

Primary and secondary mineralogy of the Ilímaussaq alkaline complex, South Greenland



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1. Introduction

The Ilímaussaq alkaline complex is among the largest known alkaline complexes in the world and has been studied since the early 19th century, when Giesecke explored Greenland for minerals. More than 230 different mineral species occur in the complex. Ilímaussaq is the type locality for 34 minerals, including 15 that have not been reported elsewhere. Some of these are rock-forming minerals and thus, although unique to Ilímaussaq, may not be considered rare.

Among the minerals first described from Ilímaussaq are two important sources for critical materials: steenstrupine-(Ce) and eudialyte (Table 1). Steenstrupine-(Ce) is the main target mineral for the Kvanefjeld multi-element project in the northern part of Ilímaussaq, whereas eudialyte is targeted at the Tanbreez project in the southern part known as Kringlerne (Fig. 1).

The first detailed mapping and petrological studies of the complex were published by Ussing (1912), who also defined the term ‘agpaitic’ for rocks where the molar ratio (Na+K)/Al is greater than or equal to 1.2. Since then, the distinction between agpaitic and miaskitic has changed from being based on just rock chemistry to being based more on mineral paragenesis. Sørensen (1997) defined agpaitic as peralkaline rocks in which High Field Strength Elements (HFSE; e.g., Zr and Ti) are hosted in complex minerals such as eudialyte and rinkite. Rocks with high alkalinity, where HFSE are hosted in minerals such as zircon, are considered miaskitic. Khomyakov (1995) further developed the agpaitic classification by introducing the term hyperagpaitic for the most evolved syenites. Hyperagpaitic rocks are characterised by containing water soluble minerals (e.g., natrosilite and natrophosphate) and complex phosphosilicates (e.g., steenstrupine-(Ce) and vuonnemite; Khomyakov, 1995).

The Ilímaussaq complex is one of several alkaline complexes formed during Mesoproterozoic rifting in the southwestern part of Greenland, which collectively is called the Gardar province (Upton, 2013). With an age of ~1.6 Ga, Ilímaussaq is the youngest major intrusion of the Gardar province (Waight et al., 2002; Krumrei et al., 2006).

2. Geology

The Ilímaussaq complex consists of three main intrusive suites (Fig. 1): 1) an augite syenite, which forms the shell of the complex, cuts the Julianehåb granite in the south and a series of sedimentary rocks and volcanic flows in the north; 2) peralkaline granites and quartz syenites; and 3) alkaline nepheline syenites. The sodalite-rich nepheline syenite naujaite occurs under the roof whereas the kakortokites are in the lower part of the complex. The lujavrites are the youngest, forming a horizon between the naujaites and kakortokites. However, the lujavrite does not form a uniform unit, but rather penetrates the naujaite and appears to have flowed around the edges of the major naujaite unit, especially in the northern part of the complex (Ussing, 1912; Ferguson, 1964; Marks and Markl, 2015). Descriptions of the most important agpaitic rocks

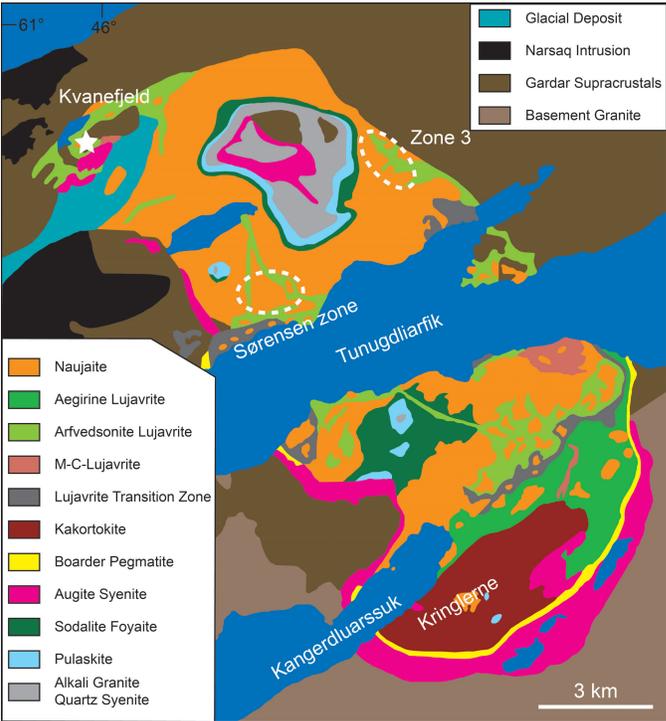


Fig. 1. Geological map of the Ilímaussaq alkaline complex. Modified after Sørensen (2001) and GME (2015).

Table 1. Simplified formulae of selected Ilimaussaq minerals.

Britholite group	$\text{Ca}_2(\text{REE}, \text{Ca})_3(\text{SiO}_4)_3(\text{OH}, \text{F})$
Catapleiite	$\text{Na}_2\text{ZrSi}_3\text{O}_9 \cdot 2\text{H}_2\text{O}$
Eudialyte s.s.	$\text{Na}_{15}\text{Ca}_6\text{Fe}_3\text{Si}(\text{Si}_{25}\text{O}_{73})(\text{O}, \text{OH}, \text{H}_2\text{O})_3(\text{Cl}, \text{OH})_2$
Gittinsite	$\text{CaZrSi}_2\text{O}_7$
Lovozerite s.s.	$\text{Na}_3\text{Ca}_3(\text{Ca}, \square)_2\text{Zr}(\text{Si}_6\text{O}_{12})\text{O}_3(\text{OH})_3$
Nacarenioibsite-(Ce)	$\text{Na}_3\text{Ca}_3\text{REENb}(\text{Si}_2\text{O}_7)\text{OF}_3$
Natrophosphate	$\text{Na}_7(\text{PO}_4)_2\text{F} \cdot 19\text{H}_2\text{O}$
Natrosilite	$\text{Na}_2\text{Si}_2\text{O}_5$
Naujakasite	$\text{Na}_6(\text{Fe}, \text{Mn})\text{Al}_4\text{Si}_8\text{O}_{26}$
Phosinaite-(Ce)	$\text{Na}_{13}\text{Ca}_2\text{REE}(\text{Si}_4\text{O}_{12})(\text{PO}_4)_4$
Rinkite	$(\text{Na}, \text{Ca})_3(\text{Ca}, \text{REE})_4\text{Ti}(\text{Si}_2\text{O}_7)\text{OF}_3$
Steenstrupine-(Ce)	$\text{Na}_{13}\text{REE}_6\text{Mn}_2(\text{Fe}^{3+}, \text{Mn}^{3+})_2(\text{Zr}, \text{Th})(\text{Si}_6\text{O}_{18})_2(\text{PO}_4)_7 \cdot 3\text{H}_2\text{O}$
Townendite	$\text{Na}_8\text{ZrSi}_6\text{O}_{18}$
Ussingite	$\text{Na}_2\text{AlSi}_3\text{O}_8\text{OH}$
Villiaumite	NaF
Vitusite-(Ce)	$\text{Na}_3\text{REE}(\text{PO}_4)_2$
Vuonnemite	$\text{Na}_{11}\text{TiNb}_2(\text{Si}_2\text{O}_7)_2(\text{PO}_4)_2\text{O}_3(\text{F}, \text{OH})$

s.s.: sensu stricto. Minerals in bold were first described from Ilimaussaq.

related to critical materials are presented below.

2.1. Kakortokite

Kakortokite is a medium- to coarse-grained nepheline syenite and forms the main magmatic layered part of the intrusion in the southern part of the complex (Fig. 2a). Most of the 29 described units consist of three layers, which are named on the basis of colour. Basal black layers of predominantly arfvedsonite are followed by red layers rich in eudialyte, then

by white layers rich in nepheline and alkali-feldspar, which are generally the thickest (Bohse et al., 1971). The layers maintain a consistent thickness throughout a unit. Although most of the units contain all three layers, some do not. Black layers have abrupt lower contacts with subjacent white layers, but typically pass gradationally upwards to red layers (Fig. 2b). The units are labelled based on their position with respect to a marker unit called zero (e.g., unit +16 is the 16th unit above the marker; unit -3 is the 3rd unit below the marker). The typical thickness



Fig. 2. Rhythmic, metre-scale magmatic layering of kakortokite. **a)** Sharp-based basal black layers of predominantly arfvedsonite grade upward to red layers rich in eudialyte, which are overlain by relatively thick white layers rich in nepheline and alkali-feldspar. **b)** Close up illustrating abrupt basal contact between black layer 0 and underlying white layer -1, and gradational transition between black layer 0 and overlying red layer 0.

of the red layers is about 1 m, but red +16 is a particularly rich eudialyte zone 3.5 m thick (Bohse et al., 1971). Although eudialyte is predominant in the red layers, it is also present in the other layers.

2.2. Naujaite

Naujaite is a cumulate rock consisting of large (up to 5 mm) euhedral sodalite crystals that floated to the top of the magma chamber. Later crystallising phases are mainly nepheline, alkali-feldspar, aegirine, arfvedsonite, and eudialyte, which result in a predominantly poikilitic texture. Individual feldspars can be up to 25 cm, and both aegirine and arfvedsonite can form crystals up to 30 cm (Ussing, 1912). The amount of eudialyte in naujaite is inconsistent and it may be completely absent. Rinkite is a common accessory mineral in naujaite, which is the most exposed rock in Ilímaussaq. The naujaites are not considered potential ores for critical materials.

2.3. Lujavrite

Lujavrite is a meso- to melanocratic agpaitic to hyperagpaitic syenite with a pronounced lamination caused by the orientation of mafic minerals and, in part, felsic minerals such as feldspars. The two most abundant varieties of lujavrite in Ilímaussaq are named on the basis of colour: in green lujavrite the predominant mafic mineral is aegirine; in black lujavrite, the main mafic mineral is arfvedsonite. The felsic minerals in lujavrite are nepheline, albite, microcline, and sodalite. Lujavrites are fine grained (up to 0.6 mm), but sodalite grains can be up to 2 mm and mafic minerals up to 1 mm. A coarser type of lujavrite is called m-c-lujavrite (m-c stands for medium to coarse grained), in which the individual grains can reach sizes of more than one cm, and are locally pegmatitic. A fourth type, naujakasite lujavrite, contains naujakasite and is associated with the highly agpaitic stage of the complex. In these lujavrites, naujakasite generally occurs at the expense of nepheline (Andersen and Sørensen, 2005). Although naujakasite has been found only in Ilímaussaq, it is common; locally the lujavrite contains up to 75 % volume naujakasite. The water-soluble mineral villiaumite is abundant in most of the lujavrites but has been leached out in some near-surface rocks. Lujavrite is the rock unit containing the highest amounts of incompatible elements and is the major ore of the Kvanefjeld deposit (Bailey et al., 2001).

3. Exploration history

The first exploration activity in Ilímaussaq was initiated by The Cryolite Company in 1888 and, in 1899, K.J.V. Steenstrup mined eudialyte in the southern part of the complex. A total of 60 t of eudialyte was mined for zirconium extraction experiments (Sørensen, 1966, 1968). In 1968, the Danish company Dansk Svovlsyre- og Superphosphat-Fabrik A/S (now Superfos) extracted approximately 100 t of eudialyte ore from the kakortokites. The renewed interest in zirconium and niobium led to detailed investigations and mapping of the kakortokites. Since the work by Superfos, several minor exploration companies have held the exploration licence to

this part of the complex. Tanbreez is the current licence holder and they report an inferred resource of 4.3 billion tonnes of eudialyte ore with an average grade of Zr-oxide (1.8%), Nb-oxide (0.2%), light rare earth elements (LREE) (0.5%) and heavy rare earth elements (HREE) (0.15%) (Tanbreez, 2015).

In the wake of the Second World War, demand for uranium led to a uranium-focused survey of Greenland. In the 1950s, the Kvanefjeld deposit was discovered after a radiometric survey. The ore consists of several types of lujavrite. Material with the highest radioactivity was collected to test extraction via conventional acid and carbon leach methods, but the ore was found to be refractory (Sørensen, 2001). A drilling program was started in 1958, and in 1962, 180 t of ore from a 20 m adit was extracted (Fig. 3). The ore was used to test sulphatizing roasting for extracting uranium and thorium at the Danish National laboratories (RISØ). In 1978, the 'Kvanefjeld Uranium Project' was initiated to examine the economic potential of a uranium mine at Kvanefjeld. In 1980-81, a 1 km adit was made at Kvanefjeld (Fig. 3). Each blast was checked with a scintillometer, and high-grade ore was shipped to RISØ where a 1:10-scale pilot plant had been constructed. A total of 4700 metric tons of ore were processed by pressurized carbonate leaching, which resulted in recoveries of up to 80% (Sørensen, 2001). The Kvanefjeld project was terminated in 1983 when a political decision was made for Denmark to maintain its status as a nuclear-free zone. At the end of the project, 69 holes had been drilled with a total core length of more than 10 km, and 20,000 tonnes of ore had been mined.

In 2007, Kvanefjeld received renewed interest as a multi-element deposit. Greenland Minerals and Energy Ltd. (GME) set up a drilling program and metallurgical studies. In addition to U, Th, and rare earth elements (REE), the deposit contains significant Zn in the form of sphalerite. The new drilling program at Kvanefjeld consisted of 146 holes with a total of 35.5 km of core. The feasibility report defined a measured resource of 143 Mt @ 303 ppm U_3O_8 , 1.2% TREO, and 0.24% zinc (GME, 2015). Greenland Minerals and Energy Ltd. also discovered two additional lujavrite-hosted deposits (the

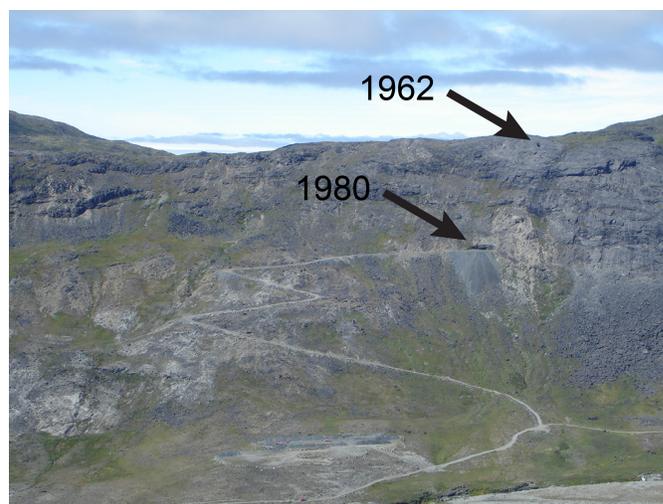


Fig. 3. View of Kvanefjeld showing the 1962 and 1980 adits.

Sørensen zone and Zone 3) north of the Tunugdliarfik (Fig. 1). The Sørensen zone has an inferred resource of 119 Mt @ 400 ppm U_3O_8 , 1.2% TREO, and 0.3% zinc, and Zone 3 an inferred resource of 47 Mt @ 358 ppm U_3O_8 , 1.2% TREO, and 0.3% zinc (GME, 2015).

4. Primary ore minerals

Eudialyte is concentrated in specific magmatic layers in the kakortokite. In contrast, steenstrupine-(Ce) is distributed throughout the lujavrites, making all arfvedsonite lujavrite a potential ore. Steenstrupine-(Ce) and lovozerite group minerals are the main target minerals in the lujavrites. In addition to steenstrupine-(Ce) and lovozerite group minerals, the lujavrites may contain significant amounts of other primary REE-minerals such as vitusite-(Ce) and phosinaite-like minerals (Pekov and Ekimenkova, 2001; Friis, Unpublished data). Below is a description of the three most important primary ore minerals in the Ilímaussaq complex.

4.1. Eudialyte

Eudialyte was described as a new species from Ilímaussaq by Stromeyer (1819). It occurs in all of the main apgaitic rock units, but is particularly abundant in the kakortokites, where it typically forms euhedral crystals that are generally less than 5 mm, but locally reach several cm. In pegmatites, massive eudialyte can form over several metres and be up to 50 cm thick. Pfaff et al. (2008) showed a decrease in the Fe/Mn ratio of eudialyte when moving from unit zero through the units to +16 and that the trend continued into the lower lujavrites. Pfaff et al. (2008) further showed that the Na-content in the eudialyte increases up section in the kakortokites, but that the increase in Na is more pronounced for eudialytes in the aegirine lujavrites. This is consistent with the observation that Na content in eudialyte from the aegirine lujavrites at Kvanefjeld is higher than Na in eudialyte in other lujavrites from the same locality (Friis, 2012; Andersen and Friis, 2015).

4.2. Steenstrupine-(Ce)

Steenstrupine-(Ce) was first described as a new species from Ilímaussaq by Lorenzen (1881) from material collected by Steenstrup. Steenstrupine-(Ce) is widespread in pegmatites and ussingite veins in the naujaite, and is a rock-forming mineral in many arfvedsonite-lujavrites. Steenstrupine-(Ce) has not been observed in the kakortokites and generally does not coexist with eudialyte. Steenstrupine-(Ce) is a phosphosilicate, which is a group of minerals characteristic of hyperagpaitic environments (Khomyakov, 1995). The chemical composition of steenstrupine-(Ce) is complex and can vary significantly. For example, Makovicky and Karup-Møller (1981) showed that practically all Na may be replaced by H^+ owing to large channels in the structure (Moore and Shen 1983). The structure of steenstrupine-(Ce) contains several different coordinations, which enable the incorporation of various elements or elements in different valence states (Makovicky and Karup-Møller, 1981). For example, steenstrupine-(Ce) from the Kola

Peninsula has both divalent and trivalent Fe (Malczewski et al., 2005). High Th (up to 5 wt.% oxides; Makovicky and Karup-Møller, 1981; Khomyakov and Sørensen, 2001) and U (0.2-1% U_3O_8 ; GME, 2015) results in most steenstrupine-(Ce) being metamict and hence challenging to analyze.

The steenstrupine-(Ce) crystals in the lujavrites are typically euhedral and up to 0.6 mm. Each lujavrite may contain several types of steenstrupine-(Ce), such as inclusion-free metamict, partly altered, anisotropic rim, or poikilitic varieties (Fig. 4). A general correlation between different types of steenstrupine-(Ce) in a sample seems to be lacking, except for the poikilitic type, which is always present with at least one other type. Typical inclusions are arfvedsonite, lovozerite group minerals, and britholite group minerals. Khomyakov and Sørensen (2001) investigated steenstrupine-(Ce) with anisotropic rims and found that the rims have a lower Th and higher REE than the cores (1.22 vs. 4.37 wt.% ThO_2 and 33.15 vs. 30.00 wt.% REE_2O_3). Although steenstrupine-(Ce) is a rare mineral, only described in minor amounts at a few other alkaline complexes, it is abundant in some of the lujavrites, making it a rock-forming mineral with a modal abundance of up to 10% (Makovicky et al., 1980).

4.3. Lovozerite

Lovozerite group minerals in Ilímaussaq were first described in a few samples from the Kangerdluarssuk Fjord where they occur associated with ussingite and steenstrupine-(Ce) (Danø and Sørensen, 1959). Lovozerite was considered a rare mineral in Ilímaussaq until a new member of the lovozerite group (townendite) was discovered in flotation tails (Grey et al., 2010). Since the discovery of townendite, lovozerite group minerals have been observed in many lujavrite samples. Three types have been observed: 1) colourless (in plane polarized light) euhedral grains up to 0.1 mm, with a composition close to that of lovozerite *sensu stricto*; 2) pink euhedral to subhedral grains that have higher Mn, Y, and U contents than the colourless type, but still classify as lovozerite *sensu stricto*; and 3) brown grains that show clear evidence of alteration. The composition of the latter type is varied and probably reflects the alteration rather than a true lovozerite group composition (Friis, 2012). The lovozerite crystals typically occur on grain boundaries or partly to fully embedded in arfvedsonite and steenstrupine-(Ce). Because of the relative abundance of lovozerite in the lujavrite and its U content, it is an important ore mineral and its presence could explain the low recovery of U from ore during tests in the 1980s.

5. Alteration

Most of the rocks in Ilímaussaq show signs of alteration, which varies from thin zones along fractures and grain boundaries to complete replacement of all primary minerals. The replacements are not necessarily related to external fluids, but may have resulted from exsolution of fluids, in the final stages of crystallization, that reacted with previously formed minerals. Evidence for such an alteration has been observed

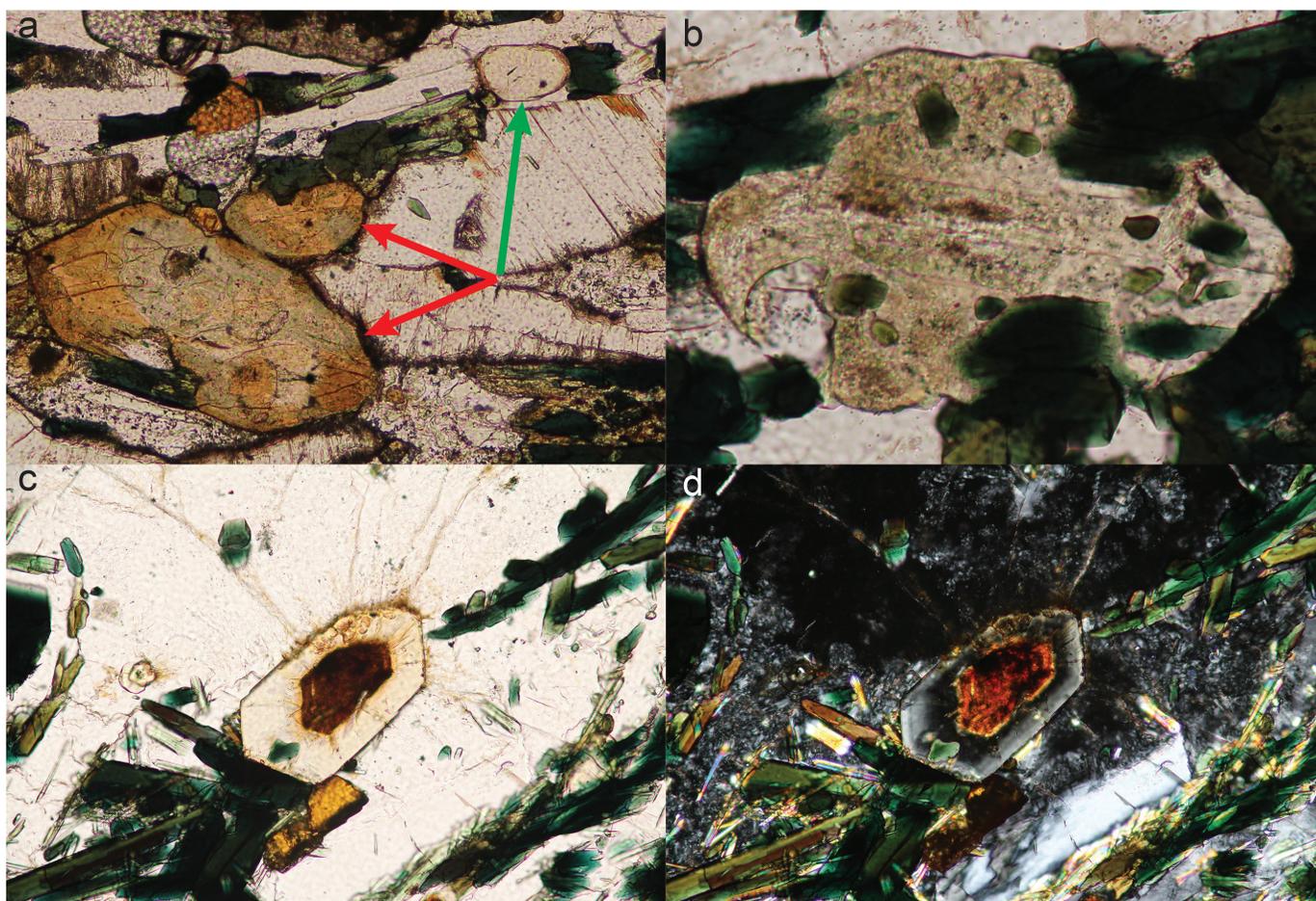


Fig. 4. Photomicrographs of steenstrupine-(Ce) in lujavrites from Kvanefjeld. **a)** Two altered crystals (red arrows) of which the larger is 0.5 mm. The smaller crystal (green arrow) is a metamict steenstrupine-(Ce). **b)** A 0.3 mm poikilitic steenstrupine-(Ce) crystal with inclusions of arfvedsonite. **c+d)** A 0.4 mm steenstrupine-(Ce) crystal with an altered core and anisotropic rim.

in many black lujavrites, which contain isolated spheroidal areas where the primary minerals have been replaced. The alteration is easily identified because the rock has patches of lighter green where the arfvedsonite is replaced by aegirine (Sørensen et al., 2003). In rocks of this type, feldspars and feldspathoids typically alter to natrolite or analcime. Secondary feldspar and sodalite may also be alteration products (Markl et al., 2001). Alteration in Ilímaussaq was produced under high pH conditions (Markl and Baumgartner, 2002) as indicated by ussingite in some zones and in late magmatic veins.

The alteration of HFSE minerals in Ilímaussaq is characterised by redistribution of the HFSE rather than by additions or removals. The most common Zr-alteration product after eudialyte is catapleiite, but locally zircon may form (Ussing, 1912; Karup-Møller et al., 2010; Borst et al., 2015). Borst et al. (2015) describe a third alteration, where late-stage fluids interacted with already formed catapleiite, which then altered to gittinsite. The REE liberated by decomposition of eudialyte typically form separate phases (e.g., nacareniobsite-(Ce) and fergusonite-(Y)) preferentially rich in either LREE or HREE (Karup-Møller et al., 2010). The same is the case for the alteration of steenstrupine-(Ce), where REE fractionate

into monazite-(Ce) and xenotime-(Y), whereas thorite is the most common secondary U-Th mineral. What is characteristic for the alteration of both steenstrupine-(Ce) and eudialyte is that the REE are contained within the grain boundary of the original crystal; evidence of these elements leaving the system is lacking. The fractionation mechanism of REE into mainly LREE and Y+HREE phases was further explored by Friis (2015) who illustrated that all the known Y-minerals in Ilímaussaq are alteration products. The alteration of eudialyte releases Cl and the alteration of villiaumite-rich lujavrite releases F, elements that both can form complexes and could transport, the REE. Secondary Cl minerals are uncommon, supporting the hypothesis that Cl leaves the system during alteration. However, Migdisov and Williams-Jones (2014) showed that at high pH, fluorine and chlorine complexes with REE form solid phases. Borst et al. (2015) and Friis (2015) explain the low mobility of HFSE during alteration in Ilímaussaq as a result of the high pH (>9; Markl and Baumgartner, 2002). Therefore, fluorine and chlorine complexes are unlikely to have transported REE over large distances during alteration of the Ilímaussaq rocks.

Alteration of the Ilímaussaq rocks changes only the mineralogy, which influences the ore processing, but not

grade. This change in mineralogy poses challenges to planning a mining project. When exploring for critical materials in complex alkaline environments, detailed mineralogical studies are essential because alterations may not be easily derived from assays.

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