

# Geochemical overview of the Ilímaussaq alkaline complex, South Greenland

John C. Bailey, Raymond Gwozdz, John Rose-Hansen and Henning Sørensen

The Ilímaussaq alkaline complex is one of the latest members of the Mesoproterozoic Gardar igneous province. It was emplaced in three phases, (1) augite syenite, (2) alkali acid rocks and (3) the dominant agpaitic nepheline syenites. The immediate parent for the nepheline syenites is the augite syenite whose geochemical features, such as the low Zr/Nb ratio (3.7), match benmoreites from a nearby alkali basalt series.

The agpaitic nepheline syenites are characterised by exceptionally high contents of Zr, Hf, Nb, Ta, REE, Th, U, Sn, Li, Be, Rb, Zn, Pb, Sb, W, Mo, As and Ga, and the volatile elements F, Cl, Br, I and S but exceptionally low levels of Ba, Sr, Co, Cu, Ni, Sc, V and Cr. Fractionation of cumulus phases with a distinctive geochemistry, such as sodalite (rich in Cl, Br, I, B), eudialyte (Zr, Hf, Nb, Ta, W, As) and arfvedsonite (Co, Sc, V, Cr), caused exhaustion of these elements in the residual magmas. The agpaitic magmas underwent extreme fractionation with the final lujavrite forming after 99% crystallisation of the augite syenite. Zirconium was apparently soluble in Ilímaussaq melts up to a concentration of c. 9000 ppm. Whole-rock analyses define a number of discrete Zr–U arrays which are restricted to certain intervals of the cumulate stratigraphy and are taken as evidence for liquid layering in the Ilímaussaq magma chamber. The distribution of the less common trace elements (Bi, Cd, Co, Ge, In, I, Hg, Sc, Se, V, W) and of the more abundant trace elements which failed to form minerals (Br, Cs, Ga, Hf, Rb, Sr) is summarised.

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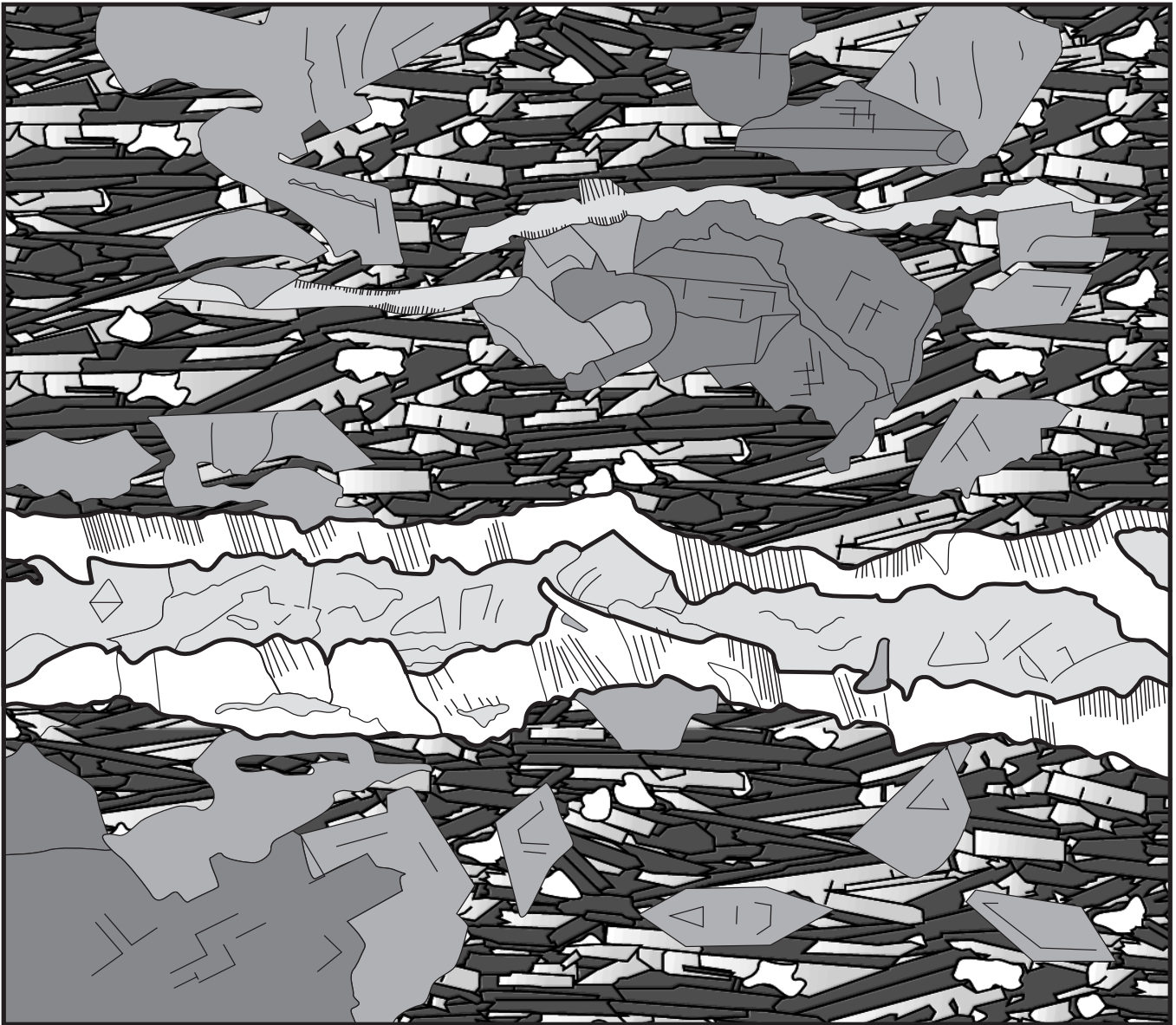
Keywords: agpaitic, alkaline rocks, cumulates, geochemistry, Greenland, Ilímaussaq, liquid layering, solubility, trace elements, zirconium

The aim of this paper is to review the main geochemical features of the Ilímaussaq complex. We emphasise studies on the dominant agpaitic rock types of the complex rather than the less voluminous pegmatites and veins. The influences of petrological processes such as extreme fractionation, crystallisation of cumulus versus intercumulus materials, and possible layering of the Ilímaussaq magma chamber are considered. This geochemical overview is largely based on work in progress and preliminary interpretations by the authors but also includes published work by a number of investigators.

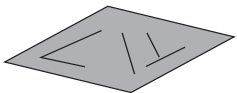
## Petrology of the Ilímaussaq complex

A summary of the geological setting and rock types of the Ilímaussaq complex is presented by Sørensen (2001, this volume) and only a brief overview of the setting and petrological evolution is given here.

The Ilímaussaq complex is one of the youngest members of the Mesoproterozoic Gardar province of South Greenland which comprises a suite of extrusive and intrusive igneous rocks produced in an extensional continental environment (Upton & Emeleus 1987). The complex was intruded into basement granitoids and the unconformably overlying sandstones and largely basaltic lavas of the Eriksfjord Formation, part of the Gardar supracrustals (Fig. 1).



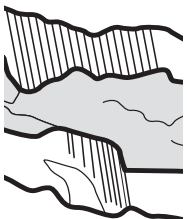
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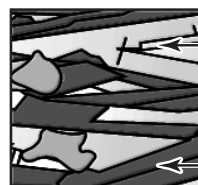
Naujakasite



Villiaumite



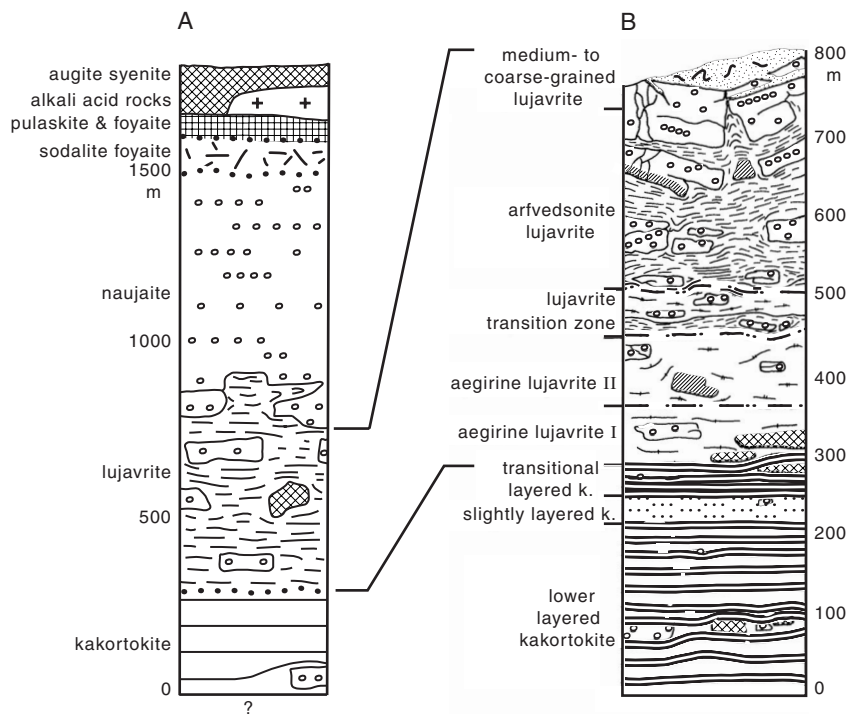
Trona  
Natrophosphate  
Trona



Microcline and albite

Arfvedsonite

Fig. 2. **A**: Schematic section through the Ilímaussaq alkaline complex. **B**: The lowermost 800 m of the complex showing the detailed igneous stratigraphy of the kakortokites and lujavrites. Modified from Andersen *et al.* (1981).



The Ilímaussaq complex was emplaced in three main phases. Phase 1 is composed of a mildly alkaline augite syenite while phase 2 consists of peralkaline quartz syenite and alkali granite. Phase 3 starts with slightly peralkaline Si-undersaturated rocks which evolve into the areally dominant, agpaite nepheline syenites of the complex (Fig. 2; Table 1). The agpaite rocks at Ilímaussaq are characterised by an agpaite index (molar  $(\text{Na}_2\text{O} + \text{K}_2\text{O})/\text{Al}_2\text{O}_3 > 1.2$  and by complex Zr-Ti silicates such as eudialyte and rinkite.

Phase 3 consists of a series of roof cumulates, and a series of floor cumulates which are overlain by an intermediate sequence of rocks whose cumulate character decreases upwards so that the later rock types are intrusive into the earlier materials. The roof rocks crystallised from the top downwards, forming the succession pulaskite, foyaite, sodalite foyaite and naujaite. Floor cumulates contemporaneous with the greater part of the roof cumulates are thought to occur below the deepest exposed levels.

The earliest exposed floor cumulates are the lower layered kakortokites which exhibit a striking macro-rhythmic layering. Each of the 29 three-layer units starts

with a black arfvedsonite-rich layer; this is followed with a few exceptions by a red eudialyte-rich layer and then a thicker white layer rich in alkali feldspar. The 29 units are numbered from -11 upwards through 0 to +17 (Bohse *et al.* 1971). Based on analyses of individual layers, we have computed a bulk kakortokite composed of 12.7% black, 11.5% red and 75.8% white layers. The lower layered kakortokites are overlain by slightly layered kakortokite and then by transitional layered kakortokite where the black, red and white layers are labelled F to A upwards (Bohse & Andersen 1981).

There is a gradual and concordant contact to the overlying aegirine lujavrite I where detailed studies have established weakly developed macrorhythmic layering (Bailey 1995). Additional stratified and more evolved lujavrites at higher levels are termed aegirine lujavrite II, lujavrite transition zone and arfvedsonite lujavrite. The final medium- to coarse-grained lujavrite (M-C lujavrite), which occurs as intrusive sheets, approaches pegmatites in its grain size and texture. These late lujavrites are associated with a uranium deposit in the roof zone at Kvanefjeld (Sørensen *et al.* 1974).

Most of the Ilímaussaq rocks are cumulates but rare chilled margins, dykes and sills thought to be close to magmatic compositions of the evolving Ilímaussaq system are now starting to be recognised and are discussed below.

Fig. 1. Simplified geological map of the Ilímaussaq complex. Modified from Ferguson (1964) and Andersen *et al.* (1988). **M-C** lujavrite: medium- to coarse-grained.

Table 1. The major rock types of the Ilímaussaq alkaline complex

Rock type	Texture	Essential minerals*	Minor minerals
Augite syenite	hypidiomorphic to xenomorphic granular, massive or layered, medium to coarse	alkali feldspar, hedenbergite, titanomagnetite, ferropargasite, olivine, biotite	ternary feldspar, apatite, pyrrhotite, nepheline
Pulaskite and foyaite	massive, medium to coarse, platy feldspars	alkali feldspar, nepheline, hedenbergite, fayalite, aegirine-augite to aegirine, katophorite	titanomagnetite, apatite, aenigmatite, biotite, fluorite, eudialyte
Sodalite foyaite	foyaitic, coarse	alkali feldspar, nepheline, sodalite, aegirine-augite to aegirine, katophorite, arfvedsonite	fayalite, hedenbergite, apatite, aenigmatite, titanomagnetite, eudialyte, rinkite, fluorite, biotite
Naujaite	poikilitic, coarse to pegmatitic	sodalite, alkali feldspar, nepheline, aegirine, arfvedsonite, eudialyte	aenigmatite, hedenbergite, aegirine-augite, fayalite, apatite, katophorite, rinkite, polythionite, biotite, sphalerite, pectolite, villiaumite, fluorite, titanomagnetite
Kakortokite	laminated, layered, medium to coarse	alkali feldspar, nepheline, aegirine, arfvedsonite, eudialyte	sodalite, aenigmatite, magnetite, rinkite, fluorite, löllingite, sphalerite, galena
Lujavrite <sup>†</sup>	laminated, fine-grained; sometimes layered or massive, medium to coarse	microcline, albite, nepheline, sodalite, analcime, naujakasite, aegirine, arfvedsonite, eudialyte,	monazite, britholite, villiaumite, sphalerite, pectolite, steenstrupine, lovozerite, vitusite, polythionite, ussingite, lueshite, neptunite
Alkali granite, quartz syenite	hypidiomorphic granular, medium to coarse	alkali feldspar, quartz, aegirine, arfvedsonite	aenigmatite, elpidite, zircon, ilmenite, pyrochlore, neptunite, fluorite, sphalerite

\* Analcime and natrolite are secondary minerals in most rocks.

† There are several types of lujavrites. Three major groups may be distinguished: *aegirine or green lujavrite* with aegirine being the dominant mafic mineral; *arfvedsonite or black lujavrite*, fine grained, often laminated with arfvedsonite as the dominant mafic mineral; *medium- to coarse-grained lujavrite (M-C lujavrite)* with arfvedsonite as the dominant mafic mineral and generally showing foyaitic textures. *Naujakasite lujavrite* is a variety of arfvedsonite lujavrite containing naujakasite instead of nepheline and with steenstrupine instead of eudialyte.

## Analytical methods

Most of the data presented here were obtained by X-ray fluorescence analysis on glass discs (major elements) or powder tablets (trace elements), and by instrumental neutron activation analysis. These methods were supplemented by optical emission spectrometry for B, delayed neutron counting for U, Cerenkov counting for Li, photometry for F, and atomic absorption spectrometry for Li, Be and Cd. Minerals

were separated by conventional methods of magnetic separation and heavy liquids.

## Agpaitic geochemistry

Representative analyses for the main rock types in the Ilímaussaq complex are given in Table 2, and analyses of rare dykes and sills thought to be close to Ilímaussaq magma compositions are given in Table 3.



The agpaitic magmas and derivative rocks possess a number of unique geochemical features. Outstanding among these is their peralkaline, highly sodic character. In the Ilímaussaq complex, this led to the appearance of a long series of Na-bearing minerals (formulae are given by Petersen 2001, this volume). They include minerals of Na–Fe (arfvedsonite, aegirine, aenigmatite), Na–Zr (eudialyte, catapleiite), Na–Nb (pyrochlore, lueshite), Na–Ti–Nb (rinkite, neptunite, astrophyllite-niobophyllite), Na–REE (steenstrupine, tundrite, joaquinite), Na–Be (chkalovite, sorensenite, leucophane) and Na–P (vitusite, vuonnemite, Na-rich apatite). Na was linked with volatile components in minerals of Na–Cl (sodalite), Na–F (villiaumite) and Na–CO<sub>2</sub> (trona and thermonatrite). K–Na even enter the sulphide djerfisherite. Many of these minerals have limited occurrences in late pegmatites, veins and pockets but sodalite, arfvedsonite, aegirine and eudialyte are important rock-forming minerals (Table 1).

In terms of trace elements, the Ilímaussaq rocks are characterised by exceptionally high levels of the normally incompatible elements Zr, Hf, Nb, Ta, REE, Th, U, Sn, Li, Be, Rb, Zn, Pb, Sb, W, Mo, As and Ga, as well as the volatile elements F, Cl, Br, I and S. Less well documented, because of analytical difficulties, are the exceptionally low levels attained by compatible trace elements such as Sc and Co (< 0.1 ppm), V, Cr and Ni (< 0.5 ppm), Cu (< 1 ppm), Ba (< 5 ppm) and Sr (< 10 ppm).

Elements which are usually coherent geochemically can exhibit bizarre relations. Thus during evolution of the complex K/Rb falls from 460 to 35, Zr/Hf rises from 45 to 97, and Cl/Br rises from 170 to 1500. Average Mg/Li ratios of basalts (c. 2700) and granites (c. 40) can be compared with averages for nepheline syenites from Lovozero (110) and Ilímaussaq (4.5). The average Mg/Li ratio for arfvedsonite can be compared at Lovozero (19) and Ilímaussaq (1.9, with individual ratios down to 0.13). In the final arfvedsonites at Ilímaussaq, Li is a minor element whereas Mg is a trace element (Bailey *et al.* 1993). It should be emphasised that these features apply to rocks and minerals occurring in outcrops over hundreds or even thousands of metres and not to pockets of exotic pegmatite.

The extraction of a series of cumulus phases characteristic of agpaitic nepheline syenites (Table 1) has imposed distinctive element associations on the succession of cumulate rocks. In several of these cumulus phases, elements which are normally considered incompatible, e.g. during basalt fractionation, are strongly compatible and this leads to a sharp decrease in their contents in the succeeding residual magmas.

Thus the build-up of Cl, which occurs in residual phonolitic magmas at many localities, reaches a climax at Ilímaussaq with the voluminous formation of flotation cumulates of sodalite. These flotation cumulates form naujaite, the final rock type of the roof series. Chlorine contents of naujaite typically are in the range 2–3.5 wt% with an extreme value of 4.6 wt%. The earliest sodalites are particularly rich in Br, I and B and this leads to decreasing levels in subsequent rocks.

In the earliest exposed floor cumulates, the kakortokites, the Na–Zr silicate eudialyte is an important cumulus phase, particularly in the eudialyte-rich red kakortokite layers. Eudialyte not only strongly partitions Zr but also Hf, Nb, Ta, W and As so that all these elements show decreasing contents in the subsequent rocks. Moderate partitioning of Ca, Sr and Ba, and of Cl and Br, accelerates the already dwindling amounts of these elements. Even the rising levels of REE and Pb in the Ilímaussaq system receive a short-lived setback as a result of their significant incorporation in the abundant eudialyte of the kakortokites.

Also at the kakortokite stage, cumulus arfvedsonite and aegirine scavenge large amounts of the rare transition metals Co, Sc, V and Cr. Levels of Sc, for example, fall to < 0.1 ppm in the subsequent rocks.

Fluorite is a cumulus phase in the kakortokites and its fractionation leads to a reduction in F levels so that contents only recover to > 1000 ppm in the final lujavrites with the crystallisation of villiaumite.

Clearly, a number of elements which are normally incompatible in alkaline magma series (Cl, Zr, Hf, W) pass through maxima in the Ilímaussaq agpaitic system and may then evolve to significantly lower levels. Thus in the final medium- to coarse-grained lujavrite, Zr may fall to 328 ppm and Hf to 2.6 ppm. This allows new associations of minerals and elements to be established: instead of Th and U substituting in eudialyte, they concentrate in the Th-REE silicophosphate steenstrupine.

Agpaitic cumulate rocks, and presumably their parental melts, solidify at remarkably low magmatic temperatures, probably < 500°C (Sood & Edgar 1970). They may even exhibit a continuous transition through hyper-agpaitic melts to hydrothermal solutions (Khomyakov 1995). Such behaviour suggests that fractionation proceeds to an extremely high degree. The recognition of a rare suite of dykes and sills corresponding to Ilímaussaq liquids (Table 3) has allowed us to quantify this fractionation. During most though not all of the evolution, Th, U, La, Cs, Li, Be and Pb behaved as almost perfectly incompatible elements.

Table 2. Representative analyses of Ilimaussaq rock types

	Phase 1		Phase 2		Phase 3		
	fine augite syenite	coarse augite syenite	quartz syenite	alkali granite	pulaskite	foyaite	sodalite foyaite
GGU no.	154361	154334	152118	152116	154387	154384	154351
SiO <sub>2</sub> (wt%)	50.78	56.97	70.17	71.01	62.54	58.50	48.24
TiO <sub>2</sub>	2.76	1.27	0.26	0.23	0.10	0.32	0.33
ZrO <sub>2</sub>	0.02	0.06	0.19	0.46	0.21	0.27	0.51
Al <sub>2</sub> O <sub>3</sub>	13.69	16.82	10.03	8.65	16.97	16.21	18.99
Fe <sub>2</sub> O <sub>3</sub>	2.78	1.47	3.03	4.33	2.02	3.03	4.16
FeO	11.69	6.68	4.67	3.79	1.48	3.80	3.08
MnO	0.35	0.22	0.18	0.21	0.12	0.19	0.20
MgO	2.32	0.76	0.11	0.18	0.15	0.11	0.12
CaO	5.12	3.47	0.38	0.24	1.53	1.76	1.67
Na <sub>2</sub> O	3.84	5.65	5.28	4.67	8.19	7.56	14.87
K <sub>2</sub> O	4.16	5.16	4.25	3.91	4.69	5.64	3.47
P <sub>2</sub> O <sub>5</sub>	1.22	0.34	0.02	0.07	0.05	0.04	0.06
H <sub>2</sub> O <sup>+</sup>	0.36	0.56	0.34	0.28	0.83	1.26	1.24
H <sub>2</sub> O <sup>-</sup>	0.24	0.19	0.16	0.26	0.20	0.21	0.22
CO <sub>2</sub>	0.07	0.05	0.10	0.09	0.06	0.04	0.10
S	0.16	0.07	0.00	0.00	0.01	0.02	0.07
Cl	0.02	0.04	0.02	0.01	0.02	0.12	2.34
F	0.16	0.14	0.35	0.27	0.20	0.20	0.17
others	0.05	0.06	0.19	0.47	0.18	0.17	0.40
	99.79	99.98	99.35	99.12	99.55	99.45	100.24
- O	0.16	0.10	0.15	0.12	0.09	0.12	0.66
	99.63	99.88	99.20	99.00	99.46	99.33	99.58
A.I.	0.80	0.88	1.32	1.38	1.09	1.14	1.49
Cs (ppm)	< 0.1	1.2	0.35	1.2	2.3	5.3	6.3
Rb	66	68	438	557	234	315	348
Tl	< 0.5	< 0.5	2.3	0.5	1.0	1.3	1.7
Ba	2580	2320	16	48	30	42	10
Pb	12	15	69	163	36	45	89
Sr	281	395	15	54	16	27	11
La	73	77	252	537	223	244	619
Ce	163	163	510	1110	445	512	1420
Nd	83	76	205	423	185	219	572
Sm	15.0	13.9	31.2	65.6	36.3	38.2	95.9
Eu	5.08	4.53	2.13	4.16	3.08	3.60	10.1
Tb	2.06	1.88	5.73	12.1	5.89	5.82	16.6
Yb	5.21	5.30	18.5	36.8	24.7	17.9	50.9
Lu	0.68	0.80	2.80	5.38	3.69	2.43	6.42
Y	50	45	187	356	240	184	488
Th	3.4	7.9	48.5	123	23.5	27.8	54.0
U	0.55	1.9	15.6	44.1	8.9	9.8	15.5
Zr	208	272	1790	4560	2790	2070	4110
Hf	5.8	11.4	38.0	108	52.6	42.5	98.9
Sn	7.3	12	38	88	25	27	62
Nb	55	93	312	912	386	325	777
Ta	3.9	6.0	18.7	56.6	21.3	19.2	48.4
Li	16	80	182	156	91	132	161
Zn	148	117	402	1260	182	276	534
Cu	26	16	5	6	5	10	8
Co	10	3.8	1.1	-	1.1	3.6	0.32
Ni	< 0.5	< 0.5	< 0.5	< 0.5	3.1	0.5	< 0.5
Sc	40	18	1.9	1.2	0.21	0.52	0.09
V	< 0.5	< 0.5	2.1	1.6	1.5	< 0.5	< 0.5
Cr	8.2	1.8	3.5	3.9	1.1	1.8	1.9
Ga	23	29	41	37	56	58	66
Ge	1.9	1.9	1.8	2.1	1.3	1.4	1.6
B	< 3	8	13	13	7	5	3
Be	1.5	3.5	24	59	15	16	32
Sb	< 0.1	< 0.1	0.2	0.9	0.2	0.1	< 0.5
Mo	4.1	4.4	2	8.4	< 0.5	< 0.5	10
As	0.7	3.3	2.8	14	4.3	2.8	9.9
F	1560	1350	3550	2740	2040	1960	1730
Cl	210	450	190	120	160	1200	23400
Br	< 0.5	1.5	0.5	< 0.5	0.8	2.9	117
I	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	1.8
S	1570	690	45	40	105	160	730

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Table 1. The major rock types of the Ilímaussaq alkaline complex

Rock type	Texture	Cumulus minerals	Other minerals
<i>Phase 1</i>			
augite syenite	hypidiomorphic to xenomorphic granular, massive or layered, medium to coarse	alkali feldspar, hedenbergite, olivine, titanomagnetite, apatite, plagioclase, pyrrhotite, chalcopyrite	ferropargasite, biotite, nepheline
<i>Phase 2</i>			
alkali granite, quartz syenite	hypidiomorphic granular, medium to coarse		alkali feldspar, quartz, arfvedsonite, aegirine, aenigmatite, ilmenite, sphalerite, galena, pyrochlore, fluorite, zircon
<i>Phase 3</i>			
pulaskite and foyaite	massive, medium to coarse, platy feldspars	alkali feldspar, hedenbergite, fayalite, titanomagnetite, apatite	katophorite-arfvedsonite, aegirine-augite to aegirine, nepheline, aenigmatite, biotite, eudialyte, fluorite, analcime, natrolite
<i>Agpaitic nepheline syenites</i>			
sodalite foyaite	foyaitic, coarse	alkali feldspar, sodalite, nepheline, hedenbergite, fayalite, titanomagnetite, apatite	katophorite-arfvedsonite, aegirine, eudialyte, aenigmatite, analcime, natrolite
naujaite	poikilitic, coarse to pegmatitic	sodalite, alkali feldspar, nepheline, hedenbergite, fayalite, titanomagnetite,	katophorite-arfvedsonite, aegirine, eudialyte, aenigmatite, analcime, apatite, natrolite, fluorite, villiaumite, rinkite, sphalerite
kakortokite	laminated, layered, medium to coarse	alkali feldspar, nepheline, arfvedsonite, aegirine, eudialyte, sodalite, fluorite, aenigmatite	analcime, natrolite, rinkite, sphalerite, galena, löllingite
lujavrite †	laminated, fine-grained; sometimes layered or massive, medium to coarse	aegirine, arfvedsonite, microcline, albite, nepheline, eudialyte	analcime, natrolite, sodalite, naujakasite, steenstrupine, vitusite, monazite, villiaumite, sphalerite, galena

† See Sørensen (2001, Table 1) for further information.

Table 3. Analyses of rocks representing Ilímaussaq liquids

	1 augite syenite, chilled margin	2 evolved augite syenite, av. of 3	3 Cl-poor sodalite foyaite, av. of 3	4 Fe-rich phonolite dyke	5 Fe-rich phonolite dyke	6 aegirine lujavrite av. of 3	7 arfvedsonite lujavrite av. of 2	8 medium- to coarse-grained lujavrite weighted av.
SiO <sub>2</sub> (wt%)	53.24	62.33	51.01	51.83	52.27	52.38	52.25	52.71
TiO <sub>2</sub>	2.44	0.50	0.34	0.55	0.46	0.22	0.23	0.35
ZrO <sub>2</sub>	0.04	0.11	0.36	0.55	0.78	0.95	0.25	0.13
Al <sub>2</sub> O <sub>3</sub>	14.79	15.65	17.38	14.57	14.09	13.20	12.23	13.29
Fe <sub>2</sub> O <sub>3</sub>	2.64	2.12	4.73	7.56	11.03	10.90	6.06	4.04
FeO	8.66	3.54	4.62	4.61	1.15	1.96	8.72	8.21
MnO	0.24	0.16	0.25	0.48	0.30	0.37	0.64	0.60
MgO	1.60	0.31	0.13	0.14	0.07	0.10	0.12	0.12
CaO	4.94	1.69	1.97	2.54	2.06	1.20	0.27	0.30
Na <sub>2</sub> O	4.68	6.74	10.08	8.81	9.27	10.72	9.25	9.20
K <sub>2</sub> O	4.26	5.53	3.93	4.87	4.12	2.82	3.23	4.69
P <sub>2</sub> O <sub>5</sub>	0.74	0.08	0.05	0.08	0.09	0.18	0.54	0.41
H <sub>2</sub> O <sup>+</sup>	0.29 <sup>†</sup>	0.68	4.19	1.56 <sup>†</sup>	1.18 <sup>†</sup>	3.44	3.65	3.70
H <sub>2</sub> O <sup>-</sup>	0.19	0.20	0.16	0.47	0.36	0.28	0.24	0.13
CO <sub>2</sub>		0.23	0.07			0.09	0.19	0.07
S	0.15	0.00	0.02	0.12	0.03	0.06	0.06	0.14
Cl	0.03	0.01	0.09	0.33	0.08	0.05	0.03	0.05
F	0.10	0.15	0.41	0.84	1.80	0.08	0.14	0.16
others	0.46	0.08	0.31	0.42	0.71	0.89	1.64	1.47
	98.99	100.11	100.10	100.33	99.85	99.89	99.74	99.77
- O	0.12	0.07	0.20	0.49	0.79	0.07	0.10	0.15
	98.87	100.04	99.90	99.84	99.06	99.82	99.64	99.62
A.I.	0.83	1.09	1.20	1.36	1.40	1.57	1.53	1.52
FeO*	11.04	5.45	8.88	11.41	11.08	11.77	14.17	11.85
Zr (ppm)	284	836	2690	4040	5740	7010	1860	939

Analysts: J. Kystol, J.C. Bailey and R. Fuge.

<sup>†</sup> Loss on ignition corrected for other volatiles where known.

A.I.: Apgaitic index, (Na<sub>2</sub>O+K<sub>2</sub>O)/Al<sub>2</sub>O<sub>3</sub> mol.

FeO\*: Total Fe as FeO.

1: GGU 153394 (new analysis of sample U-106 from Ussing's collection in the Geological Museum, Copenhagen).

2: Average of GGU 152122, 152130, 154378.

3: Average of GGU 57070, 154303, 154347.

4: Sample GGU 42475 (Larsen & Steenfelt 1974), new analysis.

5: Sample ARM 62/8010 (Martin 1985). New analysis.

6: Average of GGU 152128, 154302, 66143.

7: Average of GGU 152127, 154363.

8: 1:1 average of (a) 3 large surface samples (GGU 154397, 154399, 154724) and (b) 21 smaller samples from drill cores.

GGU prefixed to sample numbers: samples in the collections of the Geological Survey of Denmark and Greenland.



Based on their increasing contents, it was calculated that the final rock type, medium- to coarse-grained lujavrite, was formed after 99% crystallisation of the augite syenite.

Agpaitic melts are relatively depolymerised with a high ratio of non-bridging oxygens to tetrahedrally co-ordinated cations (Larsen & Sørensen 1987; Ewart & Griffin 1994), and possess enhanced solubility for high-field-strength elements such as Zr (Watson 1979; Farges *et al.* 1991; Hess 1991). Worldwide, contents of Zr in nepheline syenites (and phonolites) increase from around 50 (250) ppm to 1400 (1750) ppm as their agpaitic index increases from 0.8 to 1.1 (Fig. 3). Only two phonolitic series are known to us with significantly higher agpaitic indices. Firstly, agpaitic phonolites in the Igaliko dyke swarm (Pearce 1988), 25 km NE of the Ilímaussaq complex, range up to 6250 ppm Zr with a somewhat aberrant dyke possibly extending the trend to 7600 ppm. Secondly, the rare phonolitic dykes and sills in and around the Ilímaussaq complex show Zr contents rising to 8700 ppm (Fig. 3).

In the foyaite at Ilímaussaq, combining the maximum Zr content in the (001) sector of interstitial aegirine (1.33%, Larsen 1976) with the Zr partition coefficient appropriate to this sector (*c.* 1.33, Wolff & Toney 1993) suggests a saturation level of *c.* 1.0% Zr. The same calculation method yields saturation levels of 0.89% Zr in sodalite foyaite and 0.85% Zr in naujaite. While such Zr contents may only reflect metastable crystallisation in anomalous interstitial micropockets, they are in broad agreement with the highest value so far reported from dykes and sills.

Zirconium contents around 9000 ppm are probably close to the solubility limit for Zr at Ilímaussaq since higher Zr whole-rock contents are only found in cumulates rich in eudialyte. Fractionation of eudialyte produced late lujavritic liquids at Ilímaussaq with Zr contents falling to < 500 ppm. The rise and fall of Zr contents at Ilímaussaq is confirmed by analyses of both cumulus and intercumulus pyroxenes (Larsen 1976).

Enhanced solubility is also evident for (1) F prior to the crystallisation of fluorite, e.g. contents of 0.84% and 1.80% F in two phonolite dykes; (2) P in late lujavrites, e.g. up to 0.39% in arfvedsonite lujavrite and up to 0.40% in medium- to coarse-grained lujavrite; (3) REE + Y in late lujavrites, e.g. up to 1.1% in arfvedsonite lujavrite and up to 1.5% in medium- to coarse-grained lujavrite.

Components which are normally volatile or prone to escape in hydrothermal solutions from many magmas were soluble to a high degree and were largely

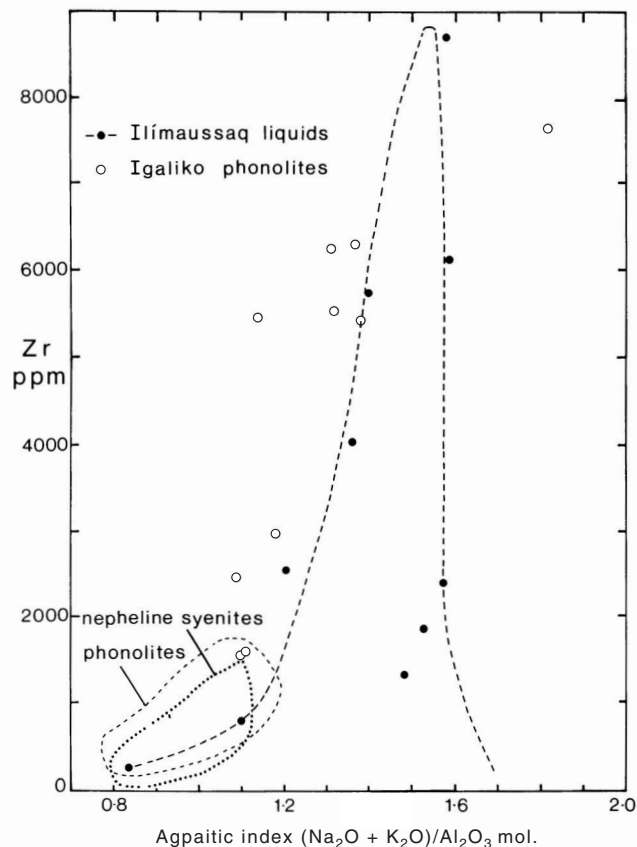


Fig. 3. Zr contents versus agpaitic index showing the main fields for nepheline syenites and phonolites (authors' compilation of 210 localities), the agpaitic phonolite series in the Igaliko dyke swarm (Pearce 1988; new analyses by the authors), and the proposed trend for Ilímaussaq liquids (Table 3).

retained by the agpaitic magmas at Ilímaussaq, e.g. H<sub>2</sub>O, Cl, Br, I, S, Mo, As, Tl, B and Cs (cf. Kogarko 1974). Fugacities of oxygen, water and sulphur are inferred to be low, even though their contents are high (Larsen 1976; Karup-Møller 1978; Konnerup-Madsen & Rose-Hansen 1982; Larsen & Sørensen 1987).

The geochemistry of the proposed magma compositions at Ilímaussaq has enabled us to estimate bulk partition coefficients for all studied elements during the main fractionation stages of the complex and to integrate these coefficients with the geochemistry of the various cumulus phases. Some distortion, however, results from the fact that these phases have often modified their chemistry during continued growth at the intercumulus stage.

Such modelling has allowed us, for example, to recognise that, during the early stages at Ilímaussaq, P behaved as a compatible element and was fractionated in apatite. However, after crystallisation

of the earliest agpaitic nepheline syenite, the sodalite foyaite, bulk partition coefficients for P fell to  $< 0.03$  and the build-up of P contents in the late lujavrites, which led to the appearance of the phosphates and silico-phosphates vitusite, monazite, steenstrupine and vüonne-mite, can be explained solely by fractional crystallisation.

It is widely recognised that fractionation of magmas is attended by a number of characteristic geochemical trends. Thus K/Rb ratios normally decrease and this trend is also present in the Ilímaussaq rocks with ratios falling from around 500 to 35. Other well-established trends, however, such as the fall in Ba/Rb ratios show reversals at Ilímaussaq. Thus the Ba/Rb ratio sharply rises in the kakortokitic rocks; this can be attributed to the high Ba/Rb ratio of cumulus eudialyte (*c.* 10) compared to Ilímaussaq magmas at this stage (*c.* 0.1). In general, as noted by Gerasimovsky (1969) and Ferguson (1970), there is a good correlation between fractionation trends and the chronology inferred from field relations. In addition, the reversals in the trends of trace element ratios noted by Ferguson (1970) can now be explained on the basis of cumulus processes and mineral geochemistry.

Comparing averages for agpaitic nepheline syenites from various localities, it is found that the Ilímaussaq rocks have lower K/Rb, K/Cs and Sr/Ca but higher Li/Mg, Zn/Fe and Ga/Al ratios than similar materials from Lovozero (Kola Peninsula) and Pilanesberg (South Africa) (Bailey *et al.* 1978). It can thus be inferred that the Ilímaussaq rocks arose from a more extreme fractionation history. This is also reflected in the fact that hyper-agpaitic conditions are not restricted to pegmatites and veins, as at Lovozero and Khibina (Khomyakov 1995), but are already present in the late lujavrites at Ilímaussaq (Sørensen 1997; Sørensen & Larsen 2001, this volume).

Other geochemical ratios, however, are more indicative of the differing parent magmas at these localities. It is characteristic for the more Si-undersaturated basalts that they possess lower Zr/Nb ratios but higher ratios of LREE/HREE (light/heavy REE), e.g. La/Lu. In the Gardar province, early- and mid-Gardar basalts have Zr/Nb ratios around 18 (Upton & Emeleus 1987) whereas among the late-Gardar Tugtutôq–Ilímaussaq dykes two populations of basalt with Zr/Nb ratios of *c.* 6.4 and 3.9 have been distinguished (Pearce 1988). Transitional basalts with Zr/Nb of *c.* 6.4 evolve to Si-oversaturated compositions. In contrast, the more Si-undersaturated suite has Zr/Nb ratios which are relatively constant in basanite/tephrite (3.6), basalt (2.9), benmoreite (3.8) and phonolite (3.5). These can be

matched by Zr/Nb ratios from Ilímaussaq in the chilled margin to the Ilímaussaq augite syenite (3.7) which is equivalent to a benmoreite.

Among the agpaitic nepheline syenites from different localities, one can compare lujavrites with a similar degree of differentiation (Bailey *et al.* 1978). Zr/Nb ratios for lujavrites at Ilímaussaq (16) are higher than for lujavrites from Lovozero (10) and Pilanesberg (11) whereas La/Lu ratios in lujavrites at Ilímaussaq (170) are lower than for lujavrites from Lovozero (270) and Pilanesberg (280). These features suggest that the nepheline syenites at Lovozero and Pilanesberg were derived from more Si-undersaturated parental magmas than those from Ilímaussaq. Kogarko (1979) reached the same conclusion on the basis of lower Sr/Ca ratios and more pronounced negative Eu anomalies in the REE patterns at Ilímaussaq compared to those at Lovozero. She concluded that the Ilímaussaq nepheline syenites were derived from nepheline-normative alkali basalts whereas the Lovozero and Khibina nepheline syenites were derived from melilite nephelinites. At Pilanesberg, the high Sr/Ca ratios, subdued Eu anomalies, and association with kimberlite and carbonatite (Ferguson 1973), suggest that the nepheline syenites there were also derived from highly Si-undersaturated magmas.

## Rare-earth elements

The main features of REE geochemistry at Ilímaussaq were presented in earlier work by Gerasimovsky (1969), Semenov (1969), Fryer & Edgar (1977), Bailey *et al.* (1978) and Sørensen (1992). Contents of REE are high, reaching maxima in the eudialyte-rich red kakortokites and in the final lujavrites where a number of REE minerals including steenstrupine and vitusite have crystallised. With evolution in the Ilímaussaq system, REE spectra reveal (1) a general increase in total REE, (2) high LREE/HREE (light/heavy REE) ratios which increase sharply in the lujavrites and (3) a more or less uniformly negative Eu anomaly. The only rocks lacking a negative Eu anomaly are some of the earliest augite syenites which still preserve plagioclase cores within their alkali feldspars.

In the earlier agpaitic rocks – sodalite foyaite, naujaite and kakortokites – REE contents correlate with Zr, and REE are mainly located in eudialyte. Total contents of RE<sub>2</sub>O<sub>3</sub> (including Y<sub>2</sub>O<sub>3</sub>) in eudialyte increase fairly regularly from about 1.7 wt% in the early roof rocks (sodalite foyaite) to 8.7 wt% in the late, more

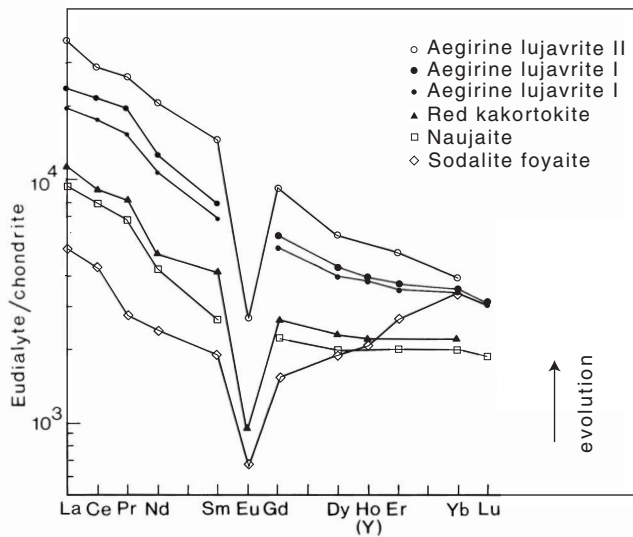


Fig. 4. REE patterns for eudialytes from the Ilímaussaq complex. Sources of data: Aegirine lujavrite II, GGU 77018 (Gerasimovsky 1969); Aegirine lujavrite I, drill core VII (GGU 108107) at 96 m (larger point) and at 146 m (smaller point) (Bailey 1995); Red kakortokite, GGU 77045 (Gerasimovsky 1969); Naujaite, drill core VII (GGU 108107) at 34 m (Bailey 1995); Sodalite foyaite, GGU 138761 (Fryer & Edgar 1977).

evolved aegirine lujavrite (Fig. 4). In this sequence,  $(La/Yb)_N$  (chondrite normalised) ratios also increase from about 1.5 to 9.4; this is largely due to an increase in  $(Gd/Yb)_N$  ratios from 0.45 to 2.3 since  $(La/Sm)_N$  ratios show no clearly defined trend. The REE patterns of eudialyte tend to show an increasing slope for HREE in the early rocks but a decreasing slope in the later rocks. Europium anomalies, as measured by  $Eu/Eu^*$  ratios, typically scatter around 0.2–0.4 but show no clear trend with evolution. The Ilímaussaq eudialytes cover most, though not all, of the REE range of the eudialyte group (Johnsen & Gault 1997). The huge tonnages, and relatively high contents of HREE compared to presently exploited REE sources, bastnaesite and monazite, mean that Ilímaussaq eudialyte has the potential to be an important future resource of REE (Sørensen 1992).

The distribution of REE between co-existing minerals can be illustrated by a sample of aegirine lujavrite I (Fig. 5). Eudialyte dominates the REE budget. The REE patterns of all minerals are characterised by LREE enrichment and a negative Eu anomaly. Compared to the bulk rock, the eudialyte REE pattern is slightly enriched in MREE (middle REE) and Eu indicating the preferential entry of Ca–Sr–Eu into this phase. Relative to the bulk rock, REE patterns of the remain-

ing minerals are slightly depleted in MREE and the negative Eu anomaly is more pronounced; the  $La/Yb_N$  ratio is similar in aegirine, higher in microcline + nepheline and analcime + natrolite, but lower in arfvedsonite. These features are broadly consistent with preferential entry of LREE into an optimal site of about 1.10 Å in feldspars, and of HREE into an optimal site of about 0.70 Å in pyroxenes and amphiboles (Jensen 1973 but with revised ionic radii from Shannon 1976).

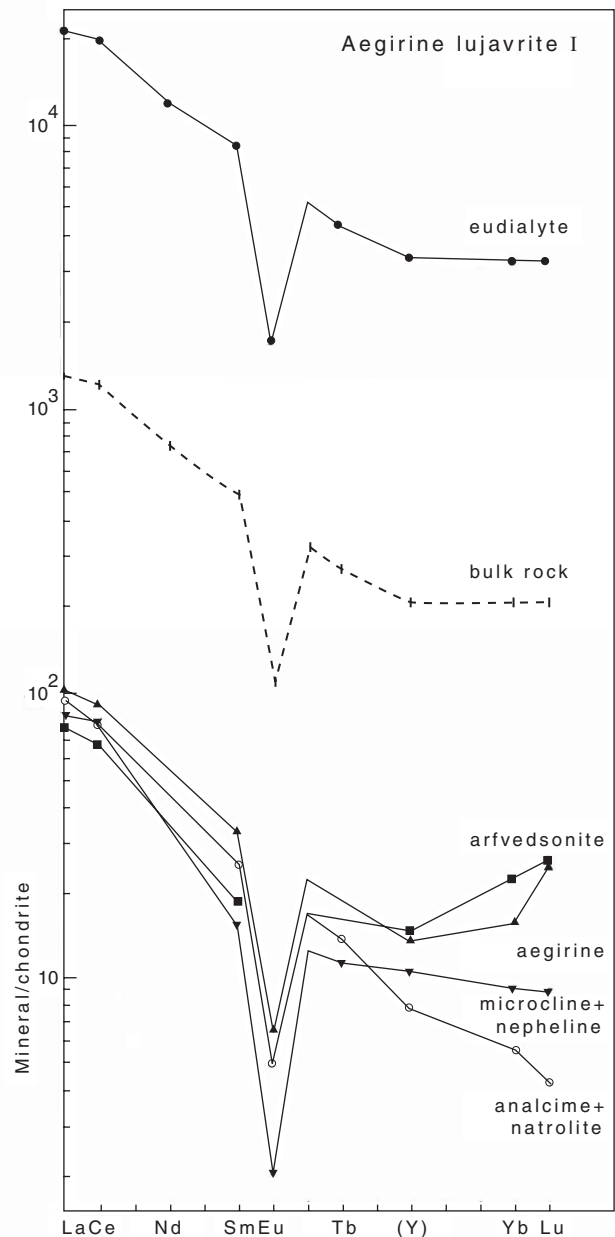


Fig. 5. REE patterns for bulk rock and separated minerals from aegirine lujavrite I, drill core VII (GGU 108107) at 191 m (Bailey 1995).

## Cumulate rocks and processes

Ever since the memoir of Ussing (1912), it has been realised that the cumulitic character of many Ilímausaq rock types has a profound effect on their geochemistry. This is most obviously seen in the exceptionally high contents of Cl in sodalite cumulates (up to 4.6% in naujaite), Zr, Hf and Nb in eudialyte cumulates (up to 3.4%, 0.11% and 0.33%, respectively, in red kakortokite), Fe and Zn in arfvedsonite cumulates (up to 20.3% and 0.082%, respectively, in black kakortokite) and F in fluorite cumulates (up to 2.1% in black kakortokite).

Varying amounts of intercumulus material, i.e. trapped liquid, and varying amounts of additional growth on the cumulus minerals are also significant. For example, in aegirine lujavrite I – which is considered an orthocumulate with virtually no adcumulus growth – Li is dominantly concentrated in the intercumulus material which varies from 25 to 48 wt% (or roughly 30 to 55 vol.%) leading to a variation in Li contents from 80 to 260 ppm (Bailey & Gwozdz 1994).

Because eudialyte more readily accepts U than Th, it has a lower Th/U ratio (*c.* 0.3–0.6) than the magmas from which it crystallises (*c.* 3). Accordingly, in cumulate rocks where additional growth of eudialyte from trapped intercumulus liquid occurs, there appears to be a build-up of Th/U ratios in the shrinking volume of intercumulus liquid. Some of this material is thought to crystallise at a late stage as autometasomatic alteration products on eudialyte and other phases. This non-

isomorphous Th and U rarely seems to escape from whole-rock samples even though it is weakly bound in mineral lattices; in consequence it can be leached out of rock powders by a weak leaching agent, a 2% ammonium oxalate solution (Bailey *et al.* 1983).

In the lower layered kakortokites, each white layer accumulated after the underlying, more densely packed, black and red layers and thus has a higher proportion of intercumulus liquid. This has led to relatively high contents of U despite only moderate levels of the main U carrier (eudialyte; Bailey *et al.* 1981), and relatively high levels of Li and Zn despite low contents of the main Li and Zn carrier (arfvedsonite). Clearly, the intercumulus liquid had high contents of U, Li and Zn. In these kakortokites, the percentage of Zn leachable in dilute hydrochloric acid, i.e. the Zn held by sphalerite rather than silicates, increases from 7% in black to 32% in red and 45% in white kakortokite. This suggests that increasing amounts of Zn crystallised as sphalerite from the enhanced amounts of intercumulus liquid in the weakly packed white layers. This is in agreement with the greater amounts of sphalerite separated from white kakortokite (Karup-Møller 1978).

## A layered magma chamber

Geochemical evidence is accumulating that the exposed Ilímausaq rocks of phase 3 crystallised from a layered magma chamber. A number of geochemical

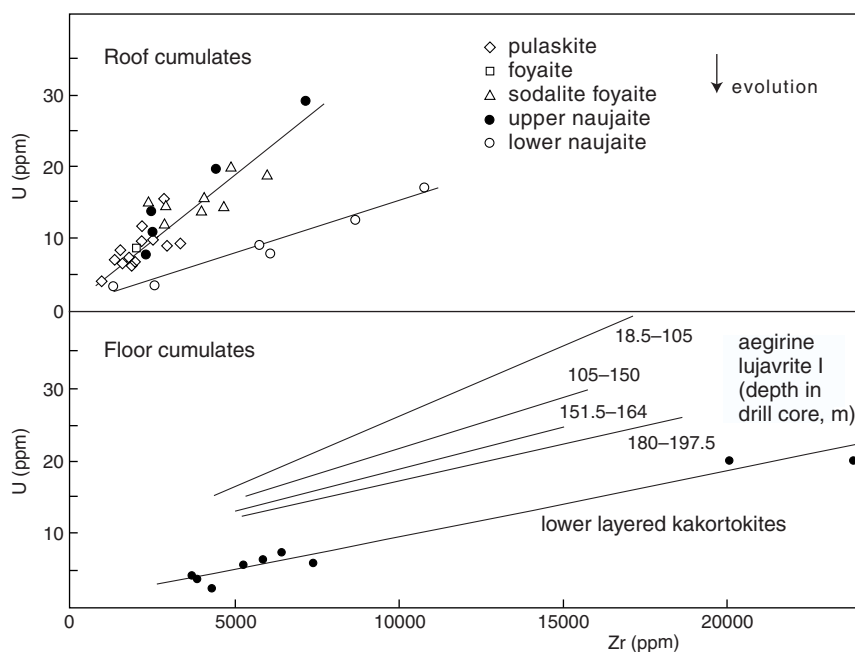


Fig. 6. Zr versus U contents for selected samples of roof and floor cumulates in the Ilímausaq complex. Data for aegirine lujavrite I in drill core VII (GGU 108107) are from Bailey (1995) but the 200 data points are not shown. The compositional arrays are largely independent of the macrorhythmic layering and are considered to indicate the presence of liquid layering in the Ilímausaq magma chamber (see text).

parameters shift in a step-like pattern with the steps occurring independently of the boundaries between the various rock types. Thus in the roof cumulates of phase 3, the pulaskite, foyaite, sodalite foyaite and upper layers of naujaite fall along a single array on a Zr–U diagram (Fig. 6). The lower and younger layers of naujaite, however, plot along a different array which has lower U contents – around three times lower – for a given Zr content.

In the floor cumulates which crystallised later, the black and red kakortokites plot along an array with even lower U contents though intercumulus liquids which are concentrated in white layers (see above) are more uraniferous. Floor cumulates from a higher, more evolved stratigraphic level – aegirine lujavrite I – are characterised by higher U contents and by several Zr–U compositional arrays at well-defined stratigraphic intervals. All samples on a given array must be controlled by a chemically uniform eudialyte – the dominant carrier of Zr and U – and the shift to the next line is attributed to the onset of crystallisation in the next chemically distinct double-diffusive liquid layer (Bailey 1995). The shifts occur independently of the macrorhythmic layering in aegirine lujavrite I.

Later lujavrites at higher stratigraphic levels are richer in U and culminate in the Kvanefjeld uranium deposit lying immediately beneath the roof of the complex (Sørensen *et al.* 1974; Kunzendorf *et al.* 1982).

Work is in progress to confirm and to establish more precisely the nature of liquid layering in the Ilímaussaq complex.

The conclusion from these Zr–U relations at Ilímaussaq is that U-rich magmas were restricted to the early uppermost layers of the roof cumulates and the late uppermost layers of the floor cumulates (Fig. 6). This is consistent with heat-flow measurements suggesting that the highly radioactive part of the complex is thin (*c.* 1 km) and the underlying rocks contain very little K, U and Th (Sass *et al.* 1972).

## Geochemical notes on selected elements

This geochemical overview would not be complete without summarising new information on the less common trace elements (Bi, Cd, Co, Ge, In, I, Hg, Sc, Se, V, W). In addition, given the mineralogical emphasis of the present work, we outline the geochemistry of elements whose contents exceed 100 ppm in certain rocks but which nevertheless failed to form minerals

in the Ilímaussaq complex (Br, Cs, Ga, Hf, Rb, Sr). *Bismuth.* Both chalcophile and lithophile tendencies are apparent at Ilímaussaq. Among sulphides, galena from a vein contained 235 ppm Bi; sphalerite from medium- to coarse-grained lujavrite contained 15 ppm and two vein sphalerites contained 15 and 20 ppm. In cuprostibite, we found 34 ppm Bi. In silicates, Bi was located in the REE–Th mineral steenstrupine where it is in the range 7–14 ppm in pegmatites and 6–17 ppm in veins. We failed to detect Bi in minerals of Nb–REE (pyrochlore < 3 ppm, rinkite < 4 ppm), Nb (nenadkevichite < 1 ppm) and Zr (eudialyte < 1 ppm). These few results suggest that Bi accumulates in the sulphides and steenstrupine of late pegmatites and veins where it may locally amount to 1–2 ppm.

*Bromine.* Median Br contents increase from around 0.5 ppm in augite syenite through 0.6 ppm in pulaskite + foyaite and 17 ppm in sodalite foyaite to 57 ppm in naujaite. Thereafter contents fall in bulk kakortokite (1.1 ppm) through aegirine lujavrite (0.5 ppm) to < 0.5 ppm in arfvedsonite lujavrite and medium- to coarse-grained lujavrite. Cl/Br ratios are around 340 in the early stages at Ilímaussaq but fall to around 170 when crystallisation of cumulus sodalite (Cl/Br 170) starts in sodalite foyaite and naujaite. Fractionation of sodalite causes a rapid depletion in Br so that the final naujaites and all subsequent rock types have higher ratios of roughly 400–1500.

Whole-rock trends are repeated in eudialytes with Cl (and Br) contents increasing from 1.1% (12 ppm) to 1.7% (40 ppm) and then decreasing to 0.6% (3 ppm). Br contents of sodalite fall from around 340 ppm to 100 ppm.

*Cadmium.* Preliminary analyses by atomic absorption on Ilímaussaq rocks suggest they contain around 0.15 ppm Cd rising to 2 ppm in the most Zn-rich samples. Sphalerite from aegirine lujavrite and medium- to coarse-grained lujavrite contains 20 and 50 ppm Cd, respectively (Karup-Møller 1978).

*Caesium.* Median Cs contents increase from 0.9 ppm in augite syenite through 6.0 ppm in pulaskite + foyaite, 5.0 ppm in sodalite foyaite + naujaite, 6.6 ppm in bulk kakortokite, 7.9 ppm in aegirine lujavrite and 8.7 ppm in arfvedsonite lujavrite to 72 ppm in medium- to coarse-grained lujavrite.

In aegirine lujavrite I, a sample from drill core VII (GGU 108107) at 191 m depth contained Cs in analcime + natrolite (42 ppm), microcline + nepheline (1.8 ppm)



and arfvedsonite (1.7 ppm) but Cs was undetected in aegirine and eudialyte. Contributions from these minerals sum to 9.3 ppm and indicate that *c.* 90% of the Cs is held in the interstitial zeolites (Bailey 1995). Separates of analcime + natrolite from 11 different depths in this drill core showed a wide range of Cs contents from 2.2 to 44 ppm. Enhanced contents of Cs (6.1–16 ppm) were found in zeolite-rich hydrothermal coatings scraped from fracture planes.

Contents of Cs in 10 samples of medium- to coarse-grained lujavrite from Kvanefjeld drill cores increased at shallow depths. Averages for Cs (and Rb/Cs) changed from 8.3 ppm (154) at 100–150 m depth to 72 ppm (17) at 50–100 m depth and 231 ppm (4.0) at < 50 m depth. Thus samples at 100–150 m depth have Rb/Cs ratios above typical magmatic values at Ilímaussaq and appear to have lost Cs. At shallow depths, Rb/Cs ratios fall below these magmatic values and contents of H<sub>2</sub>O<sup>+</sup>, Th and U (but not K and Rb) increase. This suggests that medium- to coarse-grained lujavrite has undergone Cs-rich zeolitisation in the Kvanefjeld roof zone.

Many veins are likely to be rich in Cs since samples of the widespread phase analcime contain 200–250 ppm Cs. Lesser amounts of Cs occur in pyrochlore (42 ppm), lueshite (13 ppm), neptunite (8.1 ppm), natrolite (< 0.2–7.3 ppm), tetranatrolite (4.7 ppm), tugtupite (1.3 ppm), quartz (1.0 ppm) and ussingite (0.7 ppm) but not chkalovite (< 0.5 ppm) or epistolite (< 1 ppm)

*Cobalt.* Median Co contents decrease from augite syenite (6.0 ppm) through pulaskite + foyaite (0.47 ppm) to sodalite foyaite + naujaite (0.36 ppm). Co levels rise in the bulk kakortokite (1.3 ppm) but fall again in aegirine lujavrite (0.25 ppm), arfvedsonite lujavrite (< 0.05 ppm) and medium- to coarse-grained lujavrite (0.20 ppm).

The Fe/Co ratio for the average nepheline syenite at Ilímaussaq (137000) is higher than all values in our survey of nepheline syenites and phonolites. Significantly, low Co contents and high Fe/Co ratios were already present in the earliest rock type at Ilímaussaq – the augite syenite.

In kakortokites, median Co contents decrease from black (3.3 ppm) through red (1.5 ppm) to white layers (0.93 ppm) reflecting the fact that Co is mainly hosted by arfvedsonite. Arfvedsonite here occurs as a cumulus phase and contains higher Co (4.2–4.8 ppm) than in the preceding interstitial arfvedsonite from the roof-zone naujaite. Co is notably present, substituting for Fe, in accessory löllingite (0.92%) and westerveldite

(0.38%) from a white kakortokite (GGU 154327; Karup-Møller 1978). The Fe/Co ratios for löllingite (29) and westerveldite (110) are much lower than in the host rock (85 000) and emphasise the chalcophile behaviour of Co in these phases.

Scavenging of Co by arfvedsonite from the magma co-existing with the kakortokites led to a fall in Co contents. Thus arfvedsonites from early black layers - 9 (4.7 ppm) and 0 (4.8 ppm) contain more Co than arfvedsonites from black layer +16 (4.2 ppm) and layer A (2.6 ppm) in the later, transitional layered kakortokites. In kakortokites, Co was also found in fluorite (0.01 ppm) but not in eudialyte (< 1 ppm) or microcline + nepheline (< 0.5 ppm).

*Gallium.* Ga contents reach exceptionally high levels in Ilímaussaq rocks, averages increasing from 27 ppm in augite syenite through 59 ppm in pulaskite + foyaite, 69 ppm in sodalite foyaite + naujaite, 60 ppm in bulk kakortokites and 102 ppm in aegirine lujavrite to 113 ppm in arfvedsonite lujavrite and 106 ppm in medium- to coarse-grained lujavrite.

Analyses of minerals from agpaitic nepheline syenites reveal around 180 ppm Ga in analcime and 270 ppm in natrolite. Lower contents are found in microcline (142–144 ppm), arfvedsonite (57–62 ppm), nepheline (28 ppm) and sphalerite (15 ppm).

Within the katophorite–arfvedsonite series, during crystallisation from pulaskite to aegirine lujavrite, Ga contents decrease from around 54 to 37 ppm along with Al contents from around 1.9 to 0.75%. In late arfvedsonites, however, from arfvedsonite lujavrite to medium- to coarse-grained lujavrite and derivative pegmatites, Al contents continue to fall from around 0.75 to 0.4% whereas Ga contents increase from around 37 to 62 ppm.

For any given rock type, Ga/Al ratios increase in the sequence aluminosilicate < arfvedsonite < aegirine < magnetite, i.e. ratios increase along with rising Fe<sup>3+</sup> contents of minerals. Thus Ga, rather than Al, preferentially enters minerals rich in Fe<sup>3+</sup> reflecting the similar ionic radii of Ga<sup>3+</sup> (0.620 Å VI) and Fe<sup>3+</sup> (0.645 Å VI).

*Germanium.* Median Ge contents decrease slightly through the Ilímaussaq rocks from around 1.8 to 1.5 ppm. The outstanding feature of Ge geochemistry at Ilímaussaq is the exceptionally low and falling contents in feldspars during magmatic evolution, from about 0.8 to 0.2 ppm. This trend is discussed elsewhere in this volume (Bailey 2001, this volume).

*Hafnium.* Median Hf contents rise from 9 ppm in augite syenite through 40 ppm in pulaskite + foyaite and 67.5 ppm in sodalite foyaite + naujaite to 219 ppm in bulk kakortokite and then fall through 101 ppm in aegirine lujavrite and 47 ppm in arfvedsonite lujavrite to 7.7 ppm in medium- to coarse-grained lujavrite. Individual samples emphasise the extremes of concentration and depletion, from 5.0 ppm in a primitive augite syenite to 1070 ppm in a red kakortokite and down to 2.6 ppm in a medium- to coarse-grained lujavrite.

Median Zr/Hf ratios are relatively constant from augite syenite (45) through pulaskite + foyaite (49) and sodalite foyaite + naujaite (45) to bulk kakortokite (44). But in the later rocks, there is a steady increase from aegirine lujavrite (73) and arfvedsonite lujavrite (86) to medium- to coarse-grained lujavrite (97). These are some of the highest Zr/Hf ratios recorded in igneous rocks.

In aegirine lujavrite I, about 97% of all Hf is held in eudialyte which shows contents decreasing overall from 1580 to 1320 ppm at higher stratigraphic levels and Zr/Hf ratios rising from 63 to 70. Hf contents of aegirine decrease from 59 to 38 ppm with Zr/Hf rising from 37 to 41.

Eudialytes studied to date from early sodalite foyaite to late aegirine lujavrite I show decreasing Hf contents from *c.* 2000 to 1320 ppm with Zr/Hf ratios rising from *c.* 50 to 70. The same overall trend is shown by arfvedsonite (Hf from *c.* 60 to 6 ppm, Zr/Hf from *c.* 37 to 86). Rising Zr/Hf trends in rocks and minerals reflect the preferential partitioning of Hf<sup>4+</sup> (0.71 Å) rather than Zr<sup>4+</sup> (0.72 Å) into eudialyte where the site dominated by Zr<sup>4+</sup> has an optimum size for cations close to 0.67 Å (Johnsen & Grice 1999).

Following its extensive fractionation in the kakortokites and aegirine lujavrite I, eudialyte is absent from the final lujavrite (medium- to coarse-grained lujavrite) and from the late pegmatites and veins. The reduced amounts of Zr and Hf substitute for REE–Th (steenstrupine), Nb–Ti (neptunite, epistolite, pyrochlore, lueshite) and perhaps Fe (aegirine, arfvedsonite). Zr/Hf ratios are exceptionally high in some of these late minerals rising to 200 or more in epistolite and lueshite.

*Indium.* Sphalerite separates from aegirine lujavrite and medium- to coarse-grained lujavrite contain 8 and 10 ppm In, respectively (Karup-Møller 1978).

*Iodine.* Median iodine contents at Ilímaussaq rise from augite syenite (< 0.5 ppm) and pulaskite + foyaite (<

0.5 ppm) to sodalite foyaite + naujaite (0.6 ppm). In the floor cumulates, iodine was undetected in bulk kakortokite (< 0.5 ppm) and aegirine lujavrite (< 0.5 ppm) but contents rise through arfvedsonite lujavrite (0.8 ppm) to medium- to coarse-grained lujavrite (1.8 ppm).

In naujaite, there are clear stratigraphic trends with iodine contents decreasing from around 1.7 ppm in the earliest, uppermost samples to 0.5 ppm in the deepest samples. This reflects the pronounced removal of iodine in the earliest cumulus crystals of sodalite.

*Mercury.* Contents of Hg range from 0.62 ppb in alkali granite to 11.7 ppb in naujaite (Stakheev *et al.* 1975).

*Rubidium.* Median Rb contents increase from 83 ppm in augite syenite through 312 ppm in pulaskite + foyaite, 364 ppm in sodalite foyaite + naujaite, 459 ppm in bulk kakortokite and 674 ppm in aegirine lujavrite + arfvedsonite lujavrite to 1280 ppm in medium- to coarse-grained lujavrite. Median K/Rb ratios decrease from 463 in augite syenite through 134 in pulaskite + foyaite, 83 in sodalite foyaite + naujaite, 79 in bulk kakortokite and 40 in aegirine lujavrite + arfvedsonite lujavrite to 35 in medium- to coarse-grained lujavrite.

In aegirine lujavrite I, Rb contents parallel K showing maxima in layers rich in microcline. These maxima have Rb contents of 775–1130 ppm whereas layers with background levels of K-feldspar only contain about 400 ppm. Preferred Rb contents in minerals are: microcline + nepheline (2570–3110 ppm Rb, K/Rb 42–49), arfvedsonite (79–104 ppm, K/Rb 215–272), eudialyte (47–116 ppm, K/Rb 47–74), analcime + natrolite (6.5–128 ppm, K/Rb 38–132) and aegirine (3–4 ppm, K/Rb 218–366) (Bailey 1995). Hydrothermal coatings scraped from fracture planes contain 93–450 ppm Rb.

The evolution of Rb contents in the rock-forming minerals at Ilímaussaq has not been systematically studied but we note that Rb contents in microcline increase from 125 ppm in augite syenite to 3100 ppm in aegirine lujavrite I. Rb contents in arfvedsonite increase from 57 ppm in naujaite to 142 ppm in medium- to coarse-grained lujavrite, and in eudialyte from 20 ppm in foyaite to 81 ppm in aegirine lujavrite I.

*Scandium.* Median Sc contents fall from 30 ppm in augite syenite through 0.52 ppm in pulaskite + foyaite to 0.16 ppm in sodalite foyaite + naujaite. The succeeding kakortokites have a bulk content of 2.0 ppm

but there is a further fall to 0.018 ppm in aegirine lujavrite and arfvedsonite lujavrite with only a slight recovery in medium- to coarse-grained lujavrite (0.075 ppm).

In augite syenite, Sc decreases from 39 ppm to 3.5 ppm as total Fe falls from 10.5 to 2.4 wt%. Sc is dominantly held in hedenbergite (226 ppm) with lesser amounts in magnetite (c. 4 ppm), apatite (1.1 ppm), olivine (< 1 ppm) and microcline (0.04 ppm).

In the lower layered kakortokites, there is a clear Sc–Fe correlation with median contents of Sc decreasing from black (5.1 ppm) through red (1.9 ppm) to white (1.5 ppm) kakortokite. In the black layers of the lower layered kakortokites, Sc ranges from 9.8 to 2.4 ppm as total Fe falls from 19.5 to 9.1 wt%. The dominant mafic phase, arfvedsonite, contains 5.8–13 ppm Sc and Sc is also present in cumulus aegirine (4.7 ppm), eudialyte (0.6–2.1 ppm) and fluorite (0.004–1.3 ppm) but not in aenigmatite or microcline + nepheline (both < 0.3 ppm).

By the time the following aegirine lujavrites crystallised, Sc appears to have been almost totally removed from the system; contents average 0.1 ppm and even fall to 0.0x ppm. A succeeding modest recovery in median Sc contents is seen in values for arfvedsonite lujavrite (0.05 ppm) and medium- to coarse-grained lujavrites (0.1 ppm).

The Fe–Sc relationship is absent in a group of Fe-poor, complex oxides and silicates with high contents of quadri- and pentavalent ions. These minerals – pyrochlore, astrophyllite, rinkite, epistolite and catapleiite – occur in pegmatites and veins and contain up to 9.3 ppm Sc. The association arises from the similarity in ionic radii between Sc<sup>3+</sup> (0.745 Å) and Ti<sup>4+</sup> (0.605 Å), Zr<sup>4+</sup> (0.72 Å), Nb<sup>5+</sup> (0.64 Å) and Sn<sup>4+</sup> (0.690 Å).

*Selenium.* Se was reported on or just above the detection limit (0.01%) during microprobe analysis of Ilímaussaq sulphides as follows: chalcocallite 0.02%, rohaite and cuprostibite – both 0.01% but thalcosite < 0.01% (Makovicky *et al.* 1980). However, we were unable to detect 1 ppm Se in cuprostibite. Instead, Se was found in a vein galena (850 ppm) and in sphalerites from medium- to coarse-grained lujavrite (33 ppm), a pegmatite (23 ppm) and two veins (28 and 47 ppm). The sulphur-bearing silicates sodalite and helvine contain < 1 ppm Se emphasising the chalcophile character of Se.

*Strontium.* Median Sr contents decrease from augite

syenite (284 ppm) through pulaskite + foyaite (25 ppm) to sodalite foyaite + naujaite (11 ppm) but there is a significant increase in bulk kakortokite (87 ppm) followed by a further decrease in aegirine lujavrite (74 ppm), arfvedsonite lujavrite (61 ppm) and medium- to coarse-grained lujavrite (45 ppm).

Thus, Sr contents appear to increase from the apgaitic roof rocks to the eudialyte-rich floor cumulates – the kakortokites. This reflects the fact that, in the kakortokites, eudialyte and fluorite with high Ca and Sr contents became cumulus phases. Their extraction led to CaO contents falling from about 2.0 to 0.2%, and Sr falling from about 100 to 40 ppm in the subsequent lujavrites. As a result, the final lujavrites move to a unique position in Ca–Sr space finishing with a very low Ca/Sr ratio of c. 30.

*Tungsten.* Median W contents increase from 1–2 ppm in augite syenite to 3 ppm in pulaskite + foyaite and 6 ppm in sodalite foyaite + naujaite to 15 ppm in bulk kakortokite. Thereafter W contents decrease to 10 ppm in aegirine lujavrite, 4 ppm in arfvedsonite lujavrite and 5 ppm in medium- to coarse-grained lujavrite.

At Ilímaussaq, W contents correlate strongly with Zr rising to a value of 75 ppm W in a red kakortokite with a particularly high content of eudialyte. Analyses of eudialytes reveal wide variations in their W contents. An interstitial eudialyte from pulaskite contains a high level of 244 ppm W. There is a remarkable increase in W contents of eudialytes from 4 ppm at the sodalite foyaite + naujaite contact through 61 ppm, 129 ppm and 146 ppm at progressively deeper, later stages. This reflects the build-up of W in residual magmas at the naujaite stage.

With the arrival of eudialyte on the liquidus in the lower layered kakortokites, the earliest available eudialyte (from layer –10) contains 295 ppm W. Contents fall progressively through 248 ppm (layer –9), 200 ppm (layer –4), 184 ppm (layer 0) and 143 ppm (layer +3) to 101 ppm in layer A of the later transitional layered kakortokites. It seems clear that eudialyte preferentially extracts W from the kakortokite magmas so that later stages have lower contents.

*Vanadium.* The Ilímaussaq rocks contain exceptionally low V contents. Overall, median values decrease from 3.4 ppm in augite syenite to 1.1 ppm in pulaskite + foyaite and 0.7 ppm in sodalite foyaite + naujaite; in the subsequent floor cumulates V concentrates in arfvedsonite-rich materials – bulk kakortokite (4.0 ppm), arfvedsonite lujavrite (5.6 ppm) and medium- to coarse-

grained lujavrite (4.2 ppm) – rather than in aegirine lujavrite (1.3 ppm). The increased V contents in the floor sequence of agpaitic rocks coincide with increasing levels of Fe, one of the characteristic features of agpaitic crystallisation.

The unusual geochemistry of V in the Ilímaussaq complex has its origins in the augite syenite, the immediate parent magma for the complex. The augite syenite contains 1.7–9.9 ppm V which is mainly held in titanomagnetite (156 ppm) and hedenbergite (6 ppm) but not in olivine (< 1 ppm) or microcline (< 0.5 ppm). The conclusion seems inescapable that the augite syenite parental magma at Ilímaussaq had experienced considerable fractionation of V before it was emplaced; this presumably occurred in a deeper magma chamber (cf. Larsen & Sørensen 1987).

In pegmatites, V ranges widely from 54–59 ppm in rinkite to 24–25 ppm in aenigmatite, 17–23 ppm in steenstrupine, 0.9–18 ppm in arfvedsonite, 0.7–6.0 ppm in polyolithionite, 2.1 ppm in aegirine and < 0.5 ppm in microcline and catapleite.

In veins, V ranges even more widely from 0.5–479 ppm in epistolite, 233 ppm in astrophyllite, 112 ppm in neptunite, 64 ppm in lueshite, 6.4–12 ppm in steenstrupine, < 0.5–8.2 ppm in pyrochlore, 4.4–4.7 ppm in sphalerite, 2.6 ppm in calcite, 1.5–1.6 ppm in arfvedsonite, 0.9 ppm in pectolite, < 0.5–0.6 ppm in aegirine, < 0.5–0.5 ppm in natrolite and < 0.5 ppm in eudialyte, sodalite, sorensonite, ussingite, tugtupite, tetranatrolite, chkalovite, analcime and albite.

*Concluding remarks.* Overall then, for the dominant agpaitic nepheline syenites of the Ilímaussaq complex, Co, Sc and V can be safely grouped with the ferromagnesian elements. The chalcophile character of Bi, Cd, In and Se is already discernible, though a quantitative assessment must await whole-rock analyses. Hafnium and W follow other high field-strength elements into eudialyte and both elements are impoverished in late pegmatites and veins. Iodine and Br closely follow their fellow halogen Cl being largely held by the sodalites of sodalite foyaite and naujaite. Gallium, Rb and Sr are well-known examples of camouflaged elements which substitute in Al, K and Ca minerals, respectively; they maintain this behaviour in the Ilímaussaq complex.

## Summary

1. The agpaitic nepheline syenites of the Ilímaussaq complex are characterised by a great variety of sodic minerals, and by high contents of Zr, Hf, Nb, Ta, REE, Th, U, Sn, Li, Be, Rb, Zn, Pb, Sb, W, Mo, As, Ga, F, Cl, Br, I and S but low levels of Ba, Sr, Co, Cu, Ni, Sc, V and Cr.
2. The agpaitic nature of Ilímaussaq magmas led to the appearance of Na-rich rock-forming minerals (sodalite, albite, arfvedsonite, aegirine, eudialyte) and combinations of Na with rare elements, e.g. Na–Nb (pyrochlore), Na–REE (steenstrupine), Na–Be (chkalovite) and Na–P (vitusite).
3. During fractionation of the agpaitic magmas, separation of cumulus phases with a distinctive geochemistry, such as sodalite (rich in Cl, Br, I, B), eudialyte (Zr, Hf, Nb, Ta, W, As) and arfvedsonite (Co, Sc, V, Cr), was responsible for exhaustion of these elements in the residual magmas.
4. The agpaitic magmas were capable of dissolving high contents of Zr (c. 9000 ppm), REE, Y and P, as well as normally volatile or mobile elements such as H<sub>2</sub>O, Cl, Br, I, S, Mo, As, Tl and Cs.
5. The agpaitic magmas underwent more than 99% fractional crystallisation from the parental augite syenite. This fractionation, along with the cumulate character of most rocks, was responsible for the extreme concentration and impoverishment of the different elements.
6. Whole-rock analyses define a number of discrete Zr–U arrays which are restricted to certain intervals of the cumulate stratigraphy and are taken as evidence for liquid layering in the Ilímaussaq magma chamber.
7. The distribution of the less common trace elements (Bi, Cd, Co, Ge, In, I, Hg, Sc, Se, V, W) and of the more abundant trace elements which failed to form minerals (Br, Cs, Ga, Hf, Rb, Sr) is summarised.



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