

Li in minerals from the Ilímaussaq alkaline intrusion, South Greenland

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Li was analysed by instrumental neutron activation analysis and Cerenkov counting in 120 mineral samples (30 species) from the Ilímaussaq alkaline intrusion, South Greenland. More than 0.23 wt. % Li (0.5 wt. % Li_2O) is found in polyolithionite, neptunite, riebeckite, Na-cookeite, ephesite, arfvedsonite, gerasimovskite and astrophyllite. Arfvedsonite (200–2500 ppm Li) carries the bulk of Li in most of the highly alkaline rocks. Li-Mg and Li-F relations indicate that the distribution of Li is controlled by the structure of minerals, their absolute contents of Mg and F and the fractionation stage within the intrusion. Li is probably linked with F in the fluid state and this linkage continues into crystallising phases where Li occupies sites which also accommodate Mg. Li/Mg and Li/F ratios of Ilímaussaq rocks and minerals are higher than in equivalent materials from the Lovozero intrusion (Kola, Russia). The Li-Mg- Fe^{2+} geochemical association at Ilímaussaq ($\text{Fe}^{2+} \gg \text{Mg}$) and Lovozero ($\text{Fe}^{2+} > \text{Mg}$) contrasts with the commercially important Li-rich but Mg- Fe^{2+} -poor association found in certain granite pegmatites and greisenised granites.

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Introduction

Studies of Li distribution among the minerals of igneous rocks have emphasised that Li is concentrated in F-bearing minerals containing significant levels of Mg, such as micas, amphiboles and tourmaline, and to a lesser extent in Na-rich minerals, such as feldspars (Heier and Adams 1964; Wilson and Long 1983; Henderson et al. 1989). The aim of this publication is to present data on the distribution of Li, Mg and F in minerals of the Ilímaussaq intrusion, South Greenland, and to discuss this distribution in terms of the crystallographic properties of Li and the petrological history of the intrusion. Comparisons are made with similar materials from the Lovozero intrusion, Russia, and with the commercially important, Li-rich granite pegmatites and greisenised granites.

Geological background

The Ilímaussaq intrusion (Fig. 1) is the youngest of a number of syenitic bodies in the late Precambrian Gardar province of South Greenland. The intrusion has been described by Ussing (1912), Sørensen (1962, 1969, 1970), Ferguson (1964), Hamilton (1964), Bohse et al. (1971), Engell (1973), Larsen (1976), Bailey et al. (1981a) and Larsen and Sørensen (1987). It consists of an outer envelope of augite syenite (phase 1) and two thin sheets of peralkaline granite under the roof (phase 2) which were followed by a layered series of peralkaline nepheline syenites (phase 3). The flotation cumulates of the layered series are rich in sodalite (sodalite foyaité, naujaite) whereas the bottom cumulates (kalkortokites) contain abundant arfvedsonite, eudialyte and microcline. The intervening, residual lujavrites were emplaced in the sequence: aegirine lujavrite I, aegirine lujavrite II, lujavrite transition zone, arfvedsonite lujavrite and then medium- to coarse-grained (M-C) lujavrite. Locally, there are lujavrite varieties

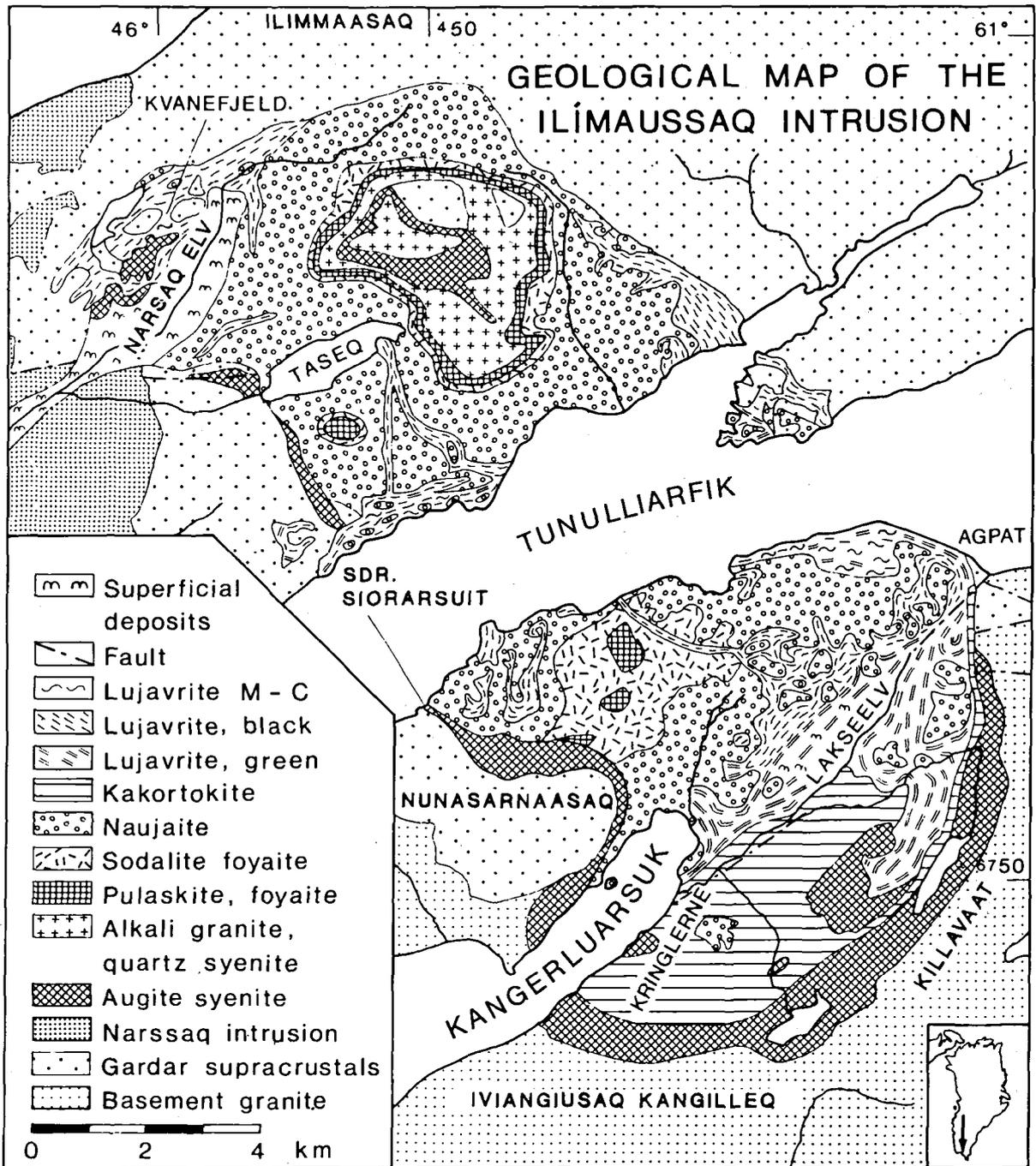


Fig. 1. Geological summary map of the Ilímaussaq intrusion based on Ferguson (1964).

which are rich in naujakasite and villiaumite and, in the roof zone, the lujavrites have notable contents of U and Th (Sørensen et al. 1974).

The minerals investigated were mainly taken from the peralkaline nepheline syenites and their pegmatitic and hydrothermal derivatives.

Existing data

The Ilímaussaq intrusion constitutes a major Li anomaly in the world of igneous rocks. Li contents in chilled marginal augite syenite are only about 23 ppm

Table 1. Previously published contents of Li, F and Mg in Ilímaussaq minerals.

Mineral	Li (ppm)		F (%)		Mg (%)	
	range	median	range	median	range	median
Aegirine	20–160	50	–	–	0.00–0.87	0.11
Albite	1–18	9	–	–	–	–
Analcime	6–7	6.5	–	–	–	–
Arfvedsonite	100–1015	940	0.29–1.23	0.88	0.02–0.72	0.19
Biotite-lepidomelane	n00	n00	–	–	–	–
Katophorite	300	330	n.d.–0.80	0.40	0.15–0.98	0.57
Chkalovite	0.5–10	5	–	–	n.d.–0.10	0.05
Na-Cookeite	2300	2300	–	–	–	–
Eudialyte	10–330	60	0.28	0.28	0.05–0.16	0.09
Feldspar	25	25	–	–	–	–
Mafics	160	160	–	–	–	–
Microline	5–17	11	–	–	tr.	tr.
Microline + nepheline	11	11	–	–	–	–
Neptunite	6600	6600	–	–	0.18	0.18
Polyolithionite	2.72–4.34%	3.15%	6.4–8.59	7.61	n.d.–0.04	0.02
Potash feldspar	2	2	–	–	–	–
Riebeckite	1500–3900	2700	0.40	0.40	0.16	0.16
Sodalite	15–90	32	–	–	–	–
Steenstrupine	100–300	200	0.27–1.32	1.24	0.03–0.19	0.05
Ussingite	30	30	0.09	0.09	0.34	0.34

–: data unavailable, tr.: trace, n.d.: not detected

Results compiled from: Bøggild (1953), Sørensen (1962), Hamilton (1964), Gerasimovsky (1969), Semenov (1969), Povarennykh et al. (1970), Engell (1973), Sørensen et al. (1974), Larsen (1976), Makovicky and Karup-Møller (1981).

but rise to 90–300 ppm in subsequent agpaitic rocks (sodalite foyaite, naujaite, kakortokite, aegirine lujavrite) and to 600–1000 ppm in the residual arfvedsonite lujavrites (Gerasimovsky 1969; Bailey et al. 1981b). Concentrations of several thousand ppm probably occur in scattered pegmatites containing Li-mica.

The literature contains Li results on 45 mineral samples (20 species) from Ilímaussaq (Table 1). Data for Li, Mg and F have only occasionally been determined on single mineral specimens. The variety of analytical techniques employed explains some of the observed range of values. The presence of impurities in mineral separates has received no (published) attention, though the widespread occurrence of Li-rich phases indicates that this is a potential cause of serious errors.

Eight Ilímaussaq minerals contain 0.23 wt. % or more of Li (equivalent to 0.50 wt. % or more of Li₂O) (Bøggild 1953; Sørensen 1962; Semenov 1969; this paper).

Polyolithionite, or according to more recent studies lepidolite, $\text{KLi}_2\text{Al}(\text{F}, \text{OH})_2\text{Si}_4\text{O}_{10}$, is a widespread accessory mineral in naujaite and lujavrite and their derivatives. It also occurs in foyaite, kakortokite and in quartz-feldspar veins cutting alkali granite. High temperature polyolithionite is lamellar and occurs in rocks and pegmatites. Flaky varieties are found in the central zones of pegmatites and in albitization zones. Earthy polyolithionite occurs in low temperature hydrothermal zeolite vugs and contains 2.72% Li in contrast to 3.0–4.2% in the earlier varieties (Semenov 1969).

Neptunite, $\text{KNa}_2\text{Li}(\text{Fe}, \text{Mn})_2\text{Ti}_2\text{OSi}_4\text{O}_{11}$, is most characteristic for the naujaite and M-C lujavrite hy-

drothermal veins but also occurs in quartz veins in alkali granite and in cross-cutting kakortokite pegmatites. It is typically associated with altered arfvedsonite and eudialyte. Li (0.66%) does not substitute for Fe or alkalis but is an essential component of the neptunite formula (Semenov 1969).

Riebeckite, $\text{Na}_2\text{Fe}_3^{+2}\text{Fe}^{+3}\text{Si}_8\text{O}_{22}(\text{OH}, \text{F})_2$, is common in late, usually hydrothermal derivatives of naujaite and alkali granite. Li contents range from 0.15–0.39%. Li⁺ may substitute for Fe²⁺ (Semenov 1969).

Na-cookeite, $\text{Al}_2(\text{OH})_2\text{AlSi}_3\text{O}_{10}^{1-}\text{LiAl}_2(\text{OH})_6^{1+}$, occurs in the central hydrothermal zone of an M-C lujavrite pegmatite in the Narssaq river (Semenov 1969). It is Li-poor (0.23%) and Na-rich compared with previously described cookeites, and is probably a special polymorphic modification of cookeite.

Ephesite, $\text{NaLiAl}_2(\text{OH})_2\text{Al}_2\text{Si}_2\text{O}_{10}$, with 1.21% Li also occurs in the hydrothermal zone of an M-C lujavrite pegmatite at Narssaq river (Semenov 1969; Drits and Semenov 1975). Ephesite is a member of the margarite group and Li fills the vacant octahedral position as Ca²⁺ is replaced by Na⁺ (Schaller et al. 1967).

These five minerals are mainly developed at the hydrothermal stage. Arfvedsonite, $\text{Na}_3\text{Fe}_4^{+2}\text{Fe}^{+3}\text{Si}_8\text{O}_{22}(\text{OH})_2$, however, is an abundant rock-forming mineral in the peralkaline rocks and their derivatives. Only in aegirine lujavrite I, the earliest of the lujavrite varieties, do Li values in arfvedsonite rise above 0.23%. The poikilitic arfvedsonite, up to 8 vol. %, is considered to have crystallised from a Li-rich interstitial coexisting magma (Bailey and Gwozdz, submitted).

Table 2. Concentration of Li, F and Mg in Ilmaussaq minerals. Li and F by instrumental neutron activation analysis and Cerenkov counting; Mg by X-ray fluorescence analysis.

Mineral	Sample number	Rock type	Local. ¹⁾	Collec. ²⁾	Li ppm	F %	Mg %
Acmite	21070A	Aegirine lujavrite	K	OL	26	n.d.	n.d.
Acmite	21070	Aegirine lujavrite	K	OL	47	n.d.	0.04
Aegirine	108107-191*	Aegirine lujavrite I	K		10	n.d.	0.02
Aegirine	108107-166	Aegirine lujavrite I	K		26	n.d.	0.03
Aegirine	108107-146	Aegirine lujavrite I	K		21	n.d.	0.03
Aegirine	108107-127	Aegirine lujavrite I	K		30	n.d.	0.02
Aegirine	108107-106	Aegirine lujavrite I	K		32	n.d.	0.03
Aegirine	108107-096	Aegirine lujavrite I	K		16	n.d.	0.03
Aegirine	108107-069	Aegirine lujavrite I	K		37	n.d.	0.02
Aegirine	108107-046	Aegirine lujavrite I	K		13	n.d.	0.03
Aegirine	108107-022	Aegirine lujavrite I	K		17	n.d.	0.04
Aegirine	108107-004	Aegirine lujavrite I	K		27	n.d.	0.03
Aegirine	149541	Lujavrite pegmatite	Kv		25	n.d.	0.04
Aegirine	21070	Aegirine lujavrite	Tu	OL	23	n.d.	0.02
Aegirine	21178	Aegirine lujavrite	K	OL	33	n.d.	0.02
Aegirine	18453	Aegirine lujavrite	Kv		16	n.d.	0.05
Aegirine	65930	Hydrothermal vein	T	JE	24	n.d.	-
Analcime	71	Steenstrupine-analcime arfvedsonite pegmatite	K	HS	12	0.06	0.01
Analcime	65914	Analcime vein	T	JE	3.8	0.13	0.01
Analcime + natrolite	108107-191	Aegirine lujavrite I	K		32	n.d.	n.d.
Analcime + natrolite	108107-166	Aegirine lujavrite I	K		24	n.d.	0.01
Analcime + natrolite	108107-146	Aegirine lujavrite I	K		30	0.11	0.01
Analcime + natrolite	108107-127	Aegirine lujavrite I	K		27	n.d.	0.01
Analcime + natrolite	108107-106	Aegirine lujavrite I	K		38	n.d.	0.01
Analcime + natrolite	108107-096	Aegirine lujavrite I	K		21	n.d.	0.02
Analcime + natrolite	108107-069	Aegirine lujavrite I	K		23	n.d.	0.02
Analcime + natrolite	108107-046	Aegirine lujavrite I	K		15	n.d.	0.03
Analcime + natrolite	108107-022	Aegirine lujavrite I	K		21	n.d.	0.02
Arfvedsonite	154351	Sodalite foyaite	K		489	0.57	0.05
Arfvedsonite	149594	Naujaite pegmatite	K		281	0.59	0.21
Arfvedsonite	154717B	Naujaite pegmatite	K		243	-	-
Arfvedsonite	154309	Naujaite	K		255	0.24	n.d.
Arfvedsonite	57041	Naujaite	K	HM	462	0.55	0.05
Arfvedsonite	154331	Naujaite	K		420	0.68	0.08
Arfvedsonite	108133	Black lujavrite	K		289	0.36	0.04
Arfvedsonite	108131	Black lujavrite	K		370	0.17	0.06
Arfvedsonite	108107-33.7	Naujaite xenolith in aegirine lujavrite I	K		233	0.15	0.51
Arfvedsonite	154343	Black kakortokite	K		513	0.47	-
Arfvedsonite	109201	Black kakortokite	K		378	0.45	-
Arfvedsonite	109240	Red kakortokite	K		410	0.33	-
Arfvedsonite	154321	Black kakortokite	K		725	0.20	0.12
Arfvedsonite	108107-191	Aegirine lujavrite I	K		2020	0.16	0.04
Arfvedsonite	108107-166	Aegirine lujavrite I	K		2200	0.27	0.05
Arfvedsonite	108107-146	Aegirine lujavrite I	K		1990	0.28	0.04
Arfvedsonite	108107-127	Aegirine lujavrite I	K		2520	0.23	0.05
Arfvedsonite	108107-106	Aegirine lujavrite I	K		2200	0.22	-
Arfvedsonite	108107-096	Aegirine lujavrite I	K		2090	0.18	0.04
Arfvedsonite	108107-069	Aegirine lujavrite I	K		2250	0.22	0.04
Arfvedsonite	108107-046	Aegirine lujavrite I	K		1980	0.16	0.06
Arfvedsonite	108107-022	Aegirine lujavrite I	K		2350	0.28	0.09
Arfvedsonite	108107-004	Aegirine lujavrite I	K		1970	0.24	0.04
Arfvedsonite	117479	Lujavrite-naujaite contact pegmatite	K		1240	0.28	0.09
Arfvedsonite	65132	Sheared country rock	Kv		2010	0.42	1.7
Arfvedsonite	163055	Central vein in pegmatite	K		1680	0.33	0.06
Arfvedsonite	71	Steenstrupine-analcime arfvedsonite pegmatite	K	HS	1770	0.08	0.05

cont.

Table 2. – Cont.

Mineral	Sample number	Rock type	Local. ⁺⁾	Collec. ^{**)}	Li ppm	F %	Mg %
Arfvedsonite	154370	Aegirine lujavrite	Tu		459	0.37	0.13
Arfvedsonite	119437–092.28	M-C lujavrite	Kv		1570	0.09	0.02
Arfvedsonite	154399	M-C lujavrite	Kv		1490	0.20	–
Astrophyllite (Nb-rich)	119411–7265	Hydrothermal vein	Kv		3000	0.71	2.9
Astrophyllite	LØK AST	Hydrothermal vein	K	LL	223	1.07	0.12
Beta-natrolite	–	Hydrothermal vein	T		1.7	n.d.	0.01
Chkalovite	155233	Ussingite vein	T		8.2	n.d.	n.d.
Epistolite		Ussingite vein	T		36	0.77	0.08
Epistolite	108130	Ussingite vein	T		38	0.97	0.09
Epistolite	155231	Ussingite vein	T		49	0.62	0.17
Epistolite	77512	Ussingite vein	Kv		421	0.13	0.18
Eudialyte	154352	Sodalite foyaite	Tu		268	0.10	–
Eudialyte	154303	Sodalite foyaite	Tu		174	n.d.	–
Eudialyte	60755	Kakortokite	K		61	n.d.	0.02
Eudialyte	57041	Naujaite	K	HM	13	0.06	–
Eudialyte	154331	Naujaite	K		12	n.d.	–
Eudialyte	108107–033.7	Naujaite xenolith in aegirine lujavrite I	K		17	n.d.	0.02
Eudialyte	109269	Red kakortokite	K		8.2	n.d.	n.d.
Eudialyte	109202	Red kakortokite	K		9.7	n.d.	n.d.
Eudialyte	117815	Red kakortokite	K		7.1	n.d.	n.d.
Eudialyte	108136	Red kakortokite	K		18	0.18	0.01
Eudialyte	143610	Feldspar-eudialyte vein	K		8.4	n.d.	–
Eudialyte	108107–191	Aegirine lujavrite I	K		8.6	n.d.	0.03
Eudialyte	108107–166	Aegirine lujavrite I	K		8.4	n.d.	0.02
Eudialyte	108107–146	Aegirine lujavrite I	K		8.5	0.05	0.02
Eudialyte	108107–096	Aegirine lujavrite I	K		7.5	n.d.	0.02
Eudialyte	108107–046	Aegirine lujavrite I	K		13	0.12	0.04
Eudialyte	108107–022	Aegirine lujavrite I	K		15	0.14	0.04
Eudialyte	108107–004	Aegirine lujavrite I	K		32	0.09	0.02
Eudialyte	57033	Aegirine lujavrite I	K	HM	10	n.d.	0.01
Gerasimovskite	77480	Hydrothermal vein	T		12400	2.34	0.01
Igdloite	185101	Hydrothermal vein	Kv		20	0.14	0.05
Kaersutite	77102	Lamprophyre	Kv		106	0.09	3.0
Microcline + nepheline	108107–191	Aegirine lujavrite I	K		5.9	n.d.	n.d.
Microcline + nepheline	108107–146	Aegirine lujavrite I	K		3.0	n.d.	0.01
Microcline + nepheline	108107–096	Aegirine lujavrite I	K		8.2	n.d.	0.01
Microcline + nepheline	108107–046	Aegirine lujavrite I	K		4.9	n.d.	0.01
Microcline + nepheline	108107–022	Aegirine lujavrite I	K		2.0	n.d.	n.d.
Murmanite	65127	Sheared country rock	Kv		346	0.13	–
Naujakasite	149421C	Naujakasite lujavrite	Kv		5	–	0.07
Nenadkevichite	LØK 27A	Hydrothermal vein	T	LL	306	0.62	0.04
Neptunite	65450	Hydrothermal vein	T		5440	0.09	n.d.
Neptunite	65450	Hydrothermal vein	T		6700	0.14	–
Niobophyllite	65112P	Sheared country rock	Kv		157	1.81	n.d.
Niobophyllite	65112P	Hydrothermal vein	Kv		218	0.58	–
Polyolithionite	64684	Naujaite pegmatite	Kv		35400	4.75	n.d.
Polyolithionite	154717A	Naujaite pegmatite	K		33000	–	0.02
Polyolithionite	QEQ	Naujaite pegmatite	K		32400	–	0.04
Polyolithionite	150318	Naujaite pegmatite	K		32000	4.59	n.d.
Polyolithionite	18617	Naujaite pegmatite	K		31300	3.89	0.02
Polyolithionite	64684A	Naujaite pegmatite	Kv		31000	4.97	0.06
Polyolithionite	20770	Naujaite pegmatite	K		29700	4.48	0.03
Polyolithionite	LØK 13R	Kakortokite pegmatite	K	LL	34500	–	0.06

cont.

Table 2. – Cont.

Mineral	Sample number	Rock type	Local. ^{+))}	Collec. ⁺⁺⁾	Li ppm	F %	Mg %
Pyrochlore	65468	Hydrothermal vein	Kv		52	1.41	0.02
Pyrochlore	65450(2)	Hydrothermal vein	Kv		80	1.57	0.02
Pyrochlore	65450(5)	Hydrothermal vein	Kv		101	1.81	0.01
Pyrochlore	65450	Hydrothermal vein	Kv		226	1.00	n.d.
Pyrochlore	65451	Hydrothermal vein	Kv		257	1.66	0.02
Quartz	152140	Alkali granite	Kv		1.2	n.d.	0.01
Sodalite	154331	Naujaite	K		131	0.80	–
Sodalite	108107–033.7	Naujaite xenolith in aegirine lujavrite I	K		33	n.d.	0.01
Sodalite	108118	Hydrothermal vein	Kv		24	n.d.	0.04
Sorensenite	155232	Hydrothermal vein	Kv		11	n.d.	0.05
Steenstrupine	71	Steenstrupine-analcime arfvedsonite pegmatite	K	HS	200	2.39	0.03
Steenstrupine	119408–33.52	Hydrothermal vein	Kv		286	2.66	n.d.
Steenstrupine	18467	Hydrothermal vein	K		145	1.02	0.04
Tugtupite	64750	Hydrothermal vein	T		3.9	n.d.	0.01
Ussingite	65914	Hydrothermal vein	Kv		3.6	0.07	0.02
Ussingite	163056	Ussingite vein in recrystallized naujaite	Tu		98	n.d.	0.03
Villiaumite	119439–393.20	Arfvedsonite lujavrite	Kv		221	–	n.d.

Sample numbers: five- and six-figure numbers are GGU numbers; other numbers and letters are sample numbers of individual collectors; *108107–191 means drill hole 7, 191 m depth (all 1081-drill holes are located outside Kvanefjeld); 119437–092.28 means drill hole 37, 92.28 m depth (all 1194-drill holes are located on the Kvanefjeld).

^{+))} Localities: K Kangerluarsuk area; Kv Kvanefjeld area; T Taseq area; Tu Tunulliarfik area.

⁺⁺⁾ Collectors: JE John Engell; OL Ole Larsen; LL Laust Løkkegaard; HM Harry Micheelsen; HS Henning Sørensen; other samples collected by the authors.

– not determined; F n.d. not detected (<0.05%); Mg n.d. not detected (<0.01%).

Gerasimovskite, $\text{TiNb}(\text{OH})_9$, with 1.24% Li is a weathering product of epistolite in ussingite veins of the Taseq area, north of Tunulliarfik.

Astrophyllite, $(\text{K}, \text{Na})_3(\text{Fe}, \text{Mn})_7(\text{Ti}, \text{Zr})_2\text{Si}_8(\text{O}, \text{OH})_{31}$, with 0.30% Li is found in considerable amounts in the border pegmatite of the kakortokites. The Ilímaussaq astrophyllite is an intermediate member of the Ti-Nb substitution series between astrophyllite and niobophyllite (Semenov 1969).

The alkaline rocks of the Lovozero massif (Kola, Russia) contain other Li-rich micas – tainiolite and biotite (Semenov 1959; Vlasov et al. 1966). Equivalent minerals have not been confidently reported from the Ilímaussaq intrusion.

Li crystallochemistry

Lithium exhibits mixed crystallochemical properties (Cocco et al. 1969).

(1) Li-O bonds. Because of its small ionic radius, Li^+ (0.82 Å) tends to follow Mg^{2+} (0.80 Å), and to a lesser extent Fe^{2+} (0.86 Å) and Al^{3+} (0.61 Å), into octahedral

sites of silicate structures. However, Li^+ does not simply substitute for Mg^{2+} etc. but all these ions compete to enter ferromagnesian minerals where the optimal size of the relevant site varies slightly (Jensen 1973): clinopyroxene (0.79 Å), orthopyroxene and hornblende (0.78 Å), olivine (0.77 Å) and biotite (0.76 Å). Entry of variously charged ions into these optimal sites must satisfy charge balancing within the mineral and this has been confirmed by analysis. Thus plots of 2Mg^{2+} versus $\text{Li}^+ + \text{Al}^{3+}$ in many Li-bearing pyroxenes, amphiboles and micas lie close to a 1:1 negative correlation line. $\text{Li}^{\text{VI}} + \text{Si}^{\text{IV}} = (\text{Fe}^{2+})^{\text{VI}} + \text{Al}^{\text{IV}}$ and other substitution mechanisms have also been suggested (Henderson et al. 1989).

Coordination octahedra around Li^+ can occur either separately or linked together in various ways. The average $\text{Li}^{\text{VI}}\text{-O}$ bond length is 2.14 Å. Li in tetrahedral coordination with O is less important in nature but does occur in petalite. The Li-phosphate ambygonite has an intermediate coordination number.

(2) Li-F bonds. Li readily loses its outermost electron resulting in an extremely reactive chemical character. Li-halide bonds are very stable. In the NaCl structure type, which includes the Ilímaussaq mineral villiaumite

(NaF), Li^+ is in 6-fold coordination and the Li-F bond length is 2.014 Å. LiF_4 tetrahedra, structurally equivalent to SiF_4 tetrahedra, occur in phenakite and cryolithionite.

(3) Li-Na and Li-K substitution. Because of the small size of Li^+ (0.82^{VI} Å) with respect to that of the next alkaline ions Na^+ (1.10^{VI} Å, 1.24^{VIII} Å) and K^+ (1.46^{VI} Å, 1.59^{VIII} Å, 1.68^{XII} Å), only limited substitution is expected. However, experimental work shows that in high-Al micas interlayer Li may replace about one-third of K^+ though natural examples have not yet been recorded (Robert et al. 1983).

Trace element geochemical studies have confirmed these crystallochemical controls and indicated their varying importance in different rock types. Thus Li-Na substitution in plagioclase is probably responsible for more than 50% of the Li in dioritic rocks (Vorontsov and Lin 1966), whereas Li-(Fe^{2+} , Mg) substitution is dominant in the main Li carriers (hornblende, biotite, muscovite) of many granites. In this evolution from intermediate to acid rocks, the proportion of Li-O bonds decreases relative to the abundance of Li-F and Li-OH bonds. Némec (1969) found that F values in pegmatitic muscovites generally correlate with Li values. Bailey (1977) has summarised the close geochemical correlation between Li and F in granitic rocks and minerals; both elements lower granite solidus temperatures and promote water solubility.

Analytical methods

Li and F were analysed by instrumental neutron activation using a fast transfer system and Cerenkov counting coupled to the Danish Nuclear Research Reactor DR2, Risø National Laboratory (Heydorn et al. 1977; Bailey et al. 1981b)). For Li, empirical values for accuracy, precision and lower limit of detection were found to be ± 10 –20%, ± 10 –15% and 0.2 ppm, respectively. The counting techniques were optimised for Li, and F results can only be considered accurate to approximately $\pm 20\%$. The detection limit for F is about 500 ppm. Mg was analysed either directly on powder pellets of the minerals using conventional X-ray fluorescence techniques or using atomic absorption analysis after HF dissolution. Values agree within 0.01% for the two techniques and the detection limit is 0.01% Mg.

A few additional Li and F values were determined by atomic absorption analysis after HF dissolution, and a combined distillation-spectrophotometry technique, respectively. In the latter technique, powdered material was fused with sodium carbonate and subjected to steam distillation from sulphuric acid. Fluorine was extracted on anion resin, selectively eluted with a sodium acetate solution and measured on a Zeiss spectrophotometer (Hansen 1983).

Mineral separation from fine and medium grained rocks was effected by a combination of heavy liquid and magnetic techniques. Coarse material was often amenable to hand picking followed by further crushing and inspection under a binocular microscope. Percentages and types of impurities, up to the 2% level, were estimated and Li contents correspondingly adjusted.

Results

Results for Li, F and Mg on 120 Ilímaussaq mineral samples (30 species) are presented in Table 2.

Arfvedsonite

Among the rock-forming minerals, arfvedsonite contains the highest amount of Li (225–2520 ppm). Arfvedsonite Li contents increase from 225–462 ppm in sodalite foyaite and naujaite, to 378–725 ppm in kakortokite, and reach a maximum of 1970–2520 ppm in aegirine lujavrite I, the earliest of the lujavrite varieties. Arfvedsonites from later lujavrites tend to contain less Li, even though whole-rock values are the highest found in the Ilímaussaq intrusion.

Semenov (1969) found that arfvedsonite from a rare-metal naujaite pegmatite (with eudialyte, epistolite and chkalovite) contained 0.093% Li and 0.80% F. But in naujaite pegmatites free of rare-metal mineralization (i.e. with microcline, sodalite and arfvedsonite) the arfvedsonite only contained 0.009% Li and 0.29% F.

Arfvedsonite crystallizes late in sodalite foyaite and naujaite but is an early phase in kakortokites and arfvedsonite lujavrites. When arfvedsonites from the same crystallization stage are compared, then Li contents are always higher in arfvedsonites from the rock type crystallising later in the Ilímaussaq sequence. It thus seems likely that Li contents increase progressively in the late magmas. The smoothness of this increase could be interrupted if there were extensive fractionation of Li-rich arfvedsonite but there is no clear evidence for such an effect at present.

The maximal values in arfvedsonites from aegirine lujavrite I probably arise from the late crystallization of the arfvedsonite in this cumulate rock type. According to Bailey and Gwozdz (submitted), the average whole-rock Li value for aegirine lujavrite I is only about 200 ppm due to the dilution by the Li-poor cumulus phases. The interstitial material is considered to represent the trapped coexisting magma and contains 540 ppm Li. The interstitial arfvedsonite thus crystallised from a Li-rich magma and has very high Li contents.

Arfvedsonite is probably the main Li carrier in most of the apaitic rocks at Ilímaussaq, though this has only been checked quantitatively for aegirine lujavrite I. Bailey and Gwozdz (submitted) found that, for three sam-

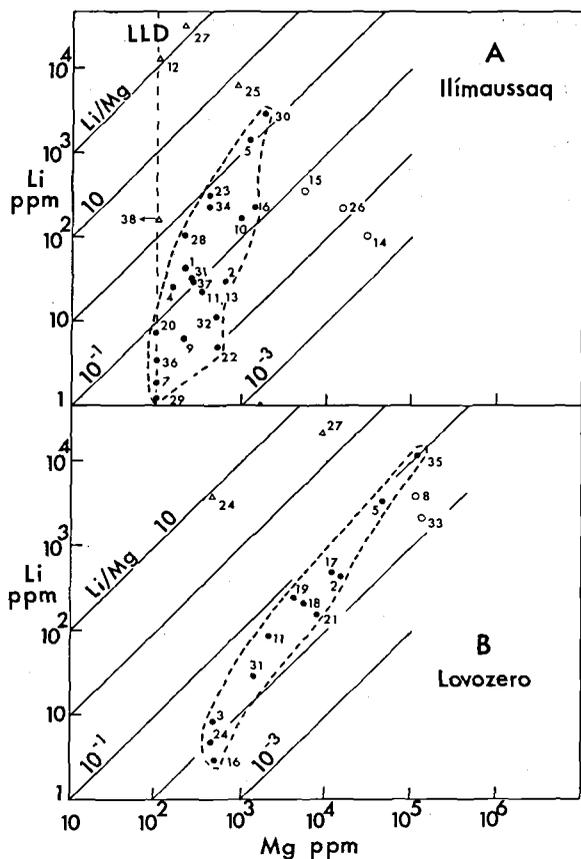


Fig. 2. Li-Mg relations for minerals from the Ilímaussaq (A) and Lovozero (B) alkaline intrusions. Only median values are plotted. Closed circles - main group; triangles - high Li/Mg minerals; open circles - low Li/Mg minerals. LLD lower limit of detection. Data for Ilímaussaq from Tables 1 and 2, this paper. Data for Lovozero from Semenov (1959, 1969), Gerasimovsky & Kholina (1965), Gerasimovsky et al. (1966), Vlasov et al. (1966). Key to numbers: 1 acmite, 2 aegirine, 3 analcime, 4 analcime + natrolite, 5 arfvedsonite, 6 astrophyllite, 7 beta-natrolite, 8 biotite, 9 chkalovite, 10 epistolite, 11 eudialyte, 12 gerasimovskite, 13 igdloite, 14 kaersutite, 15 kataphorite, 16 K-Na feldspar, 17 kupletskite, 18 labuntsovite, 19 lamprophyllite, 20 microcline + nepheline, 21 murmanite, 22 naujakasite, 23 nenadkevichite, 24 nepheline, 25 neptunite, 26 niobophyllite, 27 polyolithionite, 28 pyrochlore, 29 quartz, 30 riebeckite, 31 sodalite, 32 sorensenite, 33 spodiophyllite, 34 steenstrupine, 35 tainiolite, 36 tugtupite, 37 ussingite, 38 villiaumite.

ples of aegirine lujavrite I, the arfvedsonite contained 67 to 91% of the total Li.

There is a tendency for the F and Mg contents of the analysed arfvedsonites to decrease during the Ilímaussaq evolution (Table 2). Larsen (1976) also found a decrease for Mg among the Ilímaussaq amphiboles, though Mg values in early kakortokitic amphiboles are higher than in the preceding naujaites. Gerasimovsky et al. (1966: Table 125) found that F contents of Lovozero amphiboles increased from phase I (1.80%) to phase II (mainly 2.4%) but fell slightly in phase III (2.01%).

Other minerals

The highest contents for Li in Ilímaussaq minerals are found for polyolithionite (2.97–3.54%) and overlap previously published results. High values of Li have been confirmed in neptunite (5440–6700 ppm) and have been recorded for the first time in gerasimovskite (1.24%).

Up to several hundred ppm Li are commonly found in the Zr, Nb, Ti and Th silicates: astrophyllite, epistolite, nenadkevichite, niobophyllite, pyrochlore and steenstrupine. Niobophyllite and astrophyllite probably form a solid-solution series at Ilímaussaq (Semenov 1969) and it is notable that astrophyllite, which is the end-member rich in alkalis, also has a high level of Li (3000 ppm) compared to niobophyllite (157–218 ppm). However, a second astrophyllite sample only contains 223 ppm Li. This recalls the wide range recorded by Ganzeyev et al. (1969) who found Li values ranging from 130–5900 ppm for astrophyllite.

Surprisingly, villiaumite, whose structure and chem-

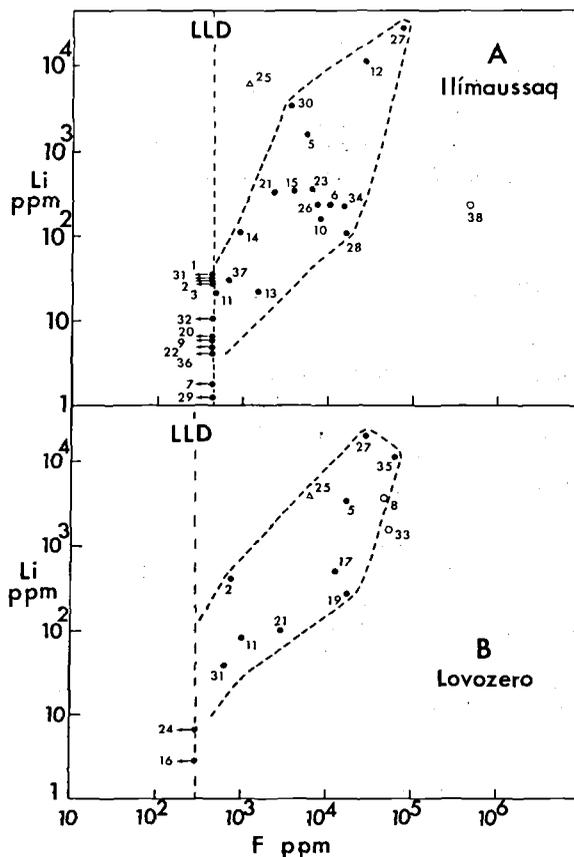


Fig. 3. Li-F relations for minerals from the Ilímaussaq (A) and Lovozero (B) alkaline intrusions. Only median values are plotted. Closed circles - main group; triangles - high Li/F minerals; open circles - low Li/F minerals. Data sources and key to numbers as in Fig. 2.

istry might be expected to readily incorporate Li, contains only 221 ppm.

Except for arfvedsonite, there are no rock-forming minerals which consistently contain more than 50 ppm Li. Aegirine (10–37 ppm), eudialyte (generally 7.5–32 ppm), microcline + nepheline (2.0–8.2 ppm), sodalite (24–33 ppm) and zeolites (3.8–38 ppm) all have distinctly low Li contents when the Li-rich nature of the intrusion is considered. They are also poor in F.

Li values in aegirine are unrelated to stratigraphic height in a 200 m drill core through aegirine lujavrite I (Bailey and Gwozdz, submitted) but show a closer correlation to the percentage of interstitial, coexisting magma in each sample.

Acmite, commonly developed after aegirine or arfvedsonite at Ilímaussaq, shows similar Li contents to aegirine (Table 2).

Eudialyte exhibits high levels of Li (174–268 ppm) in the early sodalite foyaites, but levels fall to 12–17 ppm in the succeeding naujaite and 7.1–18 ppm in kakortites. Similar low values are found in the deeper levels of the 200 m drill core through aegirine lujavrite I but the values rise to 10–32 ppm in the top 50 m. The reasons for these variations are unknown as yet. Eudialyte crystallises late in sodalite foyaite and naujaite, but early in kakortokites and aegirine lujavrite I.

Early crystallising microclines and nephelines in aegirine lujavrite I only contain 2.0–8.2 ppm Li whereas the interstitial zeolites – analcime and natrolite – contain 15–38 ppm. This is in agreement with the build-up of Li in the interstitial, residual magma of this rock type.

Bailey and Gwozdz (submitted) have pointed out that the low values of Li in the early crystallising phases of aegirine lujavrite I must imply low Li partition coefficients between these phases and the coexisting magma (about 540 ppm Li). Li thus shows a much greater preference for the melt phase and this may be an innate feature of alkali-, volatile-rich agpaite melts (cf. Steenfelt and Bohse 1975).

The lowest Li values in Ilímaussaq minerals are found for the leucocratic phases – mainly alkali-bearing silicates. These include analcime (3.8–12 ppm), beta-natrolite (1.7 ppm), chkalovite (8.2 ppm), naujakasite (5 ppm), quartz (1.2 ppm), tugtupite (3.9 ppm) and ussingite (3.6 ppm). Reliable results demand very high purity separation of these minerals. In some cases, we consider the lowest analytical values from several samples of one mineral to be the most reliable.

In general, the results of the present study confirm the previously published analyses on Li in Ilímaussaq minerals. The main differences are in the extrapolation of arfvedsonite values to higher levels in the later lujavritic rocks and the recognition of a group of Zr-Nb-Ti-Th silicates with hundreds of ppm Li. In addition, there is a reduction of Li contents in several leucocratic phases compared to earlier publications. We suspect that the investigated separates have not always been pure.

Li-Mg and Li-F relations

Li-Mg and Li-F relations for Ilímaussaq minerals are shown in Figs. 2A and 3A. Comparable plots for Lovozero (Kola, Russia) minerals are given in Figs. 2B and 3B. Only average values for minerals have been plotted.

Most of the minerals from Ilímaussaq and Lovozero fall within a major grouping with positive correlations between Li and Mg, and between Li and F. Li/Mg ratios and, to a lesser degree, Li/F ratios increase as contents of Li, Mg and F increase. Contents of these elements tend to increase in the following sequence: alkali-bearing leucosilicates – Ti-Zr-Nb silicates – pyroxenes – amphiboles – micas. Although not investigated in detail, Li shows similar, though weaker correlations with Fe^{2+} and OH (structural H_2O). A Li-Na correlation, however, cannot be demonstrated.

Outside the main grouping, we distinguish three small sets of minerals: (a) minerals with very high Li/Mg and Li/F ratios (neptunite, polyolithionite at Lovozero), (b) minerals with distinctly low ratios (biotite and spodiophyllite at Lovozero) and (c) villiumite with very high Li/Mg and very low Li/F.

For all these groupings, and for virtually every mineral species, the Lovozero samples have higher Li, Mg and F contents but the Ilímaussaq samples have clearly higher Li/Mg and slightly higher Li/F ratios. This is consistent with the whole-rock geochemical comparisons made by Gerasimovsky (1969).

The Li/Mg ratios of rocks and minerals at Ilímaussaq are remarkably high. For example, polyolithionite has an average Li/Mg ratio of roughly 3 at Lovozero but 150 at Ilímaussaq. Arfvedsonite averages 0.054 at Lovozero and about 0.53 at Ilímaussaq. The ratio rises to about 3.7 in arfvedsonite from aegirine lujavrite I and to 7.9 in the final M-C lujavrite. In these late-stage arfvedsonites, Li can be classified as a minor element whereas Mg occurs at trace element levels. The low contents of Mg in the Ilímaussaq intrusion, especially at the agpaite stage, probably explain the absence of the Mg-rich Lovozero mica, tainiolite.

Gerasimovsky et al. (1966) concluded that, for Lovozero minerals, Li is characterised by an association with Mg, Fe^{2+} , Na and F. Only the first two elements are reported as controlling the mineralogical distribution of Li at Ilímaussaq (Gerasimovsky 1969).

Discussion

The Li-Mg and Li-F relations for Ilímaussaq and Lovozero minerals, as well as the similar occurrences of particular mineral species and associations, are consistent with similar origins and evolutions for these two peralkaline SiO_2 -undersaturated intrusions. This close

relationship has been emphasised several times (a.o. Ussing 1912; Gerasimovsky et al. 1966; Semenov 1969; Sørensen 1969, 1970).

The mineralogical relationship of Li with Mg and F arises from two different crystallochemical controls. The Li-Mg correlation, and probably also the Li-Fe²⁺ correlation, is expected on the classic rules of diadochy proposed by Goldschmidt (1954). Because of its similar ionic radius but lower valency, Li⁺ is admitted into Mg²⁺-bearing minerals. Because Li is admitted, the Li/Mg ratio of a mineral species should increase during fractionation; this is well exemplified by the Ilímaussaq arfvedsonites. The Li-Mg correlation through most of the mineral species also points to replacement of Mg by Li.

Partition coefficient theory (see above) would emphasise that comparing the Li and Mg ionic radii deviations from the ideal site sizes in clinopyroxene (0.03:0.01 Å), hornblende (0.04:0.02 Å) and biotite (0.06:0.04 Å) indicates that clinopyroxene greatly favours Mg occupancy rather than Li whereas biotite only slightly favours Mg rather than Li. Thus Li/Mg ratios are expected to increase in the sequence: pyroxene – amphibole – mica, and this tendency is probably valid even for the particular pyroxenes, amphiboles and mica species found in the Ilímaussaq and Lovozero intrusions.

Tauson (1965) also emphasised that Li/Mg ratios can be linked to crystallographic controls. For example, in micas the medium-sized cations (such as Mg, Fe²⁺, Li) form a sheet-like grouping and are bound in octahedral coordination with four O and two (OH, F) anions. This bonding is much weaker in amphiboles. Since Li – (OH, F) bonding is more stable than Mg – (OH, F) bonding, the replacement of Mg by Li should occur more readily in micas than in amphiboles and Li/Mg ratios will be accordingly higher. This feature is found both at Ilímaussaq and Lovozero.

The positive Li-Mg correlation in the major grouping of mineral species at Ilímaussaq and Lovozero is the opposite to that observed for individual mineral series, e.g. arfvedsonite, or for isomorphous series where major element diadochy of Li and Mg occurs. Thus, in the two Lovozero mica series, (phlogopite)-spodiophyllite – tainiolite and tainiolite-polyolithionite, values for Li and Li/Mg increase as Mg decreases (Semenov 1959). Heterovalent substitution of the types Li⁺ + (Fe, Al)³⁺ → 2Mg²⁺ or Li⁺ + Si⁴⁺ → Mg²⁺ + Al³⁺ yields the negative correlation between Li and Mg which is visible in Fig. 2 for the Li-, Mg-rich minerals.

The increasing Li/Mg ratios with increasing Mg contents in the main grouping of minerals probably reflect the increased levels of F in the Mg-rich members of the group. Li⁺ has a greater affinity than Mg²⁺ for halogens, particularly F⁻ and should thus be preferentially incorporated in F-rich Mg²⁺-bearing minerals. Li-F linkages probably develop within the silicate melt and pass over into crystallising phases (Kogarko et al. 1968).

High F contents in magmas are known to promote the stability of Li-F micas at the expense of normal micas (Rieder 1971).

The coherent Li-Mg relation through the main group of minerals suggests that the same controls will also apply to the Li-, Mg- and F-poor phases, the alkali-bearing leucosilicates. There is no clear evidence that Li replaces Na in these minerals as suggested by Goldschmidt et al. (1934).

Outside the main group of minerals, there are phases with distinctly different Li-Mg and Li-F relations. At Ilímaussaq and Lovozero, neptunite has distinctly high Li/Mg and Li/F ratios. This is probably due to the very unusual structure of this mineral. Li is an essential component, rather than a substitutional component, and the coordination octahedra around Li occur separately rather than linked together as in amphiboles and micas (Cannillo et al. 1966).

Very low Li/Mg ratios are anticipated in the mafic phases of the early augite syenite as the Mg contents of these minerals are the highest at Ilímaussaq (Larsen 1976, 1981) but whole-rock Li values are the lowest (about 23 ppm). Biotite from Lovozero possesses low Li/Mg and low Li/F ratios (point 8 on Figs. 2B and 3B).

In the mineral villiaumite, NaF, which has a NaCl structure, Na (and probably Li) is in octahedral coordination but the bond length is smaller than Li^(VI) – O bonds. The Li/Mg ratio of villiaumite is exceptionally high but the Li/F ratio is exceptionally low (point 38 on Figs. 2A and 3A). Presumably there is a clash between the affinity of Li for F (greater than that of Na) and its limited possibility of replacing the much larger Na ion.

Li-Na substitution more clearly occurs in cookeite. This mineral usually contains 1.2–2.1% Li and 0.0–1.3% Na but at Ilímaussaq there is 0.23% Li and 3.34% Na (Semenov 1969).

Li-rich geochemical associations

The Ilímaussaq and Lovozero intrusions constitute a distinct, though rare, type of Li-rich association. Its most characteristic feature is the low content of Si in the rocks and minerals of the association. This results in the widespread substitution of Li for Fe²⁺ and Mg in apatitic minerals (arfvedsonite, aegirine, eudialyte etc.) at Ilímaussaq (Fe²⁺ >> Mg) and Lovozero (Fe²⁺ > Mg); there is only a weak development of independent Li minerals. A similar situation occurs in certain Li-rich granites and greisens where there is again sufficient Fe²⁺ and Mg and the mineralogy is dominated by a variety of Li-F-bearing micas (Henderson et al. 1989).

In major contrast to these two associations, there are certain Li-rich granite pegmatites and greisenised granites (Mulligan 1965; Anstett et al. 1990) which are characterised by very low Fe²⁺ and Mg contents and the development of many Li phases – spodumene, ambly-

gonite, lepidolite, zinnwaldite, petalite, cryolithionite, triphylite-lithiophilite and others. Some of these occurrences are commercially valuable due to the large tonnage of Li minerals. The low Fe and Mg character of this geochemical association minimises the substitution of Li and leads to its concentration in independent Li minerals.

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Dansk sammendrag

Lithium i mineraler fra den alkaline Ilímaussaq intrusion i Sydgrønland.

Polyolithionit, neptunit, riebeckit, Na-cookeit, ephesit, gerasimovskit, astrophyllit og arfvedsonit indeholder op til over 0.5 vægt % Li_2O . Hovedparten af intrusionens Li er bundet i arfvedsonit, der generelt indeholder 200–2500 ppm Li. Li-indholdet afhænger af mineralernes struktur, deres indhold af Mg og F og fraktioneringsstadiet. Li er sandsynligvis knyttet til F i fluidfasen, en relation, der fortsætter under krystallisationen, hvor Li substituerer Mg. Li/Mg og Li/F er i Ilímaussaqs mineraler højere end for tilsvarende mineraler fra Lovozero intrusion, Kola halvøen.

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