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Eudialyte decomposition minerals with new hitherto undescribed phases from the Ilímaussaq complex, South Greenland

S. KARUP-MØLLER, J. ROSE-HANSEN & H. SØRENSEN

Eudialyte is a distinctive mineral in the agpaitic group of peralkaline nepheline syenites. The paper describes the alteration of eudialyte from the marginal pegmatite of the Ilímaussaq complex, South Greenland, which is the type locality for eudialyte, as well as for agpaitic rocks.

Two types of alteration are distinguished: alteration of eudialyte to catapleiite is widespread in the complex, whereas alteration to zircon only occurs in strongly altered rocks. Additionally, the following minerals have been identified in the altered eudialyte: aegirine, K-feldspar, albite, analcime, fluorite, monazite, apatite, allanite, Y-fergusonite, Ce-fergusonite, fersmite, nacareniobsite-(Ce), uranothorianite, neptunite and grossularite. Some unidentified Nb and/or REE minerals have been distinguished.

The different styles of alteration can be explained by different late- and post magmatic conditions at different settings in the complex. Alteration to zircon is caused by fluids of external origin or by fluids expelled from intersecting lujavrite and pegmatite dykes. Alteration to catapleiite is caused by late-magmatic interstitial fluids. The alteration has resulted in distinct fractionation of some elements; Zr is, for instance, located only in zircon and catapleiite, Nb and REE in separate minerals.

Keywords: límaussaq complex, eudialyte decomposition products, marginal pegmatite, kakortokites, unidentified Nb- and REE-minerals.

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Introduction

The Ilímaussaq alkaline complex is the type locality of agpaitic nepheline syenites (Ussing 1912) as well as of eudialyte (Stromeyr 1819). Agpaitic rocks are per-alkaline nepheline syenites containing complex minerals such as eudialyte instead of simple minerals such as zircon.

The Ilímaussaq complex consists of four main intrusive phases. Phase 1 is a partial rim of augite syenite, phase 2 is alkali granite and quartz syenite in the roof zone of the complex, phase 3 consists of pulaskite, foyaite, sodalite foyaite and naujaite and phase 4 of kakortokites and lujavrites (e.g. Rose-Hansen & Sørensen 2002, Sørensen 2006, Sørensen et al. 2006).

A unit called marginal pegmatite (Ussing 1912, Westergaard 1969, Bohse et al. 1971, Steenfelt 1972) consists of agpaitic nepheline syenite and short pegmatitic veins. It forms a rim between the country rocks and the kakortokite and the lowermost lujavrites of the complex and between country rocks and naujaite in the upper part of the complex (Sørensen 2006).

The matrix of the marginal pegmatite is generally a massive-textured, homogeneous agpaitic nepheline syenite, which is locally layered. It consists of K-feldspar, nepheline, eudialyte, aegirine, arfvedsonite and locally sodalite. Where it is in contact with the lowermost lujavrites it is thoroughly altered, K-feldspar and nepheline being replaced by analcime and pigmentary material/sericite, and arfvedsonite and green aegirine by brown aegirine (Sørensen 2006).

The pegmatitic veins, which are a few metres long and up to about 0.5 m wide, consist of microcline, albite, nepheline, sodalite, eudialyte, aegirine, arfvedsonite, aenigmatite, biotite, rinkite, astrophyllite,

tundrite, lithium mica and fluorite (Bohse et al. 1971).

Eudialyte in the matrix of the marginal pegmatite is altered in various degrees to zircon or catapleiite and a host of other minerals that are described in the present paper. Selected analyses of eudialyte completed during the present study and by other scientists are listed in Table 4.

Alteration of eudialyte

Ussing (1898, 1912) gave a detailed description of the eudialyte from the Ilímaussaq complex and identified alteration of this mineral to zircon, and more commonly to catapleiite. This observation was confirmed by Bøggild (1953) who also described other types of alteration of eudialyte from the Ilímaussaq complex and the adjacent Igaliko complex. Sørensen (1962) described the alteration of eudialyte to catapleiite and noted that in some lujavrites, eudialyte was substituted by steenstrupine. Semenov (1969) described alteration of eudialyte to a yellowish powder of an amorphous substance known under the name zirfes-

ite (Kostyleva 1945) and suggested the general formula $Fe_2Zr_3Si_4O_{17} \cdot 15H_2O$ for the Ilímaussaq variety. Zirfesite has later been discredited as a mineral name. Sørensen & Larsen (2001) distinguished agpaitic eudialyte-bearing lujavrites and steenstrupine-bearing hyperagpaitic lujavrite varieties. Sørensen (2006) noted that eudialyte was substituted by zircon in the most altered parts of the marginal pegmatite. These studies were carried out by traditional microscopy and chemical analyses of separated minerals. See Table 4.

The chemical composition of eudialyte shows no significant changes throughout the kakortokite series (Bohse et al. 1971, Steenfelt & Bohse 1975, Kogarko et al. 1988, Sørensen 1992, Pfaff et al. 2008) and our analyses (Table 4).

Examples of investigation of eudialyte alteration by electron microprobe analyses are presented by Coulson (1997) from the Igaliko complex and by Mitchell & Liferovich (2006) from the Pilansberg complex, South Africa. This is also the method applied in our examination of minerals from the marginal pegmatite since the grain size of the alteration products is generally less than 10 micrometers in diameter.

Table 4. Average compositions of unidentified minerals from altered eudialyte in the marginal pegmatite of the Ilímaussaq complex

Mineral	A1	A2	Nb1	Nb1	Nb2	Uk1	Uk2
Locality	A,D,B,C	B,C	C	C	C	C	C
Element							
SiO_2	22.97	26.62	10.84	10.66	n.d.	3.68	n.d.
ZrO_2	0.16	n.d.	0.11	0.04	n.d.	n.d.	n.d.
TiO_2	n.d	n.d.	0.14	0.19	2.40	n.d.	n.d.
ThO_2	0.06	n.d.	0.05	0.06	n.d.	n.d.	n.d.
Nb_2O_5	0.06	n.d.	31.71	32.50	76.72	n.d.	n.d.
P_2O_5	0.63	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Al_2O_3	n.d.	n.d.	4.45	4.14	n.d.	2.22	n.d.
Ce_2O_3	31.71	16.04	16.21	8.35	0.06	35.91	37.07
Y_2O_3	1.24	3.79	3.06	10.78	0.17	0.01	0.08
La_2O_3	16.02	1.80	5.29	2.72	n.d	12.28	23.24
Nd_2O_3	11.60	20.04	10.55	7.34	0.03	9.99	8.44
Pr_2O_3	2.89	3.31	1.96	1.15	n.d	3.50	2.53
Sm_2O_3	2.04	5.94	2.06	2.42	n.d	0.83	0.73
Gd_2O_3	0.88	3.97	1.43	2.57	n.d	n.d	0.18
FeO	1.14	n.d.	5.23	5.75	0.32	0.95	n.d.
MnO	0.47	n.d.	0.30	0.24	20.96	0.50	n.d.
CaO	5.65	1.17	4.15	4.79	0.12	5.63	0.39
Na_2O	0.35	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
F	1.09	0.16	n.d.	n.d.	n.d.	6.05	6.77
Total	98.96	82.84	97.54	93.70	100.78	81.55	79.43

Description of the geological setting of the investigated localities

The examined material was collected at four localities (Fig. 1) within the marginal pegmatite that have been petrographically described by Sørensen (2006).

The alteration to catapleiite was studied at locality A (GI.104380) from the Kvanefjeld area in the northernmost part of the complex, and at locality D (GI. 104563) from near the south-west contact of the complex. Alteration to zircon was described at locality B (GI. 104361) from the coast of Kangerdluarsuq and locality C (GI. 109302 and 109303) from the north coast of Tunulliarfik.

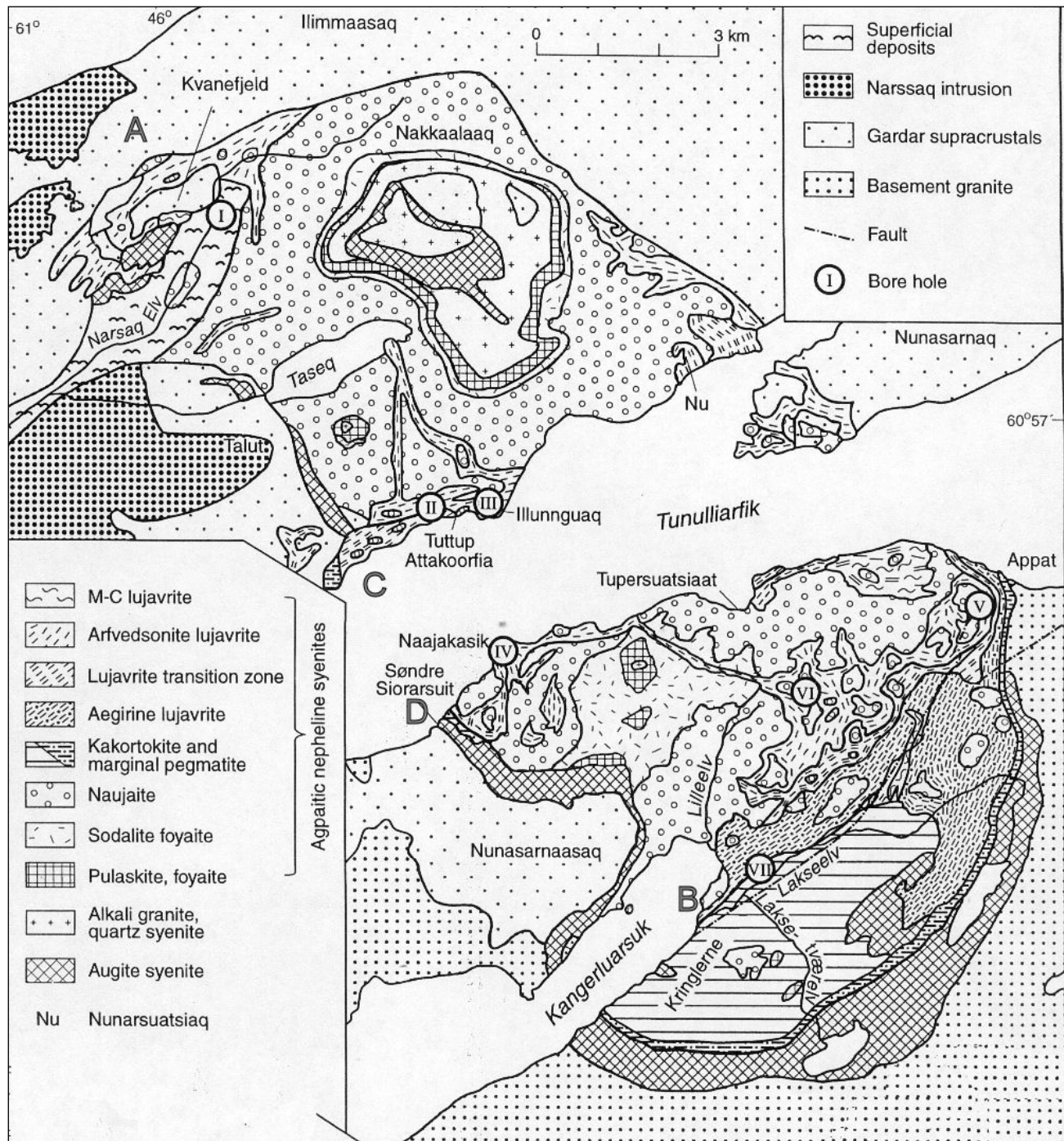At locality A, the marginal pegmatite is the mar-

Fig. 1. Simplified geological map of the Ilímaussaq complex based on Ussing (1912), Ferguson (1964), Andersen et al. (1988) and ongoing studies by the "Ilímaussaq group".

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ginal facies of the naujaite and forms a partial rim between this rock and the basaltic country rocks. Its matrix is poikilitic, similar to the texture of naujaite, but with massive-textured patches similar to the matrix of the marginal pegmatite from localities B and C. It is locally rich in eudialyte. We have examined the macroscopically massive-textured matrix variety which consists of microcline, nepheline, arfvedsonite, aegirine and eudialyte. Thin section studies reveal that arfvedsonite, aegirine and aggregates of eudialyte crystals poikilitically enclose nepheline and aegirine crystals. The eudialyte is practically unaltered.

At locality D the marginal pegmatite passes inwards into kakortokite, but has sharp contacts against the external margin of augite syenite. Masses of naujaite occur between the marginal pegmatite and the augite syenite. The sample examined is a small lenticular body of a rather fine-grained, massive-textured rock which forms a pocket in a pegmatite and is interpreted as a sample of quenched pegmatitic melt (Sørensen 2006). It is rich in eudialyte which, because of the sheltered location of the body, is only slightly altered. It belongs to a lower stratigraphic level of the complex than the pegmatite at the other three localities.

Localities B and C are both located between the lower part of the lujavrite sequence and its country rocks. At locality B, the country rocks are the augite syenite and outside it the granitic basement; at locality C, the volcanic country rocks. At both places, eudialyte, nepheline, arfvedsonite and aegirine are strongly altered. Alteration at locality B may be related to the many intersecting thin lujavrite dykes, at C to swarms of parallel pegmatite veins. The microcline is partly substituted by albite, the nepheline by sericite and analcime, the arfvedsonite by aegirine, and the eudialyte by zircon.

The matrix rock at locality C displays the same features as the examined rock at locality B, but is slightly less altered.

Mineralogy of the altered eudialyte

Methods

Electron microprobe analyses were carried out at the Department of Geography and Geology, University of Copenhagen, using a JEOL 733 superprobe in wavelength dispersive mode with an online correction program supplied by JEOL. The excitation voltage was 15 kV, beam current 15 nA and beam diameter 1 micron. Wavelengths and standards used were Na K

(albite), K K (KAlSi$_3$O$_8$), Ca K and Si K (CaSiO$_3$), Mn K (MnTiO$_3$), Fe K (Fe$_2$O$_3$), Al K (Al$_2$O$_3$), Ce L (CeO$_2$), La L (La-18), Nd L (Nd$_3$Ga$_5$), Pr L (Pr$_3$Ga$_5$O$_{12}$), Sm L (SmFeO$_3$), Y L (Y$_3$Al$_5$O$_{12}$), Gd L (GdFeO$_3$), P L and F K (apatite-wilberforce), Nb L (columbite), Th M (ThO$_2$), Cl K (sodalite). The estimated detection limit was 0.02 wt.% for all elements. Up to 10 analyses were completed on each phase in a given eudialyte alteration aggregate. Figures 2-12 are all BSF images.

Paragenesis

Table 1 presents the minerals identified during the present study, their simplified formulae and references to localities and tables with compositional data. The common rock-forming minerals are not listed.

The Nb and the REE minerals generally occur as small rounded opaque or semi-transparent grains. The grain size rarely exceeds 10 microns. This excludes X-ray analytical work and optical examination in transmitted and reflected light. The minerals have been examined using the electron microprobe. Some of the observed minerals may be new mineral species, but insufficient mineralogical data prevent the proposal of new names for these minerals.

The catapleiite alteration type

At localities A and D, eudialyte is altered to extremely fine-grained aggregates of catapleiite and a number of other minerals: aegirine, analcime, minor amounts
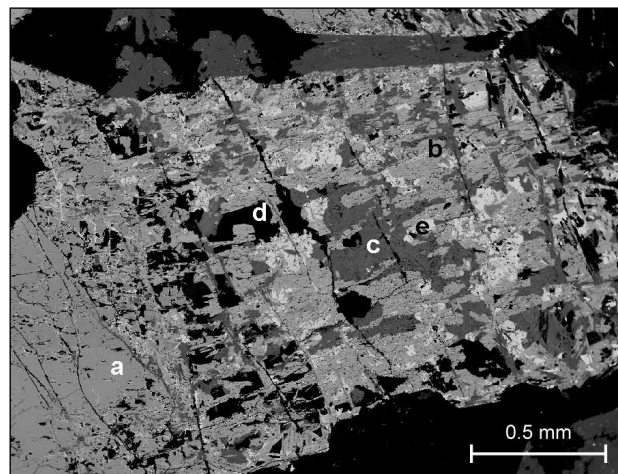

Fig. 2. A eudialyte crystal (a, grey, left part of the photo) has been completely altered (central and right part of the figure) into an aggregate of zircon (b, same grey shade as the unaltered eudialyte), aegirine (c, dark grey), analcime (d, black) and a heterogeneous mixture of fluorite, Ce-fergusonite, monazite and A1 (e, small pale grains with a random distribution). These four minerals cannot be distinguished from each other in the photograph. The boundary between altered and unaltered eudialyte is rather well defined. (Sample GI 104380, locality A).

of fluorite, Y-fergusonite, apatite, monazite, nacareniobsite-(Ce), thorianite. neptunite and A1. A typical aggregate is pictured in Fig. 2.

The zircon alteration type

At localities B and C, eudialyte is altered to zircon, K-feldspar, albite, aegirine, fluorite and grossularite, together with small quantities of monazite, Ce-fergusonite, allanite, A1 and A2, and at locality C also Y-fergusonite, fersmite, Nb1, Nb2, Uk1 and Uk2. Coexisting catapleeite and zircon is rare and has only been observed at locality C.

A representative eudialyte decomposition aggregate is shown in Figs. 3, 4 and 5. Fig. 5 covers the left part of the aggregate in Fig. 4. The outlines of the original eudialyte crystal are marked by broken strings of very small zircon crystals. Zircon is always distinctly zoned and forms irregularly-shaped patches (Fig. 5). Euhedral zircon crystals have also been observed. Besides zircon, the aggregate is composed of microcline, aegirine, fluorite and monazite. An isolated monazite cluster close to the aggregate is shown in Fig. 6.

The aggregate in Fig. 7 is composed of K-feldspar, aegirine, zircon, Ce-fergusonite and allanite. On the

Table 1. Electron microprobe analyses (XRF and INA: IG 109269 and D7-34.5 m) of eudialyte and zirfesite from the Ilímaussaq complex

	Eudialyte	Eudialyte	Eudialyte	Eudialyte	Eudialyte	Eudialyte	Zirfesite
	Kakortokite IM 109269 Layer-9, red Bohse pers. com	Naujaite D7-34.5 m # Bohse pers. com	Johnsen & Gault 1997	Kakortokite Layer +1 This paper	Kakortokite Layer +9 This paper	Kakortokite Layer +16 This paper	Semenov 1961
SiO_2	48.90	49.01	48.82	49.76	49.66	50.07	31.75
ZrO_2	13.40	13.29	11.65	11.76	11.78	11.79	21.08
ThO_2	n.d	n.a	n.d	0.02	0.02	0.01	
TiO_2	0.06	0.07	0.00	0.09	0.10	0.09	
Nb_2O_5	0.91	1.09	0.77	0.67	0.66	0.67	0.79
La_2O_3	0.34	*0.30	0.47	0.64	0.47	0.48	
Ce_2O_3	*0.77	*0.66	0.90	1.44	0.95	0.97	
Nd_2O_3	*0.32	*0.26	0.41	0.61	0.39	0.41	
Pr_2O_3	*0.09	*0.08	n.a	0.19	0.13	0.13	
Sm_2O_3	*0.06	*0.05	n.a	0.18	0.11	0.13	
Eu	*0.09	n.a.	n.a	n.a.	n.a	n.a.	
Gd_2O_3	*0.07	0.06	n.a	0.17	0.06	0.14	
Y_2O_3	*0.42	0.28	0.44	0.74	0.46	0.47	
Al_2O_3	0.38	0.68	0.50	0.19	0.24	0.24	1.23
Fe_2O_3							11.30
Total Fe as FeO	6.21	6.08		5.29	6.05	6.20	
MnO	0.85	1.07	0.47	1.16	0.60	0.65	
MgO	n.a	0.03	n.a	n.a	n.a	n.a	
CaO	9.87	10.48	9.61	9.82	10.16	9.63	3.01
Na_2O	13.26	13.24	15.46	13.27	14.87	15.34	
K_2O	0.39	0.25	0.36	0.57	0.26	0.27	
P_2O_5	n.a	n.a	n.a	0.00	0.01	0.01	
Cl	1.28	1.33	1.34	1.07	n.a	1.34	
F	n.a	n.a	n.a	0.03	0.06	0.05	
H_2O	n.a.		0.22	n.a	n.a	n.a.	15.56
HfO_2	0.23	n.a	0.58	n.a	n.a	n.a	
Ta_2O_5	0.05	0.06	n.a	n.a	n.a	n.a	
RE_2O_3							8.03
Total	97.95	98.37	92.00	97.67	97.04	99.09	92.75

* INAA analyses of separated minerals; # D7= drill core 7; n.a = not analysed. Electron microprobe analyses

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left, patchily decomposed Ce-fergusonite lies isolated in analcime (Fig. 8). In the lower central part of Fig. 7, zircon is intergrown with allanite and two small grains of the unidentified mineral Uk1 (Fig. 9). In the bottom right part of Fig. 7, allanite crystals, which are associated with zircon (Fig. 10), contain inclusions of Nb1. This mineral also occurs as disseminated grains in K-feldspar in the central part of the aggregate in Fig.7.

The Y-fergusonite occurs in some aggregates as randomly distributed grains, in places as inclusions in Nb2 (Fig. 11). The Ce-fergusonite was found in three aggregates and A1 and A2 in two. Allanite occurs as aggregates of radiating crystals. Fersmite was found

in association with zircon and grossularite. The presumed new mineral Uk2 was found in association with Ce-fergusonite (Fig. 12). The Uk2 grains in Fig.12 are embedded in microcline and a mixture of calcite, a mineral with the proposed composition $Fe_6Mn_6Al_7Si_8O_{31.5-y}(OH)_y \times nH_2O$ and a mineral in such small grains that it could not be analyzed quantitatively.

The minerals

Selected and/or average compositions of identified and unidentified, possibly new minerals are listed in
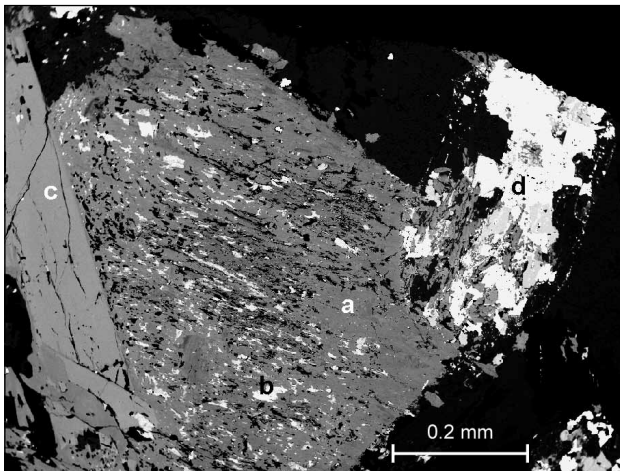

Fig. 3. The central part of the photo is composed of aegirine (a, grey) with numerous small inclusions of mainly hematite (b, white) after an original, now completely decomposed arfvedsonite crystal. To the left a primary aegirine lath (c) and to the right an eudialyte alteration aggregate (d). The latter has been enlarged in Fig. 4. (Sample GI 104361, localilty B).
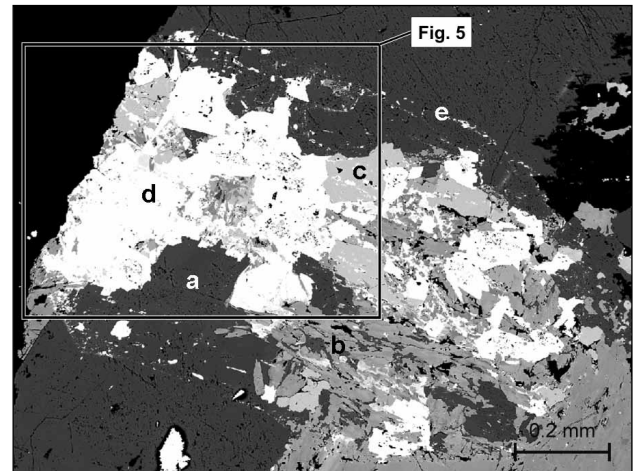

Fig. 4. Enlarged portion of Fig. 3 covering the eudialyte alteration aggregate. The aggregate is composed of microcline (a, dark grey), aegirine (b, grey), fluorite (c, light grey), monazite and zircon (d, both white). The left part of the aggregate is enlarged in Fig. 5. Very small zircon grains (e) mark the outline of the original eudialyte crystal. (Sample GI 104361).
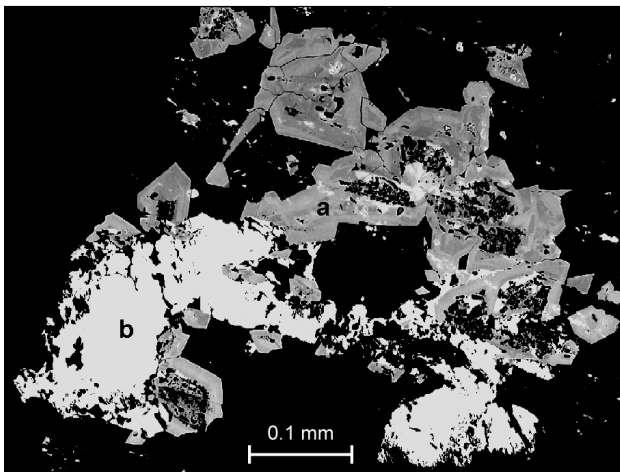

Fig. 5. In this image, the left portion of Fig. 4 is shown using a relatively low electron flux. Here (in contrast to Fig. 4) zoned zircon (a, grey) is easily distinguished from monazite (b, white). (Sample GI 104361).
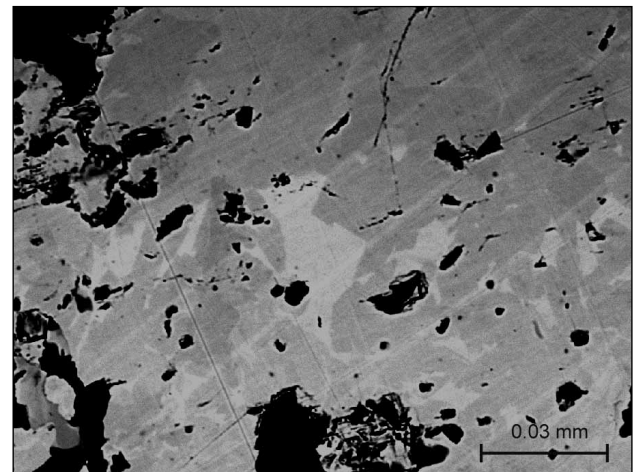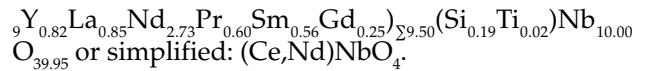

Fig. 6. Patchy aggregate of monazite. (Sample GI 104361, locality B).

Tables 2 and 3, respectively. Analyses of eudialyte from different locallties are listed in Table 4. The first author can supply detailed analytical data for all the minerals on request.

Identified minerals (Table 2)

Ce-fergusonite has a molar ratio Nb: REE close to 1:1. The proportions of the individual REE vary strongly and no obviously systematic substitutional relationships appear to exist. Based on the average value of all the analyzed Ce-fergusonites, we have calculated the following empirical formula for the mineral, ignoring the small amounts of Th, Al, Cl and F: $Ca_{0.26}(Ce_{3.6}$
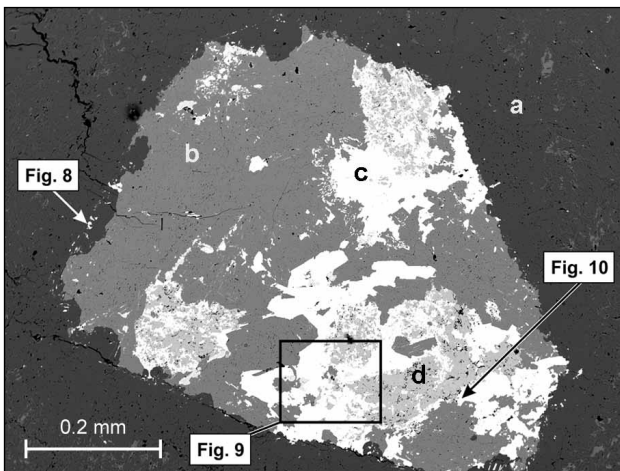

Fig. 7. Blocky eudialyte decomposition aggregate (strictly a pseudomorph after an original eudialyte crystal) in analcime (a, dark grey) is composed of K-feldspar (b, grey) and patchy areas of zircon, Ce-fergusonite and allanite (c, all three bright white) enclosing aegirine (d, light grey). For details see Figs. 8-10. (Sample GI 109302, locality C).

$_9Y_{0.82}La_{0.85}Nd_{2.73}Pr_{0.60}Sm_{0.56}Gd_{0.25})_{\Sigma 9.50}(Si_{0.19}Ti_{0.02})Nb_{10.00}$ $O_{39.95}$ or simplified: $(Ce,Nd)NbO_4$.

Y-fergusonite does not vary as much in composition as Ce-fergusonite. The low totals are ascribed to the presence of HREE which, apart from Gd, have not been analysed. The REE values vary from sample to sample, but not as much as in Ce-fergusonite. The following empirical formula for the mineral, based on the average values of all analyses recorded is: $(Ca,Fe)_{0.56}(Ce_{0.45}Y_{5.06}La_{0.07}Nd_{0.71}Pr_{0.10}Sm_{0.40}Gd_{0.63})_{\Sigma 7.42}Si_{0.2}$ $_7Ti_{0.10}Nb_{10.00}F_{0.18}Cl_{0.01}O_{37.25}$, or simplified: (Ca, Fe) $_{0.5}REE_{7.5}(Si, Ti)_{0.5}Nb_{10}O_{37.75}$ (plus HREE).

Allanite was found at localities B and C. A representative analysis of the mineral is listed in Table 2.

Fig. 8. Partly altered Ce-fergusonite. Primary Ce-fergusonite is white (a), the replacing variety is grey (b). (Sample GI 109302, locallity C).
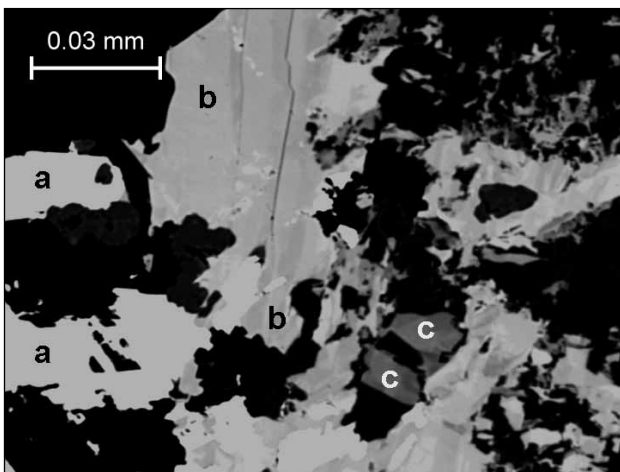
Fig. 9. Zircon (a, off-white), allanite (b, pale grey), and Uk1 (c, two dark grey grains in the lower central right part of the photo). The location of the figure is shown in Fig. 7. (Sample GI 109302, locality C).

Fig. 10. A relatively large crystal of zircon (a, white, bottom right) and three crystals of allanite (b, mottled grey) enclosing grains of Nb1 (c, white), all embedded in K-feldspar (black). (Sample GI 109302, locality C).

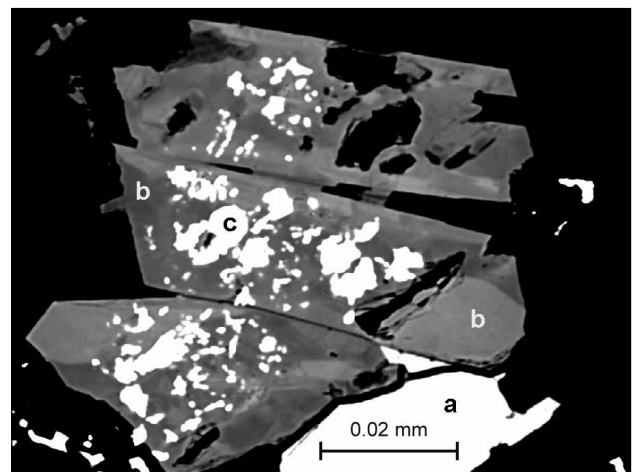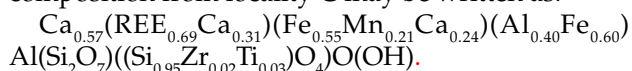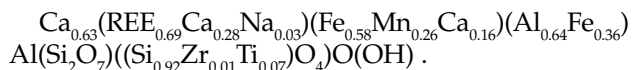Using the traditional allanite formula, its average composition from locality C may be written as:
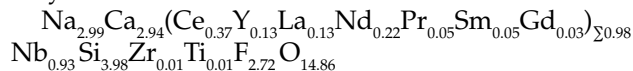
$Ca_{0.57}(REE_{0.69}Ca_{0.31})(Fe_{0.55}Mn_{0.21}Ca_{0.24})(Al_{0.40}Fe_{0.60})$ $Al(Si_2O_7)((Si_{0.95}Zr_{0.02}Ti_{0.03})O_4)O(OH)$.

For allanite from locality B, the average composition would be:

$Ca_{0.63}(REE_{0.69}Ca_{0.28}Na_{0.03})(Fe_{0.58}Mn_{0.26}Ca_{0.16})(Al_{0.64}Fe_{0.36})$ $Al(Si_2O_7)((Si_{0.92}Zr_{0.01}Ti_{0.07})O_4)O(OH)$.

The two formulae have the same total REE content. However, the proportions between

the individual REE vary significantly.

Nacareniobsite-(Ce) was only found at locality D and only in one eudialyte decomposition aggregate. Microprobe analyses of 11 grains show little variation from grain to grain. The following empirical formula may be written for the mineral.

$Na_{2.99}Ca_{2.94}(Ce_{0.37}Y_{0.13}La_{0.13}Nd_{0.22}Pr_{0.05}Sm_{0.05}Gd_{0.03})_{\Sigma 0.98}$ $Nb_{0.93}Si_{3.98}Zr_{0.01}Ti_{0.01}F_{2.72}O_{14.86}$

or simplified: $Na_3Ca_3REENb(Si_2O_7)_2OF_3$. Nacareniobsite-(Ce) was originally found at Kvanefjeld near locality A in Fig. 1 (Petersen et al. 1989).

Fersmite has the simplified composition $CaNb_2O_4$ and contains small amounts of TiO_2 (1.5 wt%), REE (2.7 wt%), FeO (0.9 wt%) and 0.2 wt % MnO and SiO_2.
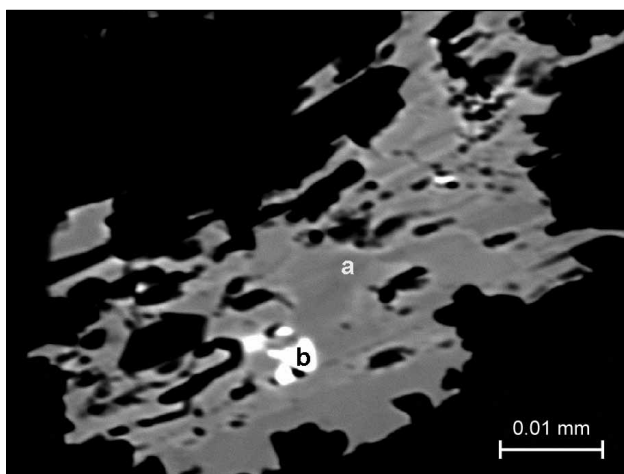

Fig. 11. Patchy grain of Nb2 (a) enclosing a grain of Y-fergusonite (b) lies in contact with allanite, zircon and K-feldspar (all black). (Sample 109302, locality C).
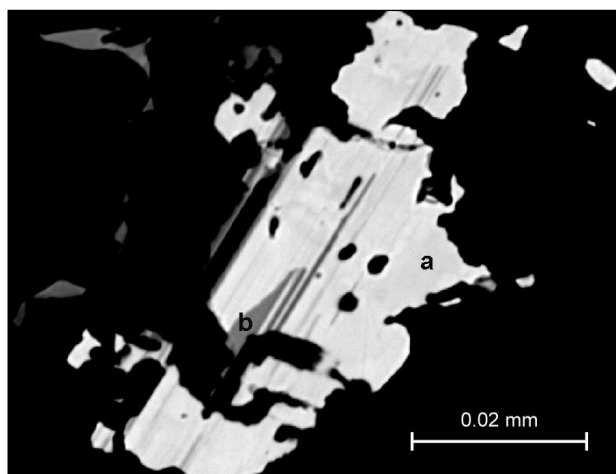

Fig. 12. Grain of mineral Uk2 (a, white), with grey lamellar phase (b). Secondary black unidentified phase replaces both Uk2 and the lamellar phase. (Sample GI 109304, locality C).

Table 2. Minerals observed in altered eudialyte from the marginal pegmatite the Ilímaussaq complex.

Mineral	Formula, based on chemical analyses this paper	Localities			
		A	D	B	C
Zircon	$Zr_{3.5}REE_{0.5}O_{10}Si_{4.0}O_{15..37}$			X	X
Catapleiite	$(Na,Ca,Fe)ZrSi_3O_8 \cdot nH_2O$	X	X		X
Ce-fergusonite	$(Fe,Nd)NbO_4$			X	X
Y-fergusonite	$(Ca,Fe)_{0.5}REE_{7.5}(Si,Ti)_{0.5}Nb_{10}O_{37.75}$	X			X
Fersmite	$CaNb_2O_6$				X
Allanite	$Ca(Ce,La,Ca)(Fe^{+2},Fe^{+3})(Al,Fe^{+3})Al(Si_2O_7)(SiO_4)O(OH)$			X	X
Nacareniobsite-Ce	$Na_3Ca_3REENb(Si_2O_7)_2OF_3$		X		
Monazite	$CeREEP_2O_8$	X		X	
Apatite	$Na_{0.5}Ca_{4.5}REE_{0.5}(PO_4)_3F(OH)$	X			
Thorianite with U	$(Th,U)O_2$ *	X	X		
A1	$(Na,Ca,Fe,Mn)_2Ce_3(REE)_3Si_6FO_{22.5}$	X	X	X	X
A2	$REE_3Si_4O_{12.5-y}(OH)_{2y} \cdot nH_2O$			X	X
Nb1	$(Fe,Mn,Ca)_{6.5}Nb_{10}REE_{10}Al_{3.5}Si_{7.5}O_{66.75}$				X
Nb2	$Mn_2Nb_2O_7$				X
Uk1	$(Fe,Mn,Ca)_{3.0}REE_{0.5}Al_{1.75}Si_4O_{14.38} \times nH_2O$				X
Uk2	$CeREEF_{1.5}O_{1.75-y}(OH)_{2y} \times nH_2O$				X

* Not analyzed for U.

Zircon is one of the major alteration products of eudialyte at localities B and C. A representative microprobe analysis is listed in Table 2. Zircon contains between 3.7 and 4.8 wt.% REE. The total content of FeO, MnO and CaO is less than 0.5 wt.%.

Catapleiite is the Zr-bearing member of the alteration products of eudialyte at localities A and D, and is a minor constituent at locality C. A representative chemical analysis is presented in Table 2. Na easily burns away under the electron beam and the measurements for this element were therefore made with a sweeping beam over an area 30 square microns in size. The lowest content of CaO (0.24 wt.%) and highest content of Na_2O (10.51 wt.%) were recorded in catapleiite from locality A and the highest content of CaO (3.11 wt.%) and correspondingly lowest content of Na_2O (5.14 wt.%) were recorded at locality C.

Monazite was identified in samples from localities A and B and the average composition of the analyses recorded is listed in Table 2. The Ce content in the analyzed grains is fairly constant. However, the REE contents vary strikingly. With increasing content of La the content of the remaining REE decreases. Two extreme compositions recorded in dark and bright areas of the monazite aggregate shown in Fig. 6, were respectively:

(a): $Ce_{0.93}La_{0.44}(Nd_{0.36}Pr_{0.09}Sm_{0.05}Gd_{0.02})_{0.52}P_{2.00}F_{0.01}O_{7.84}$ and
(b): $Ce_{0.93}La_{0.88}(Nd_{0.07}Pr_{0.04}Sm_{0.01})_{0.12}P_{2.00}F_{0.01}O_{7.88}$.

Apatite. Three microprobe analyses gave the formula: $Na_{0.42}Ca_{4.46}Si_{0.15}REE_{0.56}(PO_4)_3F_{0.99}OH_{1.62}$, or simplified: $Na_{0.5}Ca_{4.5}REE_{0.5}(PO_4)_3F(OH)$. This is very similar to the composition of apatite in naujaite (Rønsbo 2008).

Unidentified minerals (Table 3)

Mineral A1 is quite common. Most analyses have totals near 100 wt.%. The SiO_2 content is close to 23 wt.% and fairly constant. The following simplified formula is proposed: $(Na,Ca,Fe,Mn)_2Ce_3REE_3Si_6FO_{22.5}$. A detailed study of minerals A1 and A2 in the kakortokites is currently being carried out.

Mineral A2 was found at localities B and C. The average composition of A2 gives a total of only 82.84 wt.%, which suggests that it is a hydrated REE-silicate. The following simplified formula for the mineral is proposed: $REE_3Si_4O_{12.5-y}(OH)_{2y}·nH_2O$.

Mineral Nb1 has only been found at locality C. Two groups of analyses were recorded for the mineral

Table 3. Representative electron microprobe mineral analyses from the Ilímaussaq complex.

Oxide	Ce-Fergu-sonite	Y- Fergu-sonite	Allanite	Fersmite	Nacar-eniob-site-(Ce)	Zircon	Catapleiite	Grossula-rite	Monazite	Apatite
SiO_2	0.19	0.60	32.52	0.24	29.32	32.21	44.45	35.88	0.39	1.52
ZrO_2	n.d.	n.d.	n.d	n.d.	n.d.	58.30	33.66	n.d.	0.07	0.11
ThO_2	0.24	0.11	0.03	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
TiO_2	0.08	0.28	1.55	1.53	0.08	0.08	1.04	0.30	n.d.	n.d.
Nb_2O_5	46.75	49.82	n.d.	76.49	14.95	n.d.	n.d.	0.25	n.d.	n.d.
La_2O_3	2.28	0.38	7.63	0.04	1.80	0.10	n.d.	0.03	24.04	5.87
Ce_2O_3	16.45	2.95	10.81	0.14	7.35	0.89	n.d.	0.03	34.30	7.47
Nd_2O_3	13.07	3.52	1.63	0.82	4.45	1.05	n.d.	0.05	7.93	1.61
Pr_2O_3	3.02	0.73	0.63	0.17	0.96	0.07	n.d.	0.07	2.37	0.42
Sm_2O_3	2.40	1.40	0.12	0.97	1.11	0.41	n.d.	n.d.	1.06	0.12
Gd_2O_3	1.42	3.06	0.07	0.43	0.76	0.38	0.26	n.d.	0.28	0.19
Y_2O_3	11.21	24.36	n.d.	0.29	1.73	2.70	0.39	0.16	0.04	0.06
Al_2O_3	n.d.	n.d.	15.09	n.d.	n.d.	0.40	n.d.	10.40	n.d.	n.d.
FeO	n.d.	0.07	12.41	0.88	n.d.	0.05	0.26	15.21	n.d.	n.d.
MnO	n.d.	n.d.	3.77	0.21	n.d.	0.08	n.d.	4.29	n.d.	n.d.
CaO	n.d.	0.53	11.05	16.87	20.06	0.06	0.35	31.97	0.08	42.89
Na_2O	n.d.	n.d.	0.12	n.d.	11.28	0.32	6.23	n.d.	n.d.	2.26
K_2O	n.d.	n.d.	n.d.	n.d.	n.d.	0.04	0.06	n.d.	n.d.	n.d.
P_2O_5	n.d	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	31.44	36.54
Cl	0.24	0.06	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
F	n.d	0.13	n.d	n.d.	6.29	0.11	0.14	0.72	0.08	3.24
Totals	97.35	88.00	97.43	99.08	100.14	97.25	86.84	99.36	102.08	102.30

n.d. not detected

(Table 3). Characteristics for both groups are the nearly identical contents of Si, Nb, Al, Fe, Mn, and Ca. However, the proportions between the individual REE elements vary strikingly between each group, although the sum of the REE per formula unit is close to 10 (10.53 and 9.88) for both groups. We suggest the following simplified formula for the mineral: $(Fe,Mn,Ca)_{6.5}Nb_{10}REE_{10}Al_{3.5}Si_{7.5}O_{66.75}$.

Mineral Nb2 has the simplified composition $Mn_2Nb_2O_7$. It contains 2.4 wt % TiO_2 and insignificant FeO, CaO and REE.

Mineral Uk1. Two small grains are associated with allanite in the sample from locality C (Fig. 9). The sum of the elements is only 94.6 wt.%, which implies that the mineral is hydrous. The following simplified formula is proposed: $(Fe,Mn,Ca)_{3.0}REE_{0.5}Al_{1.75}Si_4O_{14.38} \times n\,H_2O$.

Mineral Uk2. On the basis of the average of nine analyses of this presumably new mineral, the following formula is proposed:
$Ce_{1.00}(La_{0.63}Nd_{0.22}Pr_{0.07}Sm_{0.02}Gd_{0.01})_{\Sigma 0.95}Ca_{0.03}F_{1.58}O_{1.75-y}(OH)_{2y} \cdot n\,H_2O$ or simplified: $CeREEF_{1.5}O_{1.75-y}(OH)_{2y} \times n\,H_2O$.

Discussion

The present study of the alteration of eudialyte from the Ilímaussaq complex confirms the observations of Ussing (1898) and Bøggild (1953) of two distinct types of alteration, one leading to zircon- and the other to catapleiite-bearing assemblages. In both cases, eudialyte is pseudomorphed by aggregates of minerals which, at the time of Ussing and Bøggild, were too fine-grained for detailed identification. Electron microprobe study has revealed a number of minerals, some of which are reported for the first time from the Ilímaussaq complex, and some may be new minerals (Table 3).

The zircon alteration type was found in thoroughly altered agpaitic rocks; nepheline and potassium feldspar being substituted by analcime and sericite, amphibole and primary aegirine by late aegirine. This type of alteration was identified in rocks of the marginal pegmatite close to the external contacts of the complex. At locality B they are penetrated by a network of lujavrite dykes and at locality C by a swarm of parallel pegmatite dykes. The eudialyte alteration aggregates consist of zircon, alkali feldspar, aegirine, analcime, grossularite and fluorite and the secondary REE- and Nb-minerals Ce- and Y-fergusonite, allanite, fersmite and some unidentified phases. The fluids responsible for this alteration may have been introduced along fractures and may have been derived from later lujavritic intrusions, or may be of external origin.

The catapleiite alteration type was found throughout the complex in rocks that are less thoroughly altered or almost unaltered. This type of alteration varies from single catapleiite plates in eudialyte in practically unaltered rocks, to pseudomorphs after eudialyte dominated by catapleiite in the most altered rocks. In naujaite, large eudialyte grains may be completely replaced by catapleiite whereas small crystals in the same rocks are unaltered. In lujavrites, eudialyte is generally altered to aggregates of catapleiite, aegirine, mica, analcime, neptunite, carbonates and other minerals (Rose-Hansen & Sørensen, 2002). In the most altered lujavrites, dusty aggregates have pseudomorphed eudialyte (Ussing 1898). In the marginal pegmatite, catapleiite is accompanied by minor monazite, apatite, nacareniobsite-(Ce) and feldspar, aegirine, neptunite, analcime and fluorite. The catapleiite alteration type appears to have been caused by low-temperature late magmatic interstitial fluids.

The late- and post-magmatic alteration is distinctly different from the hyperagpaitic orthomagmatic formation of lovozerite and steenstrupine of the Ilímaussaq complex (Sørensen 1962; Sørensen & Larsen 2001), and the formation of zirsinalite, lovozerite and terskite from the Lovozero agpaitic complex of the Kola Peninsula (Khomyakov 1995; Pekov 2000). These minerals are orthomagmatic, having formed from portions of the agpaitic magma which were enriched in Na, water and other volatiles. The formation of catapleiite appears to have been caused by low-temperature late magmatic interstitial fluids.

The zircon type of alteration has been reported by Bøggild (1953) from the mineral locality Narssarssuk in the Igdlerfigsalik centre of the Igaliko Complex located about 50 km east of the Ilímaussaq complex, and by Coulson (1997) from the North Qoroq centre from the same complex, where eudialyte is replaced by zircon, allanite, aegirine, natrolite, titanite, rinkite, mosandrite and wöhlerite, besides zirfesite. This type of alteration has been described from the Pilansberg complex, South Africa, by Mitchell & Liferovich (2006): zircon, Ce-fergusonite, Ce- and La-allanite, Ce-britholite, titanite, pyrochlore, natrolite, feldspars and a number of Ba and Mn minerals and REE carbonates. The alteration is judged to have been caused by low-temperature Na- and Cl-rich hydrothermal fluids. At Tamazeght, Morocco, eudialyte is replaced by aggregates of zircon, feldspar, aegirine, fluorite and calcite (Khadem Allah et al. 1998). In the Lovozero complex, zircon, vlasovite and woehlerite replace eudialyte in some albitized contact rocks (Pekov 2000).

The catapleiite type of alteration has been described from all eudialyte-carrying complexes. At Tamazeght

pseudomorphs after eudialyte consist of aggregates of catapleiite, feldspar, aegirine, natrolite, vlasovite, rinkite, calcite, rhodochrosite, fluorite and pyrophan- ite. The eudialyte in pegmatites intruding the sedi- mentary country rocks can be altered into zircon, and there are also cases of eudialyte overgrowing zircon (Khadem Allah et al. 1998). Some secondary minerals after eudialyte contain Zr, Nb and REE, whereas oth- ers display a sharp separation of Zr from REE and Nb.

In Fig.13 spider diagrams have been normalized against eudialyte in order to show the movement of elements during the eudialyte alteration process. Fig.13a shows the minerals belonging to the cataplei- ite alteration type and Fig.13b those belonging to the zircon alteration type. Sharp separation of Zr has taken place and most of the secondary minerals are enriched in Nb-REE-Y and F, and depleted in Si, Th, Ti and Zr.

Zirconium is almost exclusively found in zircon and catapleiite, and Th in uranothorianite, whereas
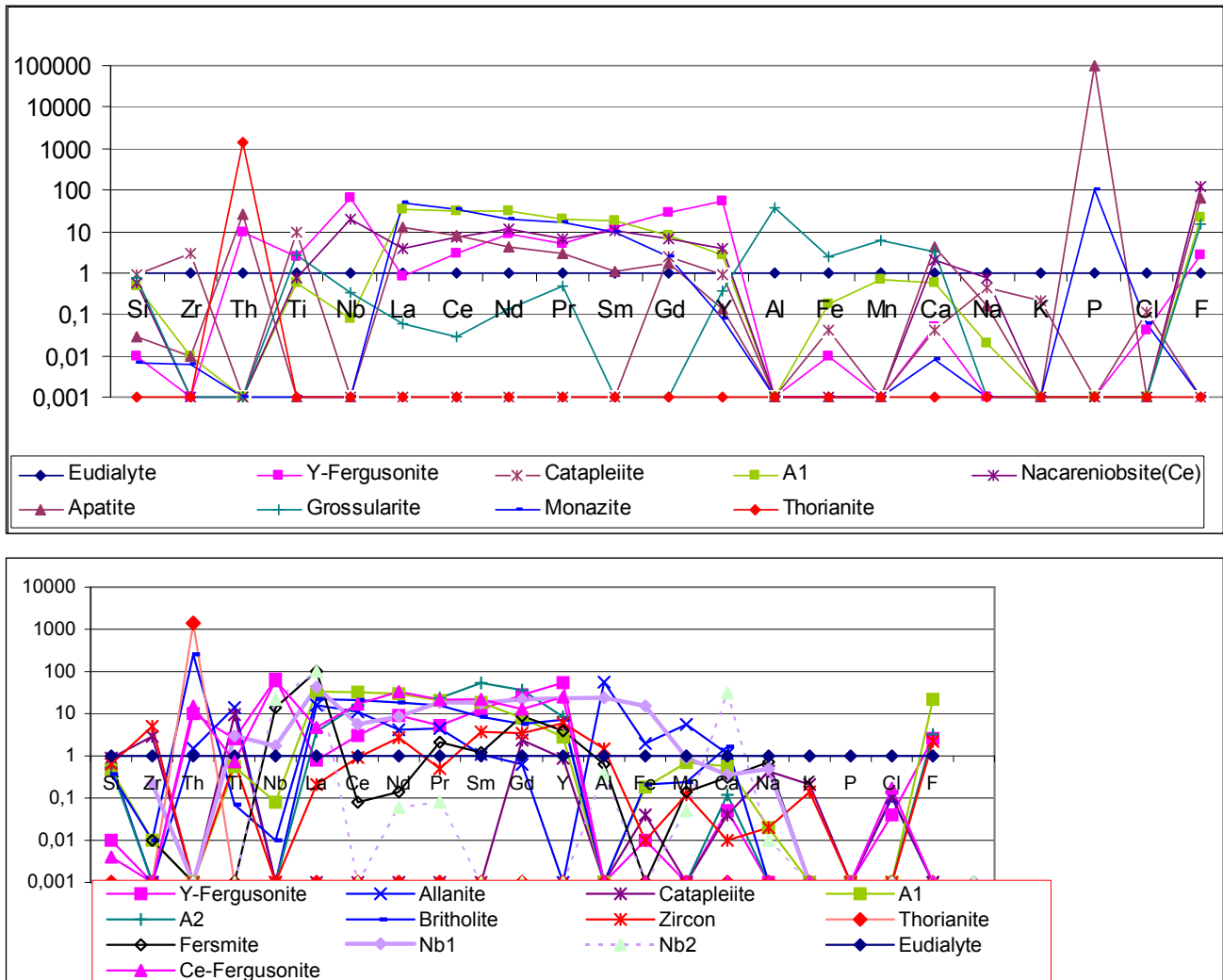

Fig. 13. Eudialyte normalized spider diagrams. Fig.13a is the catapleiite alteration type and Fig.13b the zircon alteration type. Values at 0.001 represent analyses below detection limit and lines joining these 'points' can be disregarded.

Table 5. Zr/Nb, La/Nd, Ce/y and La/Gd rations in minerals from the Ilímaussaq complex

	Eudialyte	Catap- leiite	Zircon	Ce-Fergu- sonite	Y-Fergu- sonite	Fersmite	A1	A2	Nb1	Nb2	Uk1	Uk2
Zr/Nb	10.91 - 17.84	10.71		0	0	0	0.33 - 7.00		0.001 - 0.003		1.23	2.75
La/Nd	0.98 - 1.26		0.05 -0.19	0.17 - 1.19	0.03 - 0.27	0.05 - 0.21	0.79 - 2.62	0.07 - 0.28	0.37 - 0.50		3591	463.4
Ce/Y	1.95 - 2.74			1.40 - 30.92		0.48 - 0.25	6.03 - 313.8	2.36 - 18.36	0.77 - 6.30	0.35		129.1
La/Gd	3.43 - 7.83		0.26 − 5.0	1.61	0.12	0.09	41.07	0.39	3.70			

Eudialyte decomposition minerals with new hitherto undescribed phases from the Ilímaussaq com- · 85

the REE are located in allanite, Ce- and Y-ferarusonite, monazite and most of the unidentified minerals; Nb is held by fersmite, Ce- and Y-ferrusonite, nacareniobsite-(Ce) and the unidentified minerals Nb1 and Nb2. Thus the Zr/Nb ratio, which varies from 10 to 17 in eudialyte, is very high in catapleiite and zircon and low or zero in all the other minerals containing these elements (Table 5). Na, Ca, Al, Fe and Mn are located in the feldspars, grossularite, aegirine, analcime, natrolite, grossularite, as well as in catapleiite, fersmite, allanite, apatite, nacareniobsite-(Ce), A1 and A2.

Ussing (1898, p.174) demonstrated that the bulk Zr content remained in the

pseudomorphs. This may indicate that the bulk chemical composition of eudialyte is

preserved in the pseudomorphs, although there has evidently been loss of Cl and Na and addition of F and P. Buchwald & Sørensen (1961), in a fission track examination of Ilímaussaq minerals, found that minute pigment particles in altered eudialyte are quite

radioactive, which means that U, and probably also Th, released during alteration

of the eudialyte was bound to the pigmentary material.

The alteration of eudialyte resulted in pronounced REE fractionation (Fig. 13).

Our microprobe set-up provided Gd as the only HREE. The LREE/HREE ratio and possible Eu anomaly therefore cannot be calculated; instead the La/Gd and Ce/Y ratios are presented in Table 5, together with the La/Nd ratio. The La/Nd ratio is close to 1.0 in

eudialyte and Ce-fergusonite and is very low in zircon, Y-fergusonite, fersmite, A2 and

Nb1. It is high in apatite and monazite and variable in allanite.

The Ce/Y and La/Gd ratios show lower values than eudialyte in zircon, Y-fergusonite

and fersmite, higher values in A1, apatite, monazite and Ce-allanite, and variable values

in Ce-fergusonite, A2 and Nb1.

In the eudialyte-normalized diagrams (Fig.13) it is evident that all the analyzed minerals, with the exception of Nb2, are enriched in REE relative to eudialyte. Nacareniobsite-Ce, Nb1, Nb2, Uk1, zircon, Ce-fergusonite, A1 and A2 show the same REE pattern as eudialyte. Fersmite is depleted in La, Ce, Pr and the HREE and enriched in the intermediate REE relative to eudialyte. Apatite, monazite, allanite, Uk1 and Uk2 are enriched in LREE and depleted in HREE relative to eudialyte. Y-fergusonite is the only one of the examined minerals which shows marked enrichment in the HREE.

Europium was not determined, but eudialyte displays a pronounced negative Eu anomaly similar to that of the agpaitic rocks of the complex (Bailey et al.

1978; Sørensen 1992; Pfaff et al. 2008).

In Pilansberg, three stages of eudialyte crystallization and five stages of eudialyte

alteration are distinguished (Mitchell & Liferovich 2006). Thus, zircon belongs to an

early miaskitic stage. With increasing alkalinity, eudialyte is formed, followed by

catapleiite, allanite, apatite at decreasing alkalinity.

In the rocks studied by us, only the catapleiite and zircon trends of alteration are distinguished, and we have seen no signs of successive stages of alteration. Rare catapleiite grains have only been observed in one zircon aggregate.

Identity of the unidentified minerals

The unidentified minerals described here have been compared with those from the Lovozero complex (Khomyakov 1995), Pilansberg (Mitchell & Liferovich 2006) and the Tamazeght complex (Kadar 1984; Khadem Allah, 1993). We have not succeeded in correlating the unidentified Ilímaussaq minerals with any of these minerals or with any minerals listed in *Min-Ident-Win*, Dec. 2001.

The chemical compositions of minerals A1 and A2 resemble the composition of Ce-britholite $(Ce,Ca)_{10}(SiO_4,PO_4)_6(OH,F)_2$, cerite $(Ce,Ca)_{10}(SiO_4)_6(OH,F)_5$ and the unnamed mineral mineral X (Kalsbeek et al. 1990) for which we have calculated the formula $NaCa_{1.5}REE_7(Si,P)_6O_{24}OH \times nH_2O$. Mineral X is regarded by Kalsbeek et al.. (1990) as a P_2O_5-poor member of the britholite group of minerals. Britholite is of widespread occurrence in the Ilímaussaq complex.

As we only have information on the chemical compositions and no structural data of minerals A1 and A2, we are unable to decide whether these two minerals are new species

or members of the britholite or cerite groups of minerals. In britholite, and possibly also in cerite, there is complete solid solution between Si and P.

The minerals Nb1 and Nb2 appear to be related to fersmite. The chemical analyses of nacareniobsite-(Ce) and apatite are close to the analyses of these minerals published by Petersen et al. (1989) and Rønsbo (1989, 2008).

Conclusions

Eudialyte is a major constituent of the agpaitic rocks of Ilímaussaq. It may be

perfectly fresh, but is generally altered. At least four types of alteration may be distinguished: the common catapleiite type, the rare zircon type described here, the "zirfesite" type mentioned here and the lujavrite type described by Ussing (1898, 1912).

A characteristic feature is that the crystal shape is preserved, even when the eudialyte has been completely altered, as is the case in the zircon, zirfesite and lujavrite types.

In the two first-named types, eudialyte has been pseudomorphed by alteration products, but in the lujavrite type only "shells" of eudialyte crystals are preserved. In the catapleiite type, alteration may be weak as described here, or complete as in some kakortokites where crystals are pseudomorphed by catapleiite aggregates Alteration results in a pronounced separation of elements. Zr is found only in zircon and catapleiite. REE are located in REE minerals such as monazite, apatite, allanite, A1, A2, and UK1 and UK2. Nb is bound in fersmite and Nb2. REE and Nb occur together in fergusonite, Nb1, and nacareniobsite-(Ce). Allanite and nacareniobsite-(Ce) act as collectors for REE and Ca. Allanite is low in Na and marks a miaskitic trend, nacareniobsite-(Ce) is high in Na and marks the agpaitic trend. Besides allanite and nacareniobsite, Ca is found in monazite, grossularite, fluorite, catapleiite and fersmite.

Ussing (1898) demonstrated that the Zr content of the eudialyte was most probably retained in the pseudomorphs. The same may be the case with REE, Nb and perhaps Ca. It was, however, not a completely closed system as Cl has certainly been lost and F and P introduced. A quantitative test is impossible where all the original eudialyte has disappeared, as is the case in the zircon type of alteration. The catapleiite type of alteration took place at a late magmatic stage by residual interstitial liquids, whereas the zircon type may have formed by liquids of external origin.

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