ^{IV}Mg could make up the difference. Nonetheless, the B content of the mica is low. In summary, incorporation of B in micas, dioctahedral as well as trioctahedral, is most likely restricted by the mismatch of a B-bearing tetrahedral sheet and the octahedral sheet; the influence of temperature is probably much subordinate. Even a few scattered B-bearing tetrahedra must create enough distortions in the mica structure to destabilize it, therefore limiting the amount of B that can be incorporated.

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