

# Stability of Na–Be minerals in late-magmatic fluids of the Ilímaussaq alkaline complex, South Greenland

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Various Na-bearing Be silicates occur in late-stage veins and in alkaline rocks metasomatised by late-magmatic fluids of the Ilímaussaq alkaline complex in South Greenland. First, chkalovite crystallised with sodalite around 600°C at 1 kbar. Late-magmatic assemblages formed between 400 and 200°C and replaced chkalovite or grew in later veins from an H<sub>2</sub>O-rich fluid. This fluid is also recorded in secondary fluid inclusions in most Ilímaussaq nepheline syenites. The late assemblages comprise chkalovite + ussingite, tugtupite + analcime ± albite, epididymite + albite, bertrandite ± beryllite + analcime, and sphaerobertrandite + albite or analcime(?).

Quantitative phase diagrams involving minerals of the Na–Al–Si–O–H–Cl system and various Be minerals show that tugtupite co-exists at 400°C only with very Na-rich or very alkalic fluids [ $\log(a_{\text{Na}^+}/a_{\text{H}^+}) > 6-8$ ;  $\log(a_{\text{Be}^{2+}}/(a_{\text{H}^+})^2) > -3$ ]. The abundance of Na-rich minerals and of the NaOH-bearing silicate ussingite indicates the importance of both of these parameters. Water activity and silica activity in these fluids were in the range 0.7–1 and 0.05–0.3, and  $X_{\text{NaCl}}$  in a binary hydrous fluid was below 0.2 at 400°C. As bertrandite is only stable at < 220°C at 1 kbar, the rare formation of epididymite, eudidymite, bertrandite and sphaerobertrandite by chkalovite-consuming reactions occurred at still lower temperatures and possibly involved fluids of higher silica activity.

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Beryllium minerals occur in small quantities in many magmatic rocks of broadly granitoid composition and in larger quantities in some granitic pegmatites. The earliest Be mineral in most granitic rocks is beryl; later minerals, in many cases replacing beryl, include euclase, bertrandite, phenakite (e.g. Mårtensson 1960; Černý 1963, 1968; Roering & Heckroodt 1966; Burt 1975; Hemingway *et al.* 1986; Markl & Schumacher 1997), herderite (Burt 1975) and many more. The greatest variety and abundance of Be minerals is invariably connected to pegmatitic liquids and late- to postmagmatic fluids. Table 1 provides the chemical formulae of Be minerals mentioned and discussed in the present paper.

In contrast to these ‘normal’ granitoid Be minerals, Na-rich peralkaline rocks are typically devoid of the Be minerals mentioned above, but contain Na–Be silicates such as chkalovite, tugtupite, epididymite, eudidymite or sorensonite (Semenov *et al.* 1965; Semenov & Sørensen 1966; Andersen 1967; Mandarino & Harris 1969; Sobolev *et al.* 1970; Engell *et al.* 1971 and references therein). Interestingly, no corresponding K–Be

silicate assemblages exist in K-rich, peralkaline magmatic rocks. The physico-chemical conditions under which the Na-bearing Be silicates crystallise and the compositional parameters which favour the Na-bearing or the Na-free Be mineral assemblages in the Ilímaussaq alkaline complex in South Greenland are the topic of this contribution. Of special interest is the unique occurrence of large masses of pinkish red tugtupite which is used as a semiprecious gemstone in the Ilímaussaq area.

The Ilímaussaq alkaline complex (Fig. 1) was emplaced in the late Gardar period at about 1.16 Ga (unpublished data by the author, see also Sørensen 2001, this volume) into the Mesoproterozoic Julianehåb granite and earlier Gardar sandstones and basalts. The depth of intrusion is estimated as about 3–4 km, corresponding to about 1 kbar pressure (Konnerup-Madsen & Rose-Hansen 1982, 1984). The intrusive complex was formed by three distinct magma batches which are believed to represent successively more fractionated liquids from an alkali basaltic magma chamber at depth (Larsen & Sørensen 1987; Sørensen & Larsen 1987). It

Table 1. Microprobe analyses of zoned crystals of steenstrupine from the Ilímaussaq alkaline complex

Sample No.	50-149.7 centre (5)	50-149.7 rim (6)	50-150.6 centre (2)	50-150.6 rim (1)	199104 centre (4)	199104 rim (1)
SiO <sub>2</sub>	26.94	27.75	27.54	25.98	27.40	27.06
TiO <sub>2</sub>	0.16	0.07	0.28	0.18	0.25	0.20
ZrO <sub>2</sub>	n.a.	n.a.	n.a.	n.a.	1.41	2.33
ThO <sub>2</sub>	3.95	0.42	4.13	1.40	5.04	1.83
Al <sub>2</sub> O <sub>3</sub>	n.a.	n.a.	n.a.	n.a.	0.28	0.04
La <sub>2</sub> O <sub>3</sub>	8.78	12.85	8.94	14.22	11.49	11.79
Ce <sub>2</sub> O <sub>3</sub>	14.57	15.53	13.80	13.76	15.74	16.49
Pr <sub>2</sub> O <sub>3</sub>	1.29	1.04	1.28	1.54	2.39	2.08
Nd <sub>2</sub> O <sub>3</sub>	3.72	2.80	3.50	2.78	4.01	4.22
Sm <sub>2</sub> O <sub>3</sub>	0.31	0.19	0.19	0.15	n.a.	n.a.
Y <sub>2</sub> O <sub>3</sub>	1.47	1.05	1.56	0.31	1.05	0.10
FeO	4.34	2.50	3.96	1.85	3.87	3.42
VnO	3.61	6.22	3.42	5.69	4.42	5.00
CaO	2.18	1.58	2.00	1.65	1.88	1.95
SnO	0.10	0.14	0.07	0.11	n.a.	n.a.
Na <sub>2</sub> O	8.41	12.98	7.88	13.65	2.75	6.59
K <sub>2</sub> O	0.05	0.02	0.20	0.31	n.a.	n.a.
P <sub>2</sub> O <sub>5</sub>	9.36	11.26	8.50	9.35	11.31	12.30
Nb <sub>2</sub> O <sub>5</sub>	0.10	0.24	0.13	0.67	n.a.	n.a.
U <sub>3</sub> O <sub>8</sub>	n.a.	n.a.	n.a.	n.a.	0.70	0.35
	89.34	96.64	87.38	93.60	93.89	95.75
ΣRE <sub>2</sub> O <sub>3</sub>	28.67	32.41	27.71	32.45	33.63	34.58
La <sub>2</sub> O <sub>3</sub> /Nd <sub>2</sub> O <sub>3</sub>	2.36	4.59	2.55	5.12	2.86	2.79

Analyses in oxide wt%. H<sub>2</sub>O and F have not been analysed which explains the low totals.

Figures in brackets: number of analyses.

n.a. = not analysed.

Samples 50-149.7 and 50-150.6 are from drill core 50 at 149.7 and 150.6 m, respectively. Analyst: G.N. Nechelyustov.

199104 is a GGU sample from Makovický & Karup-Møller (1981), recalculated from element wt% to oxide wt%.

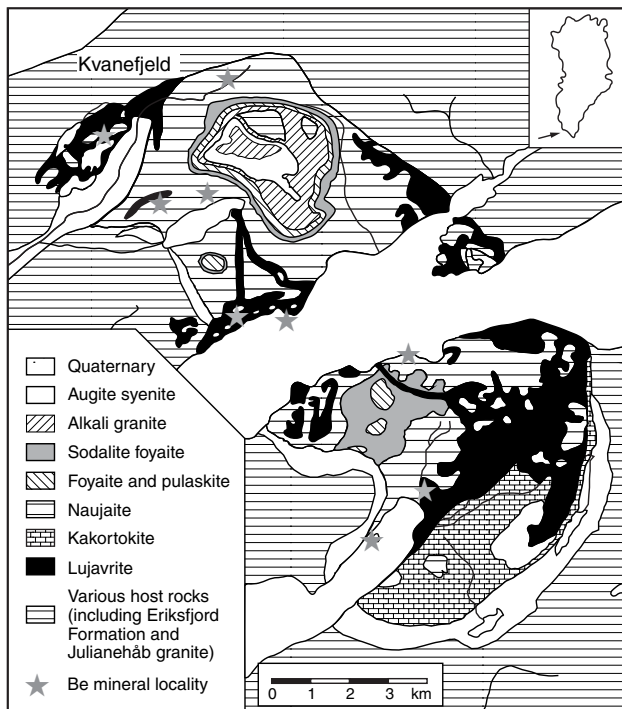


Fig. 1. Geological map of the Ilímaussaq alkaline complex based on Ferguson (1964). The stars denoting the localities where Be minerals have been found are after Engell *et al.* (1971). Note that Be minerals additionally occur at various places in the alkali granite.

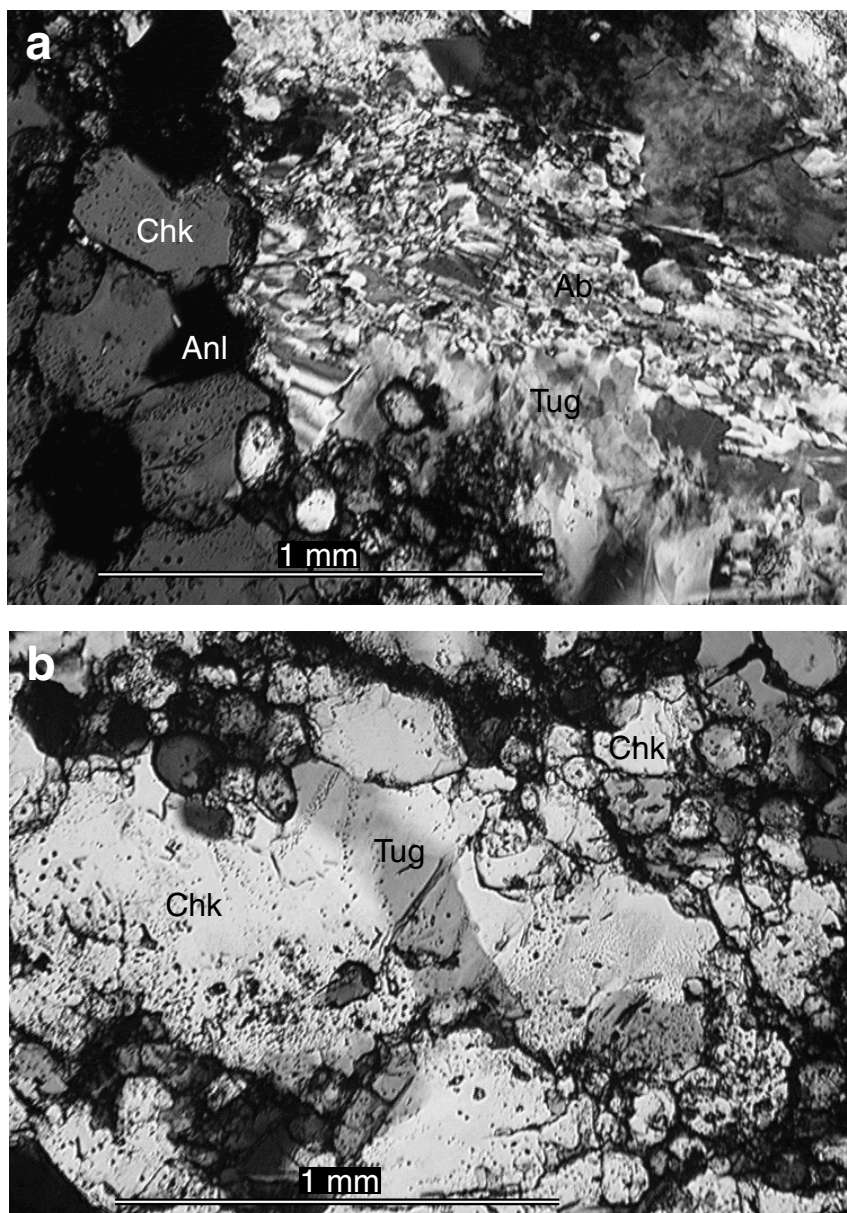
consists of a variety of rock types, ranging from early, just silica-saturated or slightly under-saturated augite syenite through agpaitic nepheline syenites (sodalite foyaite, naujaite and kakortokite) to later, extremely silica-undersaturated nepheline syenites (lujavrites) and finally to hyper-agpaitic assemblages which probably crystallised from relatively SiO<sub>2</sub>-rich fluids rather than from SiO<sub>2</sub>-undersaturated melts. In addition to this fractionation line towards lower silica activities which comprises two out of three different intrusive events at Ilímaussaq (phases 1 and 3), the second intrusive phase is represented by a small sheet of alkali granite. This rock type is interpreted as an augite syenite contaminated with SiO<sub>2</sub>-rich crustal rocks or melts, e.g. with Gardar quartzitic sandstones (Stevenson *et al.* 1997), although fluid inclusion studies (Konnerup-Madsen & Rose-Hansen 1984) do not support this hypothesis.

In the course of the 'undisturbed' fractionation process towards lower silica activities, temperature decreased from about 950°C to below 450°C, silica activity dropped from about 0.8 to below 0.2 (Larsen & Sørensen 1987; Markl *et al.* in press; Marks & Markl in press) and water activity continuously increased until saturation with a water-rich fluid was reached at the late lujavrite stage. This fluid was most probably connected to the formation of abundant aegirine-, albite- and analcime-bearing veins cutting through all other Ilímaussaq rocks, which locally contain Be silicates such as chkalovite and tugtupite. Hence, it appears that Be mineralisation is closely connected to late-stage peralkaline magmatic fluids or liquids as was described in great detail by Engell *et al.* (1971).

## Be mineral assemblages and textures in the Ilímaussaq complex

Be minerals known to occur at Ilímaussaq include chkalovite, tugtupite, sorensenite, leucophanite, sphaerobertandite(?), helvite, genthelvite, beryllite, eudidymite, epididymite, bavenite, barylite, bertrandite and Be-bearing leifite (Semenov *et al.* 1965; Semenov & Sørensen 1966; Sobolev *et al.* 1970; Engell *et al.* 1971; Sørensen *et al.* 1971, 1981; Petersen *et al.* 1991, 1994, 1995; Bohse *et al.* 2001, this volume; chemical formulae in Table 1). As far as it can be assessed from the literature and from my own field observations, the first three of these minerals are the only ones occurring in large quantities (tugtupite is even being 'mined' as an important local gemstone); the others appear to be restricted to specific local chemical

Fig. 2. Photomicrographs of tugtupite replacing chkalovite in a sample from the Kvanefjeld area. **a**: Tugtupite (**Tug**), albite (**Ab**) and analcime (**Anl**) enclosing relics of chkalovite (**Chk**). Crossed nicols. **b**: Tugtupite (**Tug**) replacing coarse chkalovite (**Chk**) in a vein-like manner. Finer grained chkalovite may be a second generation or may be recrystallised during late-magmatic deformation. Author's sample GM 1401.



environments. Epididymite and eudidymite are polymorphs and they have been assumed to have the same stability fields in the qualitative and semi-quantitative diagrams in this contribution. Helvite, genthelvite and sorensonite contain additional components (Zn, S or Sn) that complicate evaluation of their stability fields, and therefore they will not be discussed further in this contribution.

The most comprehensive accounts of Ilímaussaq Be minerals and their occurrences are given by Engell *et al.* (1971) and Sørensen *et al.* (1971). According to them, chkalovite is the earliest Be mineral in Ilímaussaq. It occurs together with ussingite or sodalite and appears to have formed due to reaction of late-magmatic fluids with nepheline-bearing wall rocks. The

assemblage chkalovite + sodalite is replaced by a variety of assemblages during infiltration with later hydrothermal fluids, the most important of which is the assemblage tugtupite + analcime + albite. This mode of occurrence is identical to that reported from the Kola peninsula (Lovozero complex; Semenov & Bykova 1960). Typical textures of tugtupite in the late-stage veins from Kvanefjeld in the northern part of the Ilímaussaq complex are shown in Fig. 2 and are more extensively documented in Sørensen *et al.* (1971). Tugtupite forms in evident equilibrium with analcime and most probably albite (although corroded grains of albite associated with tugtupite have been reported in some samples) during metasomatic recrystallisation

Table 2. Equilibrium assemblages involving Be minerals and observed reaction textures in the Ilímaussaq alkaline complex (Na–Be–Al–Si–O–H–Cl system only)

**Assemblages**

- chkalovite + sodalite
- chkalovite + ussingite
- chkalovite + albite
- chkalovite + natrolite (?)
- tugtupite + albite + analcime
- tugtupite + sphaerobrandite + analcime
- tugtupite + chkalovite + analcime + albite (?)
- eudidymite + epididymite + albite
- eudidymite + albite + quartz
- epididymite + sphaerobrandite
- bertrandite + analcime
- beryllite + analcime

**Reaction textures**

- chkalovite + sodalite → tugtupite + analcime
- chkalovite + albite → analcime + tugtupite (?)
- chkalovite → epididymite + sphaerobrandite
- chkalovite + ussingite → tugtupite + analcime
- chkalovite → beryllite
- chkalovite I → chkalovite II
- eudidymite + epididymite + albite → tugtupite + analcime
- epididymite → bertrandite (?)
- epididymite → beryllite
- analcime → ussingite

Compiled from Andersen (1966); Semenov & Sørensen (1966); Engell *et al.* (1971); Sørensen *et al.* (1971) and own observations.

of the complete host rock. As far as one can tell from the outcrops on Kvanefjeld, the most intensely red coloured varieties come from veins cutting through altered augite syenite, whereas veins cutting through altered naujaite or lujavrite appear to show lighter, pinkish or pinkish red colours. All assemblages from Ilímaussaq reported in the literature so far are collected in Table 2. Figure 1 shows the areas of Be mineralisation in the Ilímaussaq intrusion after Engell *et al.* (1971) and own observations.

In the alkali granite, epididymite was reported to form stable, probably rather late assemblages with quartz and albite (Semenov & Sørensen 1966) which are partly altered to bertrandite. In similar textures, epididymite has also been reported from the Puklen intrusion on Nunarsuit in the Gardar province (Pulvertaft 1961). In some Ilímaussaq nepheline syenites and in late-stage analcime-bearing veins, epididymite and eudidymite form by decomposition of chkalovite and are themselves replaced by still later tugtupite.

All in all, the reported and observed textures support the interpretation that chkalovite is the earliest Be mineral in the undersaturated rocks of Ilímaussaq (maybe contemporaneous with epididymite/eudidymite in the alkali granite) and most later Be minerals formed by decomposition of this early chkalovite during infiltration of later hydrous fluids (see also Sørensen *et al.* 1971). The temperature and the most important chemical and physico-chemical characteristics of these fluids may be estimated using various types of phase diagrams.

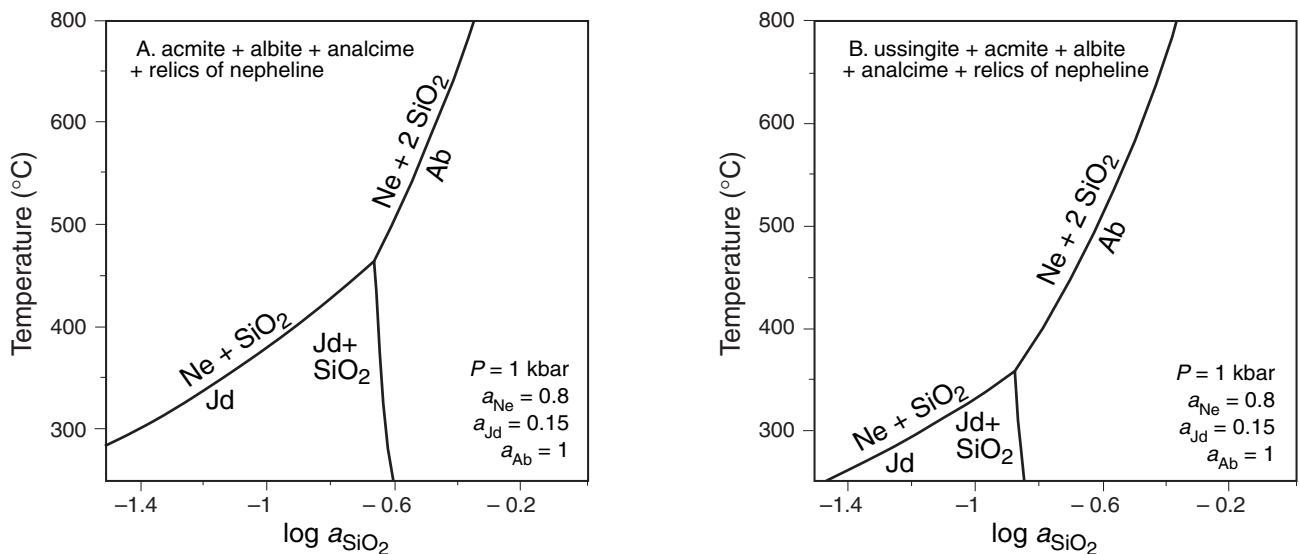


Fig. 3. Temperature–silica activity diagrams for lujavrites from Ilímaussaq (from Sommer 1999). **A:** Ussingite-free, analcime-bearing assemblages, **B:** Ussingite + analcime-bearing assemblages. Nepheline and jadeite activities are calculated from measured mineral compositions reported in Sommer (1999). **Ab:** albite, **Jd:** jadeite, **Ne:** nepheline.

Table 3. Reactions used in the construction of Figures 3 to 7

Fig. 3

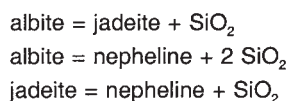


Fig. 5

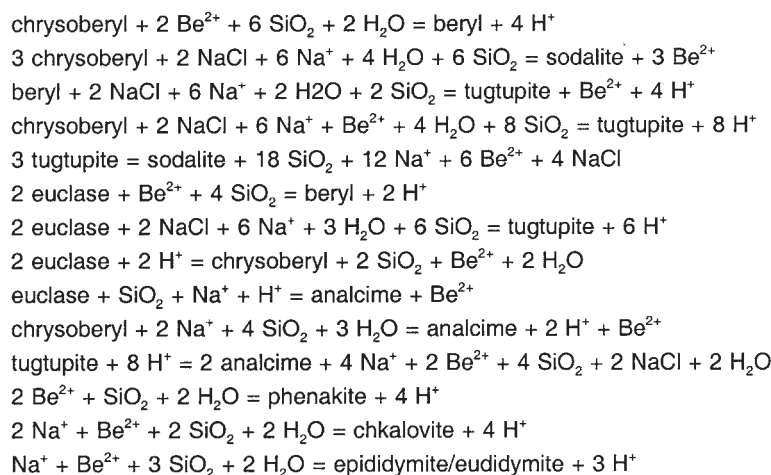


Fig. 6

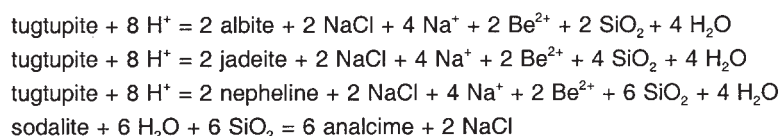
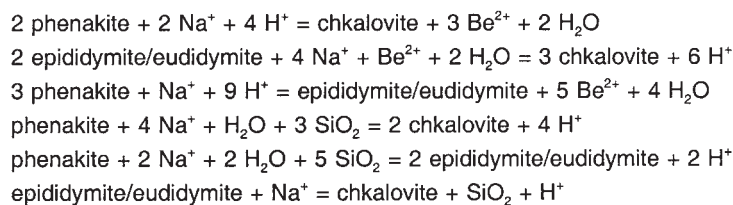


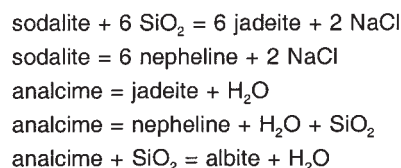
Fig. 7



### Phase diagrams for estimation of Be mineral stabilities

The phase diagrams of this section were calculated using the computer program GEOCALC of Lieberman & Petrakakis (1991) with the thermodynamic database of Berman (1988) for the anhydrous albite–jadeite–nepheline phase diagrams (Figs 3, 4), and the program SUPCRT of Johnson *et al.* (1992) with its thermodynamic database for the rest of the diagrams (Figs 5–7). The latter internally consistent database was augmented with data for sodalite from Sharp *et al.* (1989)

Fig. 4



and for the beryllium minerals beryl, chrysoberyl, euclase, bertrandite and phenakite from Hemingway *et al.* (1986) and Barton (1986). Although thereby losing its internal consistency, this method provides the only means of approximately quantifying the chemical parameters responsible for the formation of the Na–Be silicates. The results appear reasonable insofar as they agree with theoretical (Schreinemakers) predictions and observed phase assemblages, but the actual values may be slightly incorrect. Incorrect values may be the reason for the obviously incorrect feature in Fig. 5, that phenakite saturation occurs at too

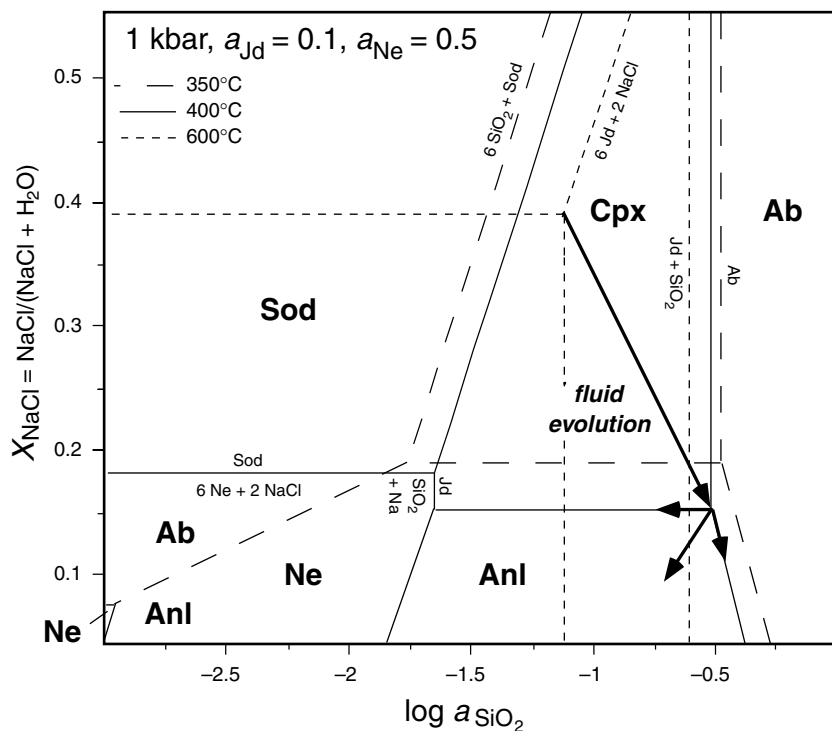


Fig. 4.  $X_{\text{NaCl}}$ -silica activity diagram for minerals in the Na-Al-Si-O-H-Cl system at various temperatures and 1 kbar. Possible fluid evolution paths during cooling are schematically represented by the thick arrows. **Anl**: analcime, **Ab**: albite, **Cpx**: clinopyroxene, **Jd**: jadeite, **Ne**: nepheline, **Sod**: sodalite.

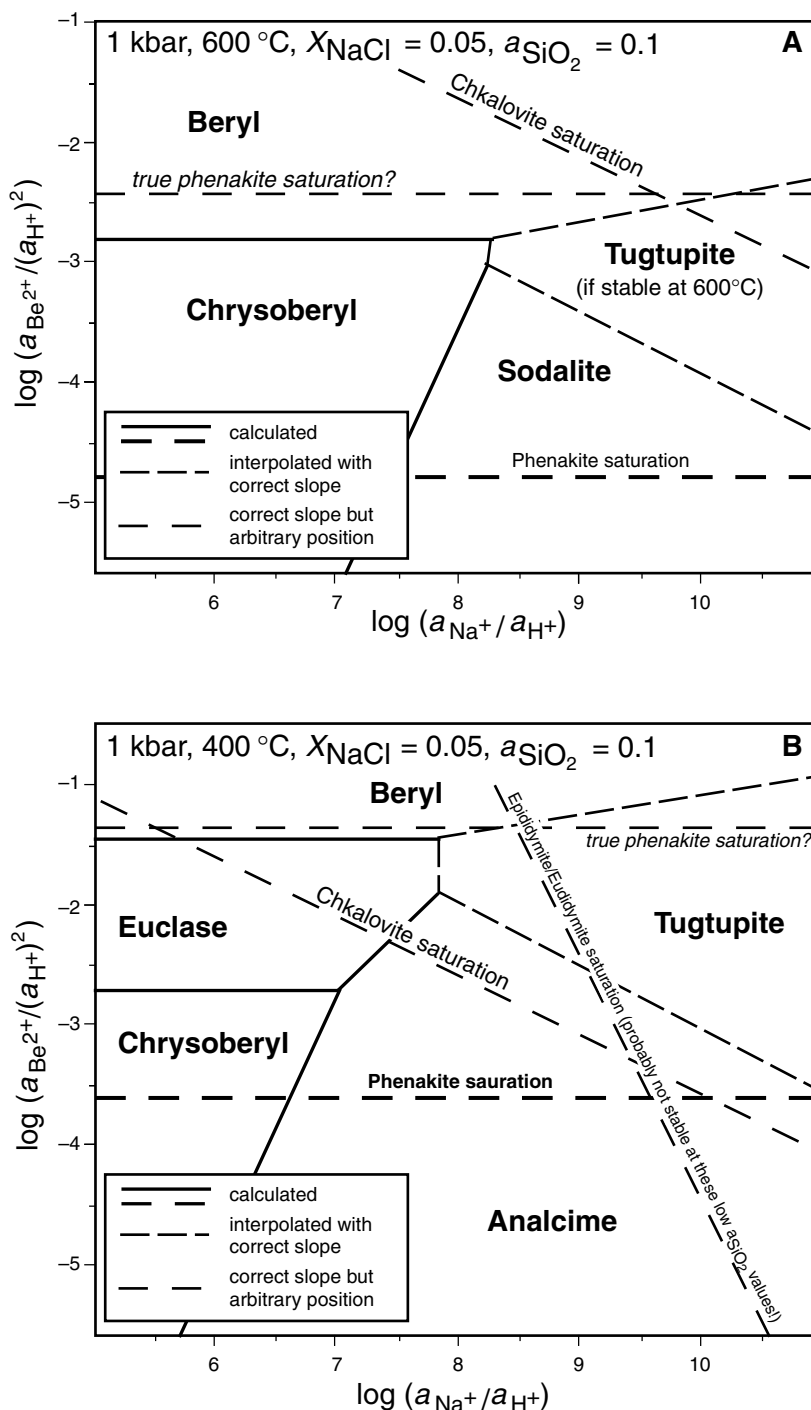
low values to permit stable occurrence of beryl or euclase at 600 and 400°C, respectively. It is assumed that the actual saturation value for phenakite in terms of the  $\text{Be}^{2+}/(\text{H}^+)^2$  activity ratio is about 2 orders of magnitude higher than the calculated value which would be consistent with the rest of the topology on Fig. 5, on which such a possible value is plotted as ‘true phenakite saturation?’. The calculated value for phenakite saturation is also plotted on Fig. 5 and the part of the diagram above phenakite and above chkalovite saturation would correctly have to be termed metastable. The interpretation below, however, assumes that phenakite saturation occurs at values permitting beryl and euclase to be stable in Fig. 5A, B, respectively (‘true phenakite saturation?’).

It was chosen to fix the pressure for all phase diagrams at 1 kbar which is the depth of intrusion of Ilímaussaq based on fluid inclusion studies (Konne-rup-Madsen & Rose-Hansen 1984). Variations in the few hundred bars range do not have a major influence on the results of this study. Hence, fixing the pressure and considering the reactions of interest (Table 3), the following major physico-chemical parameters remain to be investigated: temperature,  $a_{\text{SiO}_2}$ ,  $a_{\text{Be}^{2+}}/(a_{\text{H}^+})^2$ ,  $a_{\text{Na}^+}/a_{\text{H}^+}$ ,  $a_{\text{H}_2\text{O}}$  and  $X_{\text{NaCl}}$ . The latter two are closely connected as it was chosen to treat the fluid, as a first approximation, as a binary  $\text{H}_2\text{O}$ -NaCl mixture with mixing and solution properties calculated according to Aranovich & Newton (1996). In order

to constrain as many of the variables as possible, the problem was divided into several steps as follows.

1. Temperature and  $a_{\text{SiO}_2}$  constraints were estimated as a first step using anhydrous albite-nepheline-jadeite equilibria (Fig. 3) and results from fluid inclusion studies on lujavrites (Sommer 1999; unpublished data). The mineral compositional data for these calculations are reported in Sommer (1999); nepheline and jadeite activities were calculated from the respective solid solutions using the models of Ghiorso (<http://melts.geology.washington.edu>) and Holland (1990). These phase equilibria showed that very late-stage ussingite-bearing lujavrite (which still contains sodic clinopyroxene, albite and relics of nepheline) formed at temperatures around 350°C and at silica activities around 0.1–0.2 (referring to unit silica activity for pure quartz at the applicable  $P$  and  $T$ ). In comparison, fluid inclusion studies indicate homogenisation temperatures between 300 and 400°C for primary and some secondary inclusions in various lujavrite minerals and in tugtupite (Sommer 1999; Markl *et al.* in press). Sobolev *et al.* (1970) measured temperatures of 400–460°C for primary inclusions in tugtupite and sorensenite; secondary inclusions record temperatures between 100 and 350°C. Based on their textures, the chkalovite-sodalite assemblages are believed to have formed at some higher temperatures and a temperature of

Fig. 5. Activity–activity diagrams involving minerals from the Be–Na–Al–Si–O–H–Cl system at 600°C (**A**) and 400°C (**B**). Field boundaries and saturation lines which were calculated from thermodynamic data (see text) are plotted as thick lines; short dashed lines represent interpolated lines with correct (stoichiometric) slope; and widely dashed lines have the correct slope but are not fitted to any independently constrained invariant point. Note that tugtupite may not be stable at 600°C and that epididymite and eudidymite may not be stable at  $\text{SiO}_2$  activities around 0.1. Note also that calculated phenakite saturation occurs at unrealistically low  $\text{Be}^{2+}/(\text{H}^+)^2$  activity ratios and that the part above phenakite and chkalovite saturation in **A** and **B** is actually metastable as plotted here. ‘True phenakite saturation?’ shows a possible value for phenakite saturation consistent with the rest of the Be mineral topology. See text for discussion.



600°C was arbitrarily chosen for this stage. Studies of Sobolev *et al.* (1970) indicate still higher temperatures in the 860–980°C range, but these results are impossible to reconcile with all other estimates in the Ilímaussaq rocks based on melting experiments and phase equilibria (Sood & Edgar 1970; Markl *et al.* in press; Marks & Markl in press).

2. Based on this framework, limits on  $a_{\text{H}_2\text{O}}$  and  $X_{\text{NaCl}}$

of a fluid in equilibrium with the observed phases of the Na–Al–Si–O–H–Cl system (e.g., analcime, sodalite, clinopyroxene, albite etc.) were calculated (Fig. 4). Both variables are tied together when considering a binary  $\text{H}_2\text{O}$ –NaCl fluid only. The activity–composition relationships were derived from the formulation of Aranovich & Newton (1996) and activities were referenced to unit activities for pure  $\text{H}_2\text{O}$  and pure crystalline halite at appropriate  $P$  and

7. These calculations showed that sodalite-bearing assemblages at 600°C require an  $X_{\text{NaCl}}$  greater than about 0.39 (Fig. 4), while analcime-bearing assemblages at 400°C or below require hydrous fluids with an  $X_{\text{NaCl}}$  lower than about 0.15 corresponding to an  $\text{H}_2\text{O}$  activity of 0.7 or higher (Fig. 4). These values are limits for the formation of the two most important Be minerals at Ilímaussaq, chkalovite and tugtupite. The early chkalovite–sodalite assemblage obviously equilibrated with a fluid (or melt?) of relatively high salinity, while later tugtupite, although also Cl-bearing, formed from fluids of lower salinity.

3. These data were then used to place constraints on the fluid chemistry in terms of  $\text{Be}^{2+}/(\text{H}^+)^2$  and  $\text{Na}^+/\text{H}^+$  activity ratios by considering the typical granite association of the Na-free Be-minerals beryl, phenakite, euclase and chrysoberyl and their stability relations with respect to tugtupite and sodalite. In an Al-balanced activity–activity diagram as shown on Fig. 5, phenakite, chkalovite and epididymite/eudidymite do not have their own stability fields but are only shown by saturation lines because they contain no Al. The stability fields and saturation line for beryl, euclase, chrysoberyl, sodalite and phenakite were calculated quantitatively, while those for tugtupite, chkalovite and epididymite/eudidymite – because of the lack of thermodynamic data – were inserted with correct slopes, but at arbitrary positions constrained by calculated invariant points and by the observed mineral assemblages reported above.

Figure 5 shows that at 600°C and 1 kbar, euclase is not stable with respect to chrysoberyl and beryl, while at 400°C, all three Be–Al silicates have stability fields on such a diagram. Whether tugtupite is stable with respect to chkalovite and sodalite at 600°C is not clear – it may well be that at these temperatures, tugtupite is only stable at extremely (unrealistically?) high  $\text{Be}^{2+}/(\text{H}^+)^2$  and/or  $\text{Na}^+/\text{H}^+$  activity ratios, or that it is not stable at all. At 400°C, the best temperature estimate for tugtupite formation, the Be–Al silicates are replaced by sodalite or tugtupite when the  $\text{Na}^+/\text{H}^+$  activity ratio exceeds about  $10^8$ , while the Be concentration in the fluid appears to be of only marginal importance for this reaction. The diagrams of Fig. 5 also clearly show that phenakite is expected to be a stable phase in the Ilímaussaq Be mineral assemblages both at 600°C and at 400°C – with the exception of extremely high  $\text{Na}^+/\text{H}^+$  activity ratios. However, this evaluation at

600°C depends critically on the position of the chkalovite saturation line for which no real constraints exist at this temperature. At 400°C, phenakite should be associated with tugtupite (see Fig. 5B) but probably, in the presence of significant amounts of Zn and S in the fluid, genthelvite is stable with respect to phenakite (Burt 1988). However, genthelvite has only rarely been observed at Ilímaussaq.

4. The combined importance of both  $\text{SiO}_2$  and  $\text{Be}^{2+}/(\text{H}^+)^2$  activity ratio at fixed  $\text{Na}^+/\text{H}^+$  activity ratio and fixed fluid composition for the stability of both chkalovite and tugtupite is investigated in Fig. 6 for a temperature of 400°C. This diagram again combines quantitatively calculated stability field boundaries and saturation lines with ones not calculated but fixed at an arbitrary, but reasonable, position and with correct slope. The salinity of the fluid appears to be of surprisingly little importance for the stability of the NaCl-bearing mineral tugtupite. Silica activity, in contrast, is of much greater importance, as is the  $\text{Be}^{2+}/(\text{H}^+)^2$  activity ratio. This diagram indicates that the assemblage of tugtupite and albite should also contain phenakite as a stable Be phase at silica activities above  $10^{-0.4}$ .

The diagrams of Fig. 7 show the qualitative phase relations among the Al-free Be silicates. Here, no quantitative constraint can be put on the site of the invariant point, where phenakite, chkalovite and epididymite or eudidymite are stable together. However, the slopes of the corresponding univariant reaction curves can be readily calculated from reaction stoichiometry (see Table 3). Phenakite is stable at the highest  $\text{Be}^{2+}/(\text{H}^+)^2$  activity ratios, chkalovite at the highest  $\text{Na}^+/\text{H}^+$  activity ratios and epididymite or eudidymite at the highest  $\text{SiO}_2$  activities.

5. Bertrandite stability was calculated according to the reaction:



at 1 kbar. At this pressure, this reaction lies at about 220°C (see also Markl & Schumacher 1997). This means that the occurrence of bertrandite is clear proof of the circulation of low-temperature hydrous fluids in the Ilímaussaq rocks. It is assumed that the formation of beryllite and sphaerobertrandite (Semenov & Sørensen 1966; Andersen 1967) is also related to these low-temperature hydrous fluids.

6. The compatibility of the various Be mineral assemblages observed in various granitoid rock types can



Fig. 6. Activity–activity diagram at 1 kbar and 400°C constraining the stability of various Be minerals with regard to various Be-free minerals in equilibrium with fluids of variable salinity. Note that only phenakite saturation and the vertical field boundaries among the Na–Al silicates are thermodynamically calculated, the other lines are fixed at arbitrarily chosen invariant points. The grey arrow represents a possible fluid evolution path at constant temperature which would lead to tugtupite formation. Note that everything above the saturation lines are only meta-stable extensions.

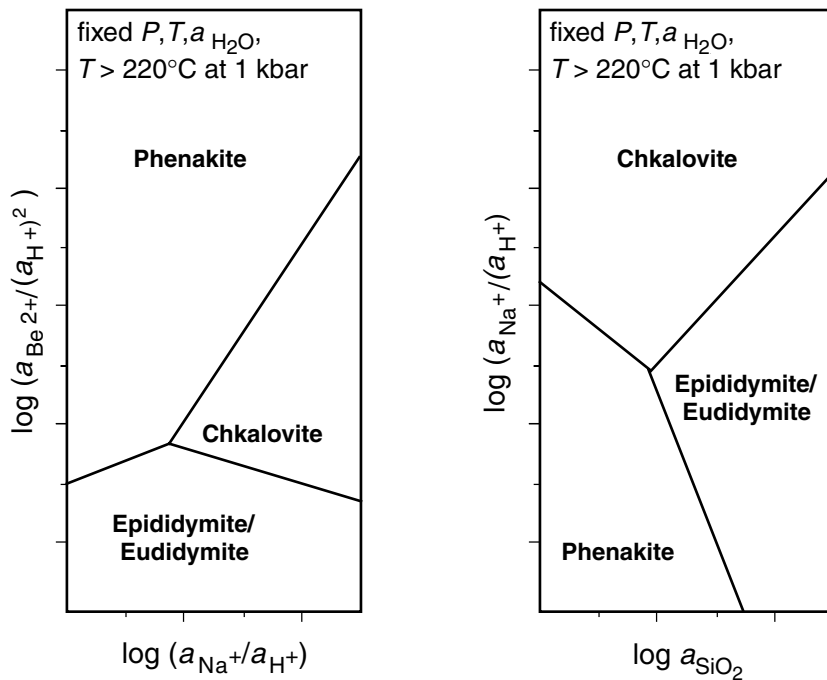
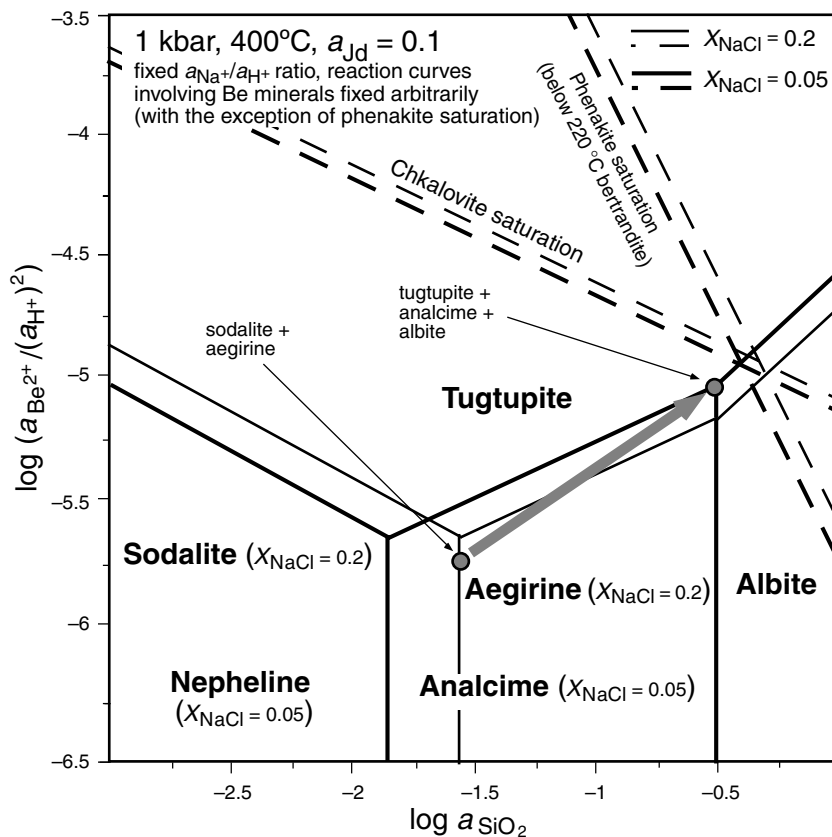


Fig. 7. Qualitative activity–activity diagrams representing the stability relations among various Al-free Be silicates. Note that the slopes are correctly calculated from reaction stoichiometry.

also be illustrated in another way. Figure 8 shows two  $\text{SiO}_2\text{-Na}_2\text{O-BeO}$  triangles with the inferred Be mineral associations at ‘higher’ (e.g. 600°C) and ‘lower’ (e.g. 200°C) temperature. These diagrams are projections from albite and an  $\text{H}_2\text{O-NaCl}$  fluid, and in this projection euclase and chrysoberyl cannot be plotted. At higher temperatures, chkalovite is stable with sodalite and, possibly, with phenakite. If stable at these high temperatures, the assemblage epididymite or eudidymite with phenakite would effectively render the co-existence of chkalovite with beryl impossible. Beryl, however, should be stable with phenakite and with epididymite/eudidymite. Tugtupite appears not to be stable at these high temperatures. All these constraints appear to agree with natural occurrences. At lower temperatures, phenakite is replaced by bertrandite, and beryl and chkalovite may become unstable depending on the exact details of the chemical environment. Tugtupite as well as epididymite/eudidymite may form by various reactions involving chkalovite, phenakite and sodalite. Under these conditions, tugtupite may form a stable assemblage with epididymite/eudidymite and bertrandite (or, at temperatures above 220°C, with phenakite). Epididymite/eudidymite may be in equilibrium with beryl and bertrandite (or phenakite).

## Discussion

### *Stability of Na-bearing versus Na-free Be silicates*

The very pronounced differences in Be mineral assemblages in peralkaline versus calc-alkaline granitoid rocks can be explained in terms of fluid chemistry which is also an approximate monitor of the melt chemistry. Na-bearing Be silicates only appear in peralkaline or alkaline rocks (Fig. 8), whereas calc-alkaline intrusives are characterised by Be- or Be-Al silicates without Na. Chkalovite and tugtupite appear to be restricted to peralkaline rocks (i.e. fluids with extremely high  $\text{Na}^+/\text{H}^+$  activity ratios, Fig. 5), whereas epididymite and eudidymite are indicative of moderately alkaline rocks or fluids forming a link to beryl-bearing assemblages. Epididymite and eudidymite are especially indicative of higher silica activities than those observed in agpaite rocks (Fig. 7). Phenakite at high and bertrandite at low temperatures are stable in both alkaline and calc-alkaline environments, unless helvite-group minerals replace them due to the presence of additional components such as Mn, Zn or S (e.g. Burt 1988). Chrysoberyl is principally expected to be stable at low  $\text{SiO}_2$  activities and in high Al environments but obviously requires additionally either very high

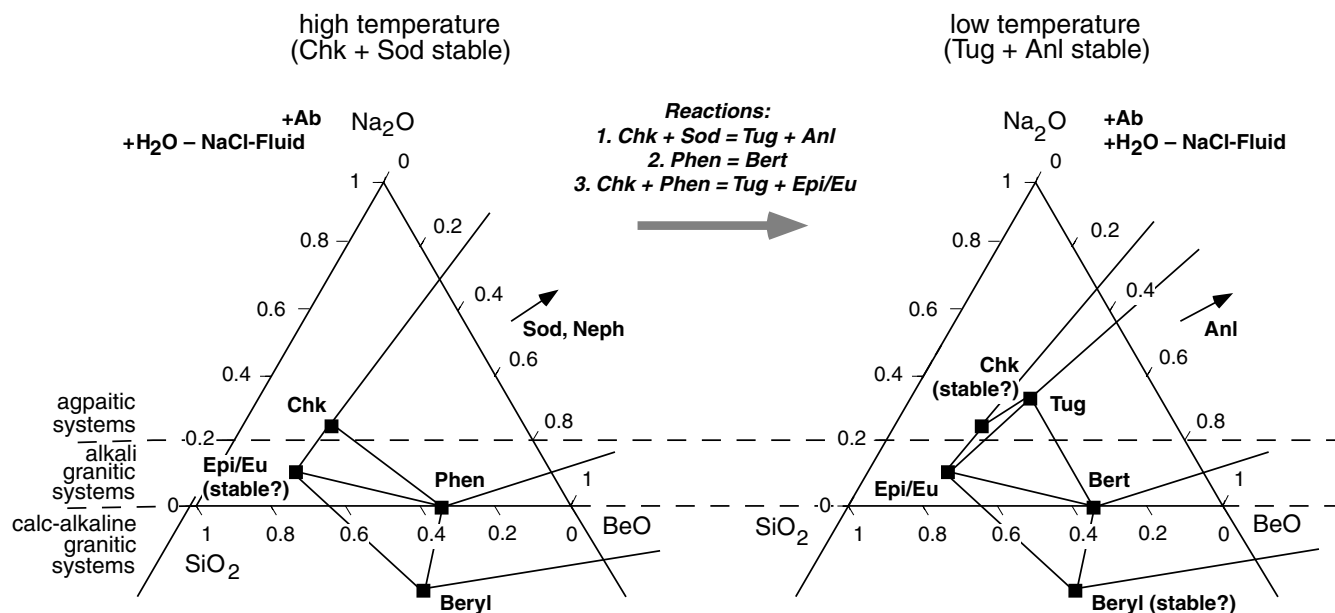
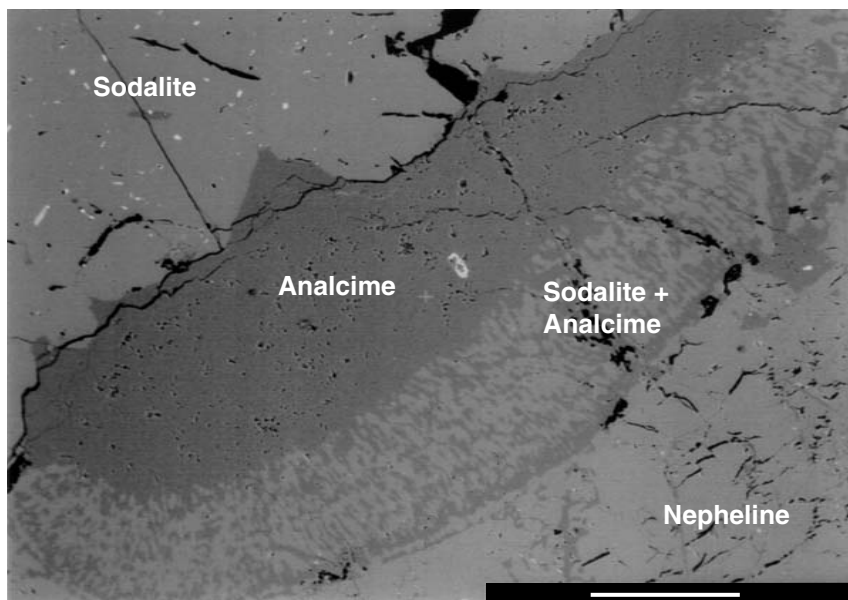


Fig. 8. Phase relations in the Be–Na–Al–Si–O–H–Cl system projected from albite (**Ab**) and a binary  $\text{NaCl-H}_2\text{O}$  fluid (variable salinity has no effect on depicted phase relations). At high temperatures (e.g. 600°C, **left diagram**), chkalovite (**Chk**) + phenakite (**Phen**) + sodalite (**Sod**) or nepheline (**Neph**) form a stable assemblage. Chkalovite + beryl is not stable if epididymite/eudidymite (**Epi/Eu**) is a stable phase at these temperatures. At lower temperature (e.g. 200°C, **right diagram**), chkalovite and beryl may no longer be stable, bertrandite (**Bert**) replaces phenakite, and tugtupite (**Tug**) forms by various reactions. Tugtupite should be stable with both epididymite/eudidymite and bertrandite as well as analcime (**Anl**).

Fig. 9. Reaction textures from an Ilímaussaq sodalite foyaite in which a primary grain boundary between sodalite (upper left) and nepheline (lower right) is replaced by analcime and an analcime + sodalite symplectite. See text for discussion. Scale bar 200  $\mu\text{m}$ . Author's sample GM 1346.

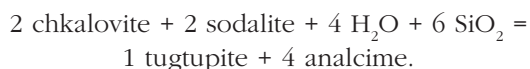


temperatures or very low  $\text{Na}^+/\text{H}^+$  activity ratios, or both, to be stable. Natural occurrences appear to be confined to high-grade metamorphic rocks or metamorphosed pegmatites of low  $\text{SiO}_2$  and low Na content (e.g. Franz & Morteani 1984).

### *Stability of tugtupite versus chkalovite*

The above considerations indicate four possible causes for the conversion from chkalovite- to tugtupite-bearing assemblages: decrease of temperature, increase of  $a_{\text{SiO}_2}$ , increase of  $a_{\text{Be}^{2+}}/(a_{\text{H}^+})^2$ , and increase of  $a_{\text{H}_2\text{O}}$  (= decrease of  $X_{\text{NaCl}}$ ).

All available field observations, fluid inclusion studies and phase equilibrium considerations detailed above indicate decreasing temperature during formation of the various Be–silicate assemblages and during conversion from chkalovite to tugtupite-bearing assemblages. Furthermore, fluid inclusions provide evidence for the existence of very water-rich fluids at temperatures in agreement with tugtupite formation estimates. The net reaction connecting the chkalovite and the tugtupite assemblages is:



At Ilímaussaq this reaction appears to occur around 400°C at  $\text{SiO}_2$  activities around 0.1 (see Figs 3, 4) and with fluids of high  $\text{H}_2\text{O}$  activity. Sobolev *et al.* (1970) reported high salinities of about 21 wt% NaCl equiva-

lent in primary fluid inclusions in tugtupite, while primary as well as secondary hydrous fluid inclusions in various minerals from various lujavrite samples record very low salinities in the 3–5 wt% NaCl equivalent range (Sommer 1999; Markl *et al.* in press). Sobolev *et al.* (1970) also report  $\text{CO}_2$  as an important species in the gas phase of these inclusions, but  $\text{CO}_2$  has never been reported by anyone else in any fluid inclusion from the entire Ilímaussaq alkaline complex, where methane is generally the most important species in any gaseous inclusion. Hence, the data of Sobolev *et al.* (1970) are considered with some caution – perhaps the fluid inclusions in tugtupite leaked  $\text{H}_2\text{O}$  and were oxidised during later cooling, which would explain both their increased salinity and the  $\text{CO}_2$ .

However, consideration of the tugtupite-forming reaction above and comparison with a reaction texture from a different Ilímaussaq rock, a sodalite foyaite (Schwinn 1999), offers a possible explanation compatible with the observation of fluid inclusions of both low and high salinity in the late magmatic stages of the Ilímaussaq complex. This texture in the sodalite foyaite is shown in Fig. 9. There, sodalite and nepheline crystals co-existed at high temperatures. At lower temperatures, an obviously  $\text{H}_2\text{O}$ -rich fluid infiltrated the rock along its grain boundaries and converted both sodalite and nepheline to analcime. The reaction zone consists of a zone of pure analcime close to sodalite, and a zone of symplectic intergrowth of analcime and sodalite close to nepheline. This texture is interpreted to reflect changes in salinity during analcime growth:

while the starting fluid was H<sub>2</sub>O-rich and therefore formed only analcime, it became progressively more saline due to consumption of H<sub>2</sub>O and liberation of NaCl from dissolving sodalite, until the salinity was so high that both analcime and a second generation of sodalite were precipitated in a symplectic intergrowth. This texture in the sodalite foyaite very likely formed during a late-stage auto-metasomatic event similar to the events that led to the formation of the analcime–tugtupite assemblages in the late veins, and hence a similar mechanism may have operated there. In the late veins, sodalite was dissolved along with chkalovite, rendering the fluid successively Be and NaCl rich, while analcime precipitated. At a certain level of Be and NaCl enrichment, tugtupite began to crystallise and enclosed some samples of the more saline fluid which Sobolev *et al.* (1970) found. While this does not explain their report of CO<sub>2</sub>, this mechanism shows how the Cl-rich mineral tugtupite could grow during influx of a hydrous, low-salinity fluid into sodalite-bearing rocks. The pre-existence of sodalite may even be a prerequisite for tugtupite formation – the similarity of their crystal structures and chemical formulae (tugtupite = sodalite + Be<sub>2</sub>Si<sub>2</sub>Al<sub>4</sub>) may also facilitate the chemical conversion of one into the other (see also Burt 1991).

### *Nature of the fluids in late-magmatic, agpaitic systems*

One type of primary fluid inclusions in lujavrites and most secondary fluid inclusions in lujavrite and all other rock types contain a hydrous fluid with about 3 wt% NaCl equivalent (Schwinn 1999; Sommer 1999; Markl *et al.* in press). In rare examples, hydrous fluid inclusions of higher salinity have been found in these rock types and also in tugtupite and chkalovite (Sobolev *et al.* 1970; Konnerup-Madsen & Rose-Hansen 1982), and these may be fluids whose compositions were modified during interaction with sodalite-bearing rocks as detailed above. The possible existence of late-stage immiscible hydrocarbon-bearing fluids (Konnerup-Madsen 2001, this volume) does not alter the results of the present contribution because the aqueous fluid would still approximately behave as a binary H<sub>2</sub>O–NaCl system.

If one considers the low-salinity hydrous fluids to be the typical late-stage fluids of agpaitic rocks (which at least for Ilímaussaq appears to be reasonable), these fluids have one very distinctive feature: they obvi-

ously have an anomalously high pH, as indicated by the stability of ussingite (NaAlSi<sub>3</sub>O<sub>8</sub> NaOH) and as indicated by the extremely high Na<sup>+</sup>/H<sup>+</sup> and Be<sup>2+</sup>/(H<sup>+</sup>)<sup>2</sup> activity ratios calculated for the stability of the observed assemblages. Interestingly, experimental mixtures of andalusite, paragonite and H<sub>2</sub>O develop extremely high pH at temperatures in the 400°C range (L. Baumgartner, personal communication 2000) which is probably caused by the interplay between aluminosilicate dissociation (which consumes H<sup>+</sup> ions) and formation of strong Na-silicate complexes (which replace NaOH-complexes more stable at higher temperatures). These features, however, may only be developed in hydrous fluids with a high Na/(Na+K) ratio and with very low CO<sub>2</sub> contents, such as in agpaitic fluids. The high pH in these fluids creates, at moderate Be and Na ion activities, the extremely high pH-normalised activities which, in turn, give rise to the formation of the observed Be silicates. Neither the absolute Be nor the absolute Na contents in agpaitic fluids have to be very high, if the pH is as high as indicated. In conclusion, the unique mineralisation of agpaitic magmatic rocks and the formation of exotic hyper-agpaitic, water-soluble minerals (e.g. Sørensen & Larsen 2001, this volume) may be an effect of the low oxidation state, low water activity and high Na/(Na+K) ratio of the parental magma which renders formation of a hydrous, CO<sub>2</sub>-poor, K-depleted, high-pH fluid phase possible in the very late stages of magmatic evolution.

### **Conclusions**

Na-bearing Be minerals in the peralkaline Ilímaussaq alkaline complex, South Greenland, formed during cooling and chemical evolution of agpaitic, late-magmatic liquids and fluids in the temperature range 600–200°C. The earliest mineral to form was chkalovite which was successively replaced by tugtupite (in large quantities) and some rare Be silicates in small quantities. Qualitative and quantitative estimates of conditions and chemical parameters during formation indicate that the early assemblages crystallised from liquids (or fluids) of high salinity, while the replacement occurred later during influx of a hydrous fluid of low salinity. Neither high Be nor high Na contents are necessary to explain the observed mineral assemblages, but the high pH of late agpaitic fluids, as demonstrated by the occurrence of the NaOH-bearing silicate ussingite, appears to be responsible for high metal ion/H<sup>+</sup>

activity ratios in the late stage fluids. High pH and high Na/(Na+K) ratios appear to be responsible for the distinct differences in Be mineral assemblages between peralkaline, alkaline and calc-alkaline granitoid rocks.

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