The Ilímaussaq alkaline complex, South Greenland: status of mineralogical research with new results

Edited by Henning Sørensen

Contributions to the mineralogy of Ilímaussaq, no. 100 Anniversary volume with list of minerals

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Cover

Igneous layering in kakortokites in the southern part of the Ilímaussaq alkaline complex, South Greenland. The central part of the photograph shows the uppermost part of the layered kakortokite series and the overlying transitional kakortokites and aegirine lujavrite on Laksefjeld (680 m), the dark mountain in the left middle ground of the photograph. The cliff facing the lake in the right middle ground shows the kakortokite layers + 4 to + 9. The kakortokite in the cliff on the opposite side of the lake is rich in xenoliths of roof rocks of augite syenite and naujaite making the layering less distinct. On the skyline is the mountain ridge Killavaat ('the comb'), the highest peak 1216 m, which is made up of Proterozoic granite which was baked and hardened at the contact to the intrusive complex. The lake (987 m) in the foreground is intensely blue and clear because it is practically devoid of life. The whole area is devoid of vegetation, with crumbly rocks typical of the nepheline syenites of the complex.

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Preface

The Mesoproterozoic Ilímaussaq alkaline complex located near the town of Narsaq in South Greenland is the type locality for agpaitic nepheline syenites and represents an enormous concentration of a number of rare elements, particularly Li, Be, Nb, Zr, REE, Y, U and Th. This explains the presence of about 220 minerals, 27 of these discovered in and first described from the complex, and nine only found there.

In 1965 it was decided to collate papers on the mineralogy (in a broad sense) of the complex published in various scientific journals into a consecutively numbered series: Contributions to the mineralogy of Ilímaussaq. The first issue appeared in 1965, no. 102 in 1999, and the series has now reached no. 113. Issue no. 100 was reserved for the anniversary volume presented here. This volume contains a brief account of the geology of the complex and the history of exploration, a list of the minerals identified so far in the complex, and papers which review selected topics and present new information on the mineralogy and geochemistry of Ilímaussaq. A companion report, Danmarks og Grønlands Geologiske Undersøgelse Rapport 2001/102, compiled by J. Rose-Hansen, H. Sørensen and W.S. Watt, presents an inventory of the literature of the complex, collated from an unusually wide variety of sources and containing about 750 entries, together with a list of the series Contributions to the mineralogy of Ilímaussaq. The report brings these both in print and on a CD-ROM, accessible for electronic searching.

We hope that this collection of papers on Ilímaussaq will be received in the same positive way as was its predecessor, 'The Ilímaussaq intrusion, South Greenland. A progress report on geology, mineralogy, geochemistry and economic geology', published as Contribution to the mineralogy of Ilímaussaq no. 63 in *Rapport Grønlands Geologiske Undersøgelse* **103** edited by J.C. Bailey, L.M. Larsen & H. Sørensen in 1981.

Contribution no. 63 was compiled after more than ten years of intensive field activity and presented new information on the geology and geochemistry of the Ilímaussaq complex together with papers on mineralogy, fluid inclusions and economic geology. Since then there has only been limited field activity in the complex. Contribution no. 100 (this volume) therefore presents results of the continued laboratory investigations of material collected during the earlier field work and drilling programmes together with discussions of a few selected topics in the light of new developments in the study of agpaitic rocks and their minerals.

The paper by J.C. Bailey, R. Gwozdz, J. Rose-Hansen and H. Sørensen contains the first general discussion of the geochemistry of the complex since V.I. Gerasimovsky's 1969 memoir on this subject. This overview of the geochemistry is based on the on-going detailed analysis of 120 large samples of the major rock types collected by blasting in order to obtain fresh material. Mineral separates have been prepared of many of the samples with the aim of analysing the rock-forming and accessory minerals, together with the bulk rocks, for more than 50 elements. The ultimate purpose is to evaluate the composition of the agpaitic melts and the distribution of a number of elements between minerals and the melt and fluid phases in order to better understand the conditions of formation of agpaitic mineral associations. The following paper by J.C. Bailey illustrates this by describing the distribution of germanium in the rocks and minerals of the complex.

Eudialyte may be called the typomorphic mineral of agpaitic rocks *sensu stricto* and is one of the minerals first found and described from Ilímaussaq. The nomenclature of the mineral has, however, been rather confusing. Based on the optical properties three varieties, eudialyte, mesodialyte and eucolite, have been distinguished, but it has been impossible to relate these differences to chemical properties. In recent years it has been demonstrated that eudialyte comprises a group of minerals. It was therefore felt timely to include a review of this mineral group in the anniversary volume (the paper by O. Johnsen, J.D. Grice and R.A. Gault).

The most highly evolved agpaitic rocks and mineral associations have been called hyper-agpaitic by Russian colleagues working in the Khibina and Lovozero complexes of the Kola Peninsula. Hyper-agpaitic rocks are extremely rich in rare minerals, a result of crystallisation of peralkaline, volatile-rich, low-viscosity melts over a broad temperature interval, ending at low temperatures. This results in long-range ordering of the crystal structures and the predominance of triclinic minerals (paper by A.P. Khomyakov). In Ilímaussaq, hyper-agpaitic mineral associations are found not only in pegmatites and hydrothermal mineralisations as in the Kola complexes but also in the most evolved lujavrites; a review is presented in the paper by H. Sørensen and L.M. Larsen.

The mineral naujakasite presents an enigmatic problem. It is composed of the most common elements, Na, Al, Fe, Mn and Si, but has nevertheless until now only been known from the Ilímaussaq complex, where it is sometimes a rock-forming mineral which may constitute more than 75 vol.% of some lujavrites. The recent discovery of a Mn-rich naujakasite, manganonaujakasite, in a pegmatite from the Lovozero complex of the Kola Peninsula is reported in the paper by A.P. Khomyakov, H. Sørensen, O.V. Petersen and J.C. Bailey. This paper also reviews the occurrence of naujakasite in hyper-agpaitic lujavrites of the Ilímaussaq complex and examines why this mineral is common in Ilímaussaq, very rare in Lovozero and absent in the Khibina complex of the Kola Peninsula.

Steenstrupine is a very important rock-forming mineral in the hyper-agpaitic mineral associations of Ilfmaussaq and is the main radioactive mineral in the Kvanefjeld uranium deposit. The mineral is generally metamict. Varieties with marginal anisotropic zones have, however, been known for a long time, but their origin was not well understood. New microprobe analyses of zoned steenstrupine crystals have demonstrated chemical differences which can explain the coexistence of metamict and crystalline steenstrupine (paper by A.P. Khomyakov and H. Sørensen).

Some rare minerals, as for instance steenstrupine, rinkite, epistolite and ussingite, are found in considerable quantities throughout the Ilímaussaq complex. Others like semenovite and chalcothallite have only been found in a few places as isolated grains. Until recently, leucophanite was considered to belong to the last-named group but is now known to occur in a number of localities within the complex (paper by H. Bohse, O.V. Petersen and G. Niedermayr).

In E.I. Semenov's pre-microprobe 1969 memoir on the mineralogy of Ilímaussaq, a number of minerals were described under preliminary names. E.I. Semenov's paper in the present volume reports the status of some of these minerals, including the occurrence of a new mineral, Na-komarovite. In spite of a low average content of 1.9 ppm thallium in the complex, four minerals having Tl as a major component have been found. The paper by S. Karup-Møller and E. Makovicky reports the discovery of a new occurrence of thalcusite and discusses the tarnishing of polished sections of this mineral.

Brown prismatic crystals of rinkite are of widespread occurrence in Ilímaussaq. It was therefore a surprise that rinkite-looking crystals in a naujaite pegmatite were found to be hiortdahlite (the paper by E.R. Robles, F. Fontan, P. Monchoux, H. Sørensen and P. de Parseval). This brings important information about a stage of reduced alkalinity, in conformity with the occurrence of fayalite and hedenbergite in the adjacent naujaite.

More than 80 water-soluble minerals are known from the Khibina and Lovozero complexes. Until recently only three such minerals were known from Ilímaussaq: villiaumite, trona and thermonatrite. Water-soluble minerals have been looked for in drill cores and the material from the Kvanefjeld adit, but unfortunately not immediately after drilling and blasting. Therefore only two additional water-soluble minerals have been found: the discovery of natrophosphate is reported in the paper by O.V. Petersen, A.P. Khomyakov and H. Sørensen.

Nacareniobsite-(Ce) and vitusite are additional examples of minerals considered to be rare at Ilímaussaq. The paper by I.V. Pekov and I.A. Ekimenkova shows that these minerals are of more widespread occurrence, and provides examples of new types of rare-earth mineralisation in the complex.

Agpaitic and hyper-agpaitic nepheline syenites contain a number of rare Na–Be-minerals such as chkalovite and tugtupite, whereas common Be minerals such as beryl, euclase and chrysoberyl are absent. The paper by G. Markl examines the stability relations of the various Be minerals and explains why the agpaitic rocks contain Na–Be minerals and the order of formation of the minerals.

The fluid phase of agpaitic nepheline syenites is rich in hydrocarbons, especially methane. The paper by J. Konnerup-Madsen reviews the evolution of the fluids and brings new isotopic evidence in support of the non-biogenic origin of these hydrocarbons.

Henning Sørensen

Brief introduction to the geology of the Ilímaussaq alkaline complex, South Greenland, and its exploration history

Henning Sørensen

The Ilímaussaq alkaline complex, the type locality of agpaitic nepheline syenites, is made up of three intrusive phases, (1) augite syenite, (2) alkali acid rocks and (3) agpaitic nepheline syenites which occupy the major part of the complex. The agpaitic phase comprises a roof series, a floor series and an intermediate sequence of rocks. The roof series crystallised from the roof downwards beginning with non-agpaitic pulaskite and ending with distinctly agpaitic naujaite. The exposed part of the floor series is made up of the layered agpaitic nepheline syenite kakortokite. The intermediate sequence consists of several types of distinctly agpaitic lujavrites which are accompanied by occurrences of uranium and other rare elements.

The complex was first visited by K.L. Giesecke in 1806 and 1809. The first detailed mapping of the complex was carried out by N.V. Ussing in 1900 and 1908. He presented a precise description of the major rock types and an illuminating discussion of the petrology of the complex in his 1912 memoir. In the period 1912–1955 there was very limited activity in the complex. Exploration for radioactive minerals in Ilímaussaq was initiated in 1955 and in subsequent years followed by geological mapping carried out by the Geological Survey of Greenland. This led to a series of detailed studies of the occurrences of not only U, but also Be, Nb, REE and Zr, and to mineralogical, geochemical and petrological studies as well as commercial evaluation and drilling.

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Keywords: agpaite, alkaline complex, Ilímaussaq, nepheline syenites, South Greenland, uranium deposit

The Ilímaussaq alkaline complex (Fig. 1) is one of a number of intrusive complexes in the Gardar igneous province, a mid-Proterozoic rift zone in South Greenland (Allaart 1973; Upton & Emeleus 1987; Kalsbeek et al. 1990; Macdonald & Upton 1993). The first detailed description, with a geological map and a discussion of the petrogenesis of the complex, was presented by Ussing (1912), who introduced the term agpaitic nepheline syenite. According to the recommendations of the IUGS Subcommission on the Nomenclature and Classification of Igneous Rocks (Le Maitre 1989), the term agpaitic should be restricted to peralkaline nepheline syenites having complex Zr-Ti silicate minerals such as eudialyte and rinkite instead of the more common minerals zircon, titanite and ilmenite. Since the appearance of Ussing's memoir

numerous papers on the geology, mineralogy, petrology, geochemistry and economic geology of the complex have been published as is apparent from the bibliography presented in a companion report (Rose-Hansen *et al.* 2001). The most recent presentations of the general geology and petrology of the complex are those of Larsen & Sørensen (1987) and Sørensen & Larsen (1987).

The complex has been dated at 1143 ± 21 Ma (recalculated from Blaxland *et al.* 1976), 1130 ± 50 Ma (Paslick *et al.* 1993), 1160 ± 5 Ma (U–Pb, G. Markl, Tübingen University, personal communication 2000), 1160.7 ± 3.4 Ma and 1161.8 ± 3.4 Ma (Rb–Sr, T. Waight, Danish Lithosphere Centre, personal communication 2000).

The complex measures 17×8 km, and the exposed

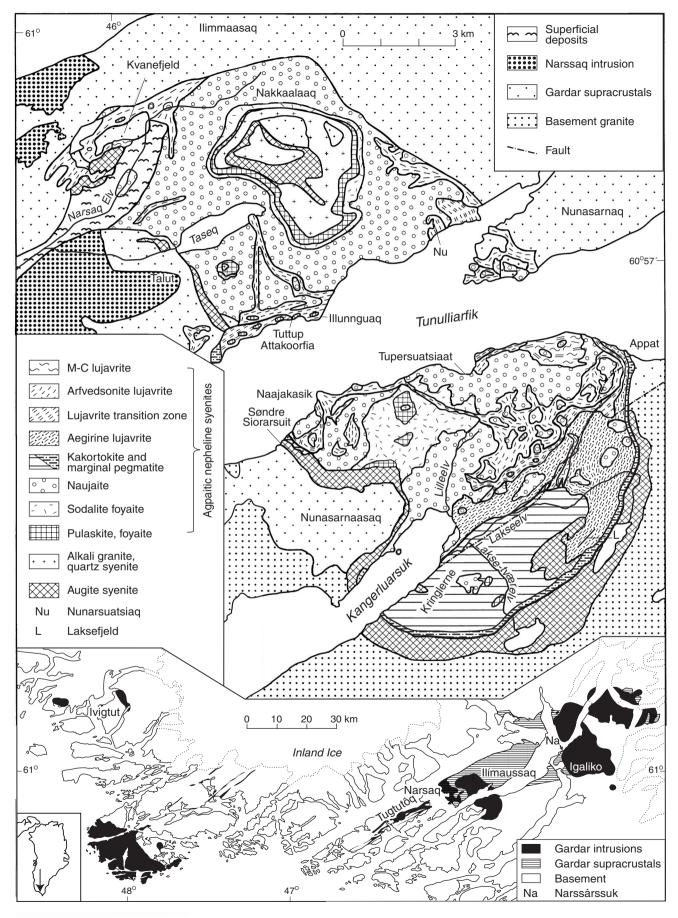


Fig. 2. The contact between the agpaitic rocks (on the right) and the augite syenite (on the left). The augite syenite is penetrated by fractures parallel to the contact, and rafts of augite syenite are enclosed in the agpaitic rock. Lakse-tværelv in the southernmost part of the complex.



vertical thickness is about 1700 m. It is estimated that the complex was emplaced 3–4 km below the contemporary surface at the discontinuity between the Ketilidian crystalline basement (*c*.1800 Ma, e.g. Chadwick & Garde 1996) and the overlying Eriksfjord Formation made up of continental sandstones and lavas of mainly basaltic composition. The basement and the overlying sandstones and lavas are intruded by numerous mainly basaltic dykes. The Eriksfjord Formation is the surface expression of Gardar activity and is preserved only in down-faulted blocks. Gardar activities embrace the period 1350 to *c*. 1120 Ma (Paslick *et al.* 1993); the Ilímaussaq complex is thus an expression of young Gardar activity.

Three intrusive phases may be distinguished in the formation of the Ilímaussaq complex (Fig. 1). The first phase is made up of augite syenite which is preserved only as a partial marginal shell and in the roof (Fig. 2). The second phase consists of alkali granite and quartz syenite which are found in the roof and as blocks engulfed by rocks of the third intrusive phase (Steenfelt 1981). The third intrusive phase occupies the major part of the complex. It is made up of a roof series, a floor series, and an intermediate sequence (Fig. 3).

The major rock types of the complex are presented in Table 1.

The roof series crystallised from the top downwards, forming the succession pulaskite, foyaite, sodalite

foyaite and naujaite (Figs 3, 4). The contacts of these rocks grade into each other, but blocks of the uppermost rocks were loosened from the temporary roof of the magma chamber and engulfed by the underlying crystallising rocks. Larsen (1976) demonstrated that there is a gradual evolution in mineralogy from pulaskite to naujaite. The primary mineral association alkali feldspar, nepheline, fayalite, hedenbergite, apatite and titanomagnetite is substituted downwards by sodalite, nepheline, alkali feldspar, aegirine, arfvedsonite, eudialyte and aenigmatite. Sodalite is an interstitial mineral in the early stages of formation of the sodalite foyaite, but gradually becomes a liquidus phase and is a flotation cumulus phase in the naujaite. This rock is poikilitic and made up of crystals of sodalite, up to 1 cm across, enclosed in grains of alkali feldspar, aegirine, arfvedsonite and eudialyte, which may measure 10 cm or more. The sodalite foyaite and naujaite are agpaitic nepheline syenites.

The floor series is made up of a layered and laminated series of kakortokite, that is an agpaitic nepheline syenite with the major minerals alkali feldspar, nepheline, aegirine, arfvedsonite and eudialyte. The bottom of the series is unknown. The lowermost visible part is made up of centimetre-thick layers with varying contents of mafic minerals, feldspar and eudialyte. It displays trough structures and cross-bedding and is overlain by a series made up of 29 three-layer units, each about 10 m thick and made up of a lower black layer rich in arfvedsonite and an upper much thicker white layer rich in alkali feldspar (Fig. 5). Between these layers, there is often a thin red layer rich in eudialyte (Bohse et al. 1971). The black, red and white layers pass gradually into each other, whereas the black layers are separated from the underlying

Fig. 1. Simplified geological map of the Ilímaussaq alkaline complex. The area to the north of Tunulliarfik is based on Ferguson (1964), the area to the south of this fjord on Andersen *et al.* (1988), with minor modifications. Index map with names of Gardar complexes mentioned in the text. **M-C** lujavrite: medium- to coarse-grained. For further explanation see Table 1.

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

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

* Analcime and natrolite are secondary minerals in most rocks.

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

white layers by sharp contacts. Bohse *et al.* (1971) have numbered the layered units, the lower part from unit no. –11 to unit 0, the upper part from units 0 to +17. The most recent discussions of the origin of the layering are given by Larsen & Sørensen (1987) and Bailey (1995); see also Upton *et al.* (1996).

The lowermost part of the naujaite and the lowermost sodalite-bearing part of the kakortokite may be contemporaneous (Sørensen & Larsen 1987), but the floor sequence which formed simultaneously with the greater part of the roof zone is unexposed.

The kakortokite and in places the naujaite are sepa-

rated from the augite syenite rim by a marginal pegmatite zone (cf. Bohse *et al.* 1971; Bohse & Andersen 1981).

The layered series of kakortokite passes gradually upwards into a thin unit of transitional layered kakortokite which again passes gradually into the intermediate sequence of lujavrites. These are agpaitic mesoto melanocratic nepheline syenites which are generally fine-grained, laminated and occasionally layered (Sørensen & Larsen 1987).

The lowermost part of the lujavrite sequence is made up of green rocks rich in aegirine and in places also in eudialyte. Bohse & Andersen (1981) distinguish a Fig. 3. A section through the complex viewed from Kringlerne plateau towards the north. Foreground layered kakortokites (K), the dark rocks in the middle ground belong to the intermediate sequence of lujavrites (L). The light grey rocks in the wall facing south towards the fjord Kangerluarsuk (Kang) are made up of the roof series with pulaskite-foyaite (P) and sodalite foyaite (SF) in the upper part, naujaite (N) in the lower part bordering the augite syenite shell (A). The contact between the roof zone and the basement granite and its overlying sandstone (S) with intercalated basalt (B) on the mountain Nunasarnaasaq (1442 m, on the left) is very sharp. The high mountains in the far distance (to the north of Tunulliarfik, see Fig. 1) have remnants of the volcanic roof (B) on top of the roof series (cf. Fig. 4). The distance to Kangerluarsuk is about 2 km.



lower again lujavrite I zone which gradually passes into the overlying aggirine lujavrite II. The upper part of the intermediate sequence is made up of black arfvedsonite-rich, fine-grained laminated rocks. Dykes and sheets of lujavrite intersect the rocks of the roof zone; in places the naujaite in the immediately overlying roof is brecciated and strongly altered by the lujavrite. On the Kvanefjeld plateau in the northernmost part of the complex (Fig. 6), lujavrites are in contact with the volcanic roof of the complex, which is strongly fenitised adjacent to the lujavrites (Sørensen et al. 1969, 1974). There are several generations of lujavrite. One of the latest phases consists of naujakasite lujavrite rich in steenstrupine (Sørensen et al. 1974; Sørensen 1997a). It is an important feature that steenstrupine substitutes for eudialyte in the most evolved lujavrites. This represents the hyper-agpaitic stage of development characterised by naujakasite, steenstrupine, ussingite, vitusite and other minerals (Sørensen & Larsen 2001, this volume). The youngest lujavrites are the so-called medium- to coarse-grained lujavrites (M-C lujavrites); they are accompanied by pegmatites and hydrothermal veins containing steenstrupine, pyrochlore, Be and Cu minerals, and by fenitisation of the volcanic roof. The Kvanefjeld uranium deposit is made up of steenstrupine lujavrites and fenitised roof rocks rich in steenstrupine.

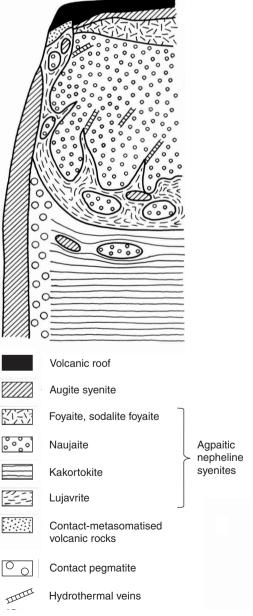


Fig. 4. Schematic section through the c. 1700 m of exposed stratigraphy of the Ilímaussaq alkaline complex.



Fig. 5. The lowermost exposed part of the complex, made up of thin layers of kakortokite with trough layering displaced by a minor fault (arrow, lower left). In the background the main series of kakortokite made up of a repetition of three-layer units, in which the black and white layers can be distinguished. The faulted white trough band is approximately 5 m wide and 15 cm thick.

The arfvedsonite lujavrites and the M-C lujavrites have the highest contents of Li, Rb, Be, REE, Zn, U, Th, etc. of all the rocks of the complex, whereas the kakortokites have the highest contents of Nb, Ta, Zr, Hf and Y (Gerasimovsky 1969; Kunzendorf *et al.* 1982; Sørensen 1992; Bailey *et al.* 2001, this volume).

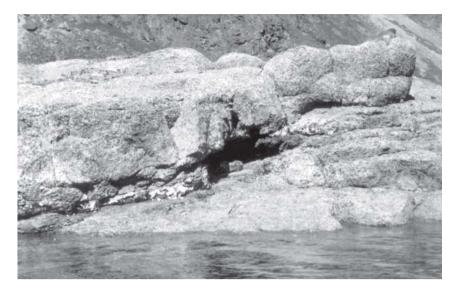
Fluid inclusions in the minerals of the agpaitic rocks of the complex are rich in methane and other hydrocarbons (cf. Konnerup-Madsen 2001, this volume).

The basalts in the part of the Gardar rift zone which contains the Ilímaussaq and Tugtutôq igneous complexes (Fig. 1) are richer in alkalis, P, Ba, Sr, Nb and LREE than the basic rocks in other parts of the Gardar province. This indicates an origin in a mantle source enriched in incompatible elements, perhaps because of metasomatism (Macdonald & Upton 1993, Upton 1996). The agpaitic nepheline syenites of the Ilímaussaq complex are considered to be products of extended fractionation of transitional to alkali basaltic melts in deep magma chambers combined with some crustal contamination (Larsen & Sørensen 1987; Stevenson *et al.* 1997; Bailey *et al.* 2001, this volume). The Tugtutôq– Ilímaussaq zone is underlain by a gravity high which may represent cumulates of olivine and other mafic minerals accumulated during the fractionation processes (Blundell 1978). In this connection it is of interest to note that the Ilímaussaq complex is marked as a magnetic low on the aeromagnetic map of the region (Thorning & Stemp 1997).

The eudialyte-rich parts of the naujaites, kakor-



Fig. 6. The Narsaq Elv valley. The mountain on the left is Kvanefjeld (685 m) made up of volcanic rocks in the upper part and lujavrites, naujaites, etc. in the lower part. The Ilimmaasaq mountain (1390 m, centre) is made up of basaltic rocks underlain by lujavrites. The mountain on the right is Nakkaalaaq 1334 m) made up of the roof series; the slope below, the Taseq slope, is made up of naujaite. The distance between Kvanefjeld and Ilimmaasaq is approximately 4 km. Fig. 7. Layered pegmatite, 0.8 m thick, in naujaite located on the small island in the inner part of Kangerluarsuk. The cave was made by Steenstrup when he collected large samples of eudialyte for the Cryolite Company in 1888. The cave, about 1 m high at the time of photographing in 1964, has since been considerably enlarged by mineral collectors. An attempt is being made to protect the island.



tokites and lujavrites represent an enormous resource of Zr, Nb and REE, especially HREE (Bohse *et al.* 1971; Sørensen 1992). These rock types are accompanied by pegmatites and hydrothermal mineralisations. Examples are ussingite veins rich in chkalovite and other Be minerals (Semenov 1969; Engell *et al.* 1971), occurrences of pyrochlore and other Nb minerals (Hansen 1968), and the above-mentioned Kvanefjeld uranium deposit (Sørensen *et al.* 1974).

History of exploration

Various aspects of the history of exploration of the Ilímaussaq complex have been presented by Sørensen (1967) who considered the history up to 1966, by Nielsen (1981) who described the exploration history of the Kvanefjeld uranium deposit, and by Bondam (1995) who compiled an overview of exploratory activities and environmental studies based on the archives of the Geological Survey of Greenland. In the following a survey of the history of exploration will be presented with emphasis on the period after 1964.

The period 1806–1912

The first scientific study of the complex was carried out by K.L. Giesecke (1761–1833) who made extensive collections of minerals in 1806 and 1809 resulting in the discovery of the minerals arfvedsonite, eudialyte and sodalite.

K.J.V. Steenstrup (1842-1913) visited the complex

several times on behalf of the Commission for the Direction of Geological and Geographical Investigations in Greenland and Kryolith-Mine- og Handels-Selskabet (hereafter referred to as the Cryolite Company) and collected numerous mineral and rock samples (Fig. 7). The minerals were examined by J. Lorenzen (1855–1884) who described the new minerals polylithionite, rinkite and steenstrupine.

G. Flink (1849–1932) visited the complex in 1883. His mineral collections from Ilímaussaq were studied by O.B. Bøggild (1872–1956) and C. Winther (1873–1968), who established the new minerals britholite, epistolite and naujakasite, the last-named mineral as late as 1933.

The first thorough geological mapping and petrological investigation of the complex was by N.V. Ussing (1864–1911) in 1900 and 1908 (Fig. 8). His memoir on the geology and petrology of the complex and neighbouring areas was published in 1912 after his untimely death. The memoir gives a detailed interpretation of the petrology of the complex, which by and large still stands today. It introduced the term agpaitic and discussed processes such as overhead stoping, magmatic differentiation, igneous layering, assimilation and feldspar solid solution series. The memoir is one of the corner stones of igneous petrology. Bøggild (1913), who assisted Ussing in the field work, discovered and named the mineral ussingite.

The period 1912–1955

Research activities in the Ilímaussaq complex were very limited from the time of Ussing's memoir in 1912



Fig. 8. N.V. Ussing (left) and O.B. Bøggild (right) and their Greenlandic assistants at the cryolite mine at Ivittuut (Ivigtut), Greenland 1900. Photo: Geological Museum archive, Copenhagen.

until mineral exploration commenced in 1955. The main events are mentioned below:

S.M. Gordon visited the complex in 1923 and published three papers about his examination of mineral localities in South Greenland (Gordon 1924).

C.E. Wegmann (1938) studied the geological chronology of South Greenland and introduced the term Gardar period. He interpreted the rocks of the Ilímaussaq complex as the results of metasomatic processes.

The Cryolite Company (Kryolitselskabet Øresund A/S) undertook investigation of eudialyte-rich naujaite and kakortokite in 1939 and 1946 (Bøgvad 1950a, b),

but concluded that exploitation of the eudialyte was not feasible at that time.

The Geological Survey of Greenland (GGU), which was established in 1946, made reconnaissance visits to the complex in 1946 and 1951.

The period 1955–1964

In 1955 the Danish government, on the recommendations of Professor Niels Bohr, the President of the Danish Atomic Energy Commission (AEK), initiated prospecting for uranium deposits in Greenland (Fig.



Fig. 9. Professor Niels Bohr expressing his thanks for the honorary citizenship of Narsaq in 1957.

Fig. 10. The team from the University of Copenhagen which in 1964 initiated the university-directed Ilímaussaq project, and its Russian partners. From left to right: B. Leth Nielsen, J. Rose-Hansen, O.V. Petersen, H. Sørensen, E. Frantzen, V.I. Gerasimovsky, T. Østergaard, E.I. Semenov, K. Hansen and behind her E. Bondesen and S. Andersen. In the background from left to right Kvanefjeld, Ilimmaasaq, Narsaq Bræ, Nakkaalaaq with the Taseq plateau and slope, and the mountain Talut.



9). GGU recommended that prospecting for uranium should begin in the Ilímaussaq complex, the only known occurrence of radioactive minerals in Greenland at that time, apart from allanite in pegmatites. A primitive Geiger counter survey was carried out by military personnel. The southern half of the complex was covered in 1955, the northern half in 1956, in which year the Kvanefjeld deposit was discovered (Nielsen 1981; Sørensen 1981; Bondam 1995). In 1957 additional detailed studies of the Kvanefjeld deposit were made with chemical assays of the uranium ore and the first attempts at developing a method of extracting the uranium from the ore. The ore was found to be refractory and impossible to treat with conventional acid or carbonate leaching methods.

The first drilling programme was carried out in 1958 resulting in 36 holes and a total core length of 3728 m. The extraction experiments were continued in the following years and a method of sulphatising roasting of the ore was designed. In 1962 180 t of ore were taken out in a 20 m long adit in the most radioactive part of the deposit for testing the method of sulphatising roasting. In the same year two of the Kvanefjeld drill holes were deepened with an additional core length of 270 m and seven holes with a total core length of 1400 m were drilled in lujavrites in other parts of the complex.

AEK constructed a base camp at Dyrnæs in connection with the drilling operations in 1958. Dyrnæs is the site of a former Norse settlement located about 5 km to the north of the town Narsaq. This camp was transferred to GGU and served as the base for the geological mapping of the whole complex and for the regional geological mapping of South Greenland. The regional mapping programme was concluded in 1963 after which most of the camp was moved to the site for the next regional mapping programme further to the north. Some of the houses, however, remained in Dyrnæs to serve as the base camp for the follow-up geological investigations of the Ilímaussaq complex, which were entrusted to staff and students from the Geological Institute and Museum of the University of Copenhagen, in co-operation with and with logistic and economic support from GGU and AEK.

A geological map of the Ilímaussaq complex at 1:20 000 was published in 1964 (Ferguson 1964). Hamilton (1964) presented the first geochemical investigation of the northern part of the complex and Ferguson (1970) presented a detailed examination of the geochemistry of the kakortokites.

A number of mineralogical papers were published in the period 1955–1964. Examples are: notes about several minerals (Danø & Sørensen 1959), the discovery of the beryllium minerals chkalovite and tugtupite (Sørensen 1960, 1963), the first description of villiaumite from the complex (Bondam & Ferguson 1962), a detailed examination of the occurrence of steenstrupine (Buchwald & Sørensen 1961; Sørensen 1962) and the discovery of a number of opaque minerals (Oen & Sørensen 1964).



Fig. 11. Diamond drilling on the Kvanefjeld plateau in 1969. A fine view of the upper part of Narsaq Elv valley with the Narsaq Bræ in centre. The thin zone of light-coloured rock above the glacier is naujaite intruding the roof of volcanic rocks in the upper part of Nakkaalaaq mountain.

The period 1964-1977

Staff and students from the University of Copenhagen were, as mentioned above, entrusted with the detailed follow-up investigations in the Ilímaussaq complex. This activity was directed by H. Sørensen, J. Rose-Hansen and in the last phase of the project by B.L. Nielsen (Fig. 10). Field teams worked every summer from 1964 to 1977. About 25 persons took part in the field work and were supported by about 15 more in the follow-up laboratory studies and publication of results. A large number of field assistants and technicians took part in this work. Hydrogeologists, ecologists and geochemists were involved in the accompanying environmental studies.

From the early stages of this activity it was clear that much could be gained if Russian scientists became involved in the work. The Ilímaussaq complex bears a close resemblance to the Khibina and Lovozero complexes of the Kola Peninsula, two complexes which have been examined in great detail by Russian scientists resulting, among other things, in the discovery of a number of new minerals. Two Russian mineralogists, Professors V.I. Gerasimovsky and E.I. Semenov, who had made impressive contributions to the study of the mineralogy and geochemistry of the two Kola complexes, were therefore invited to take part in the Greenland field work in 1964. This gave a significant impetus to the new research programme. Gerasimovsky (1969) produced chemical analyses for major and trace elements of 23 rock samples representing the main rock types of the complex and Semenov (1969) described 120 minerals from the complex, among them five new minerals: chalcothallite, cuprostibite, ilimaussite, tundrite-(Nd) and sorensenite.

Major activities in the research programme of the university geologists were:

- 1. Geological mapping of the Kvanefjeld area (Sørensen *et al.* 1969, 1974). In connection with this work six exploratory holes totalling 1621 m were drilled in 1969 (Fig. 11), two of these in the lujavrites in the northern part of the plateau which had not been investigated in the earlier phases of uranium exploration. The reasonably assured uranium reserves were estimated to be 5800 metric t U, average grade 310 ppm U; additional reasonably assured ore with a grade of 292 ppm U was estimated to 8700 metric t U. In order to facilitate the geological supervision of the drilling programme and as a general support of the geological investigations in the Kvanefjeld area, a hut was set up in 1968.
- 2. Mapping of the kakortokites in the southern part of the complex with unravelling of the layered sequence. The resources of Zr and Nb were estimated to 51.6×10^6 metric t ZrO_2 and 5.4×10^6 metric t Nb₂O₅ in the examined part of the complex (Bohse *et al.* 1971).
- 3. Geological mapping and detailed examination of veins containing beryllium minerals in the north-

ern part of the complex (Semenov 1969; Engell *et al.* 1971).

- 4. Examination of the occurrences of U, Zr, Nb and Be minerals was carried out in close co-operation with scientists from the Danish Atomic Energy Commission Research Establishment Risø (now Risø National Laboratory) and resulted in development of apparatus to be used in the field and in the laboratory. Examples are: portable beryllium prospecting instruments (Løvborg et al. 1968a; Engell et al. 1971), portable X-ray fluorescence equipment for quantitative determination of Zr and Nb in the field (Bohse et al. 1971; Kunzendorf 1971, 1973), and gamma-spectrometers for use in the field, for assaying drill holes and drill cores and for laboratory determination of contents of U, Th and K in minerals and rocks (e.g. Løvborg et al. 1968b, 1972, 1980).
- 5. From 1968 to 1976, the Danish company Superfos A/S explored the eudialyte-rich kakortokites and naujaites in the southern half of the complex and developed methods to extract Zr, Nb, REE and Y from eudialyte concentrates, but found no markets for the products. To support the bulk sampling undertaken in 1968, a house was constructed at the mouth of Lakseelv in Kangerluarsuk. In subsequent years this house served as the base for many field teams working in this part of the complex.
- 6. A new geological map over the southern half of the complex in the scale of 1:20 000 (Andersen *et al.* 1988).
- 7. Collection of 120 samples for detailed geochemical analysis of contents of about 50 elements in whole rocks and separated mineral fractions (work still in progress, see Bailey *et al.* 2001, this volume).
- 8. Many studies of minerals and rocks collected in the series *Contributions to the mineralogy of Ilimaussaq* (updated in Rose-Hansen *et al.* 2001). The following new minerals were described in the period 1964–1977: sorensenite, chalcothallite, ilimaussite, tetranatrolite (described under the name tetragonal natrolite), tundrite-(Nd), semenovite, skinnerite, cuprostibite and rohaite (see list of minerals in Petersen 2001, this volume).

- A study of fluid inclusions in the minerals of the complex initiated in co-operation with Russian colleagues (Petersilie & Sørensen 1970; Sobolev *et al.* 1970) demonstrated that fluid inclusions of the agpaitic rocks, like the rocks of the Khibina and Lovozero complexes of the Kola Peninsula, are rich in hydrocarbons. This discovery was followed by detailed studies of the rocks of the complex, e.g. Konnerup-Madsen *et al.* (1979, 1988), Konnerup-Madsen & Rose-Hansen (1982), Konnerup-Madsen (2001, this volume).
- 10. An investigation of the water balance in the Narsaq Elv valley, which intersects the northern part of the complex, was carried out as one of the Danish contributions to the International Hydrological Decade (Hansen & Pulawski 1966; Larsen 1972, 1973).
- 11. An ecological and environmental geochemical programme, the Narsaq Project, supported by the Danish Natural Science Research Council was carried out 1974-1977 (Larsen 1977; Rose-Hansen & Sørensen 1977; Rose-Hansen et al. 1977; Nielsen 1979). The project was initiated at a time when exploitation of the Kvanefjeld uranium deposit was considered possible within a few years. The purpose of the project was to describe the natural state of the environment around the Ilímaussaq complex before the opening of a uranium mine. The project also had the aim to study the distribution of rare elements, including uranium, around the complex, which may be considered a marked geochemical anomaly (Rose-Hansen et al. 1986). As described in a later paragraph, uranium mining in the area was given up for political reasons, which meant that the Narsaq Project was also discontinued.

1977 to the present

The 1977 field season marked the termination of the field activities in Ilímaussaq by staff and students from the University of Copenhagen. The Dyrnæs base camp was thereafter used to support other activities such as the *Kvanefjeld Uranium Project* and the *Syduran Project* (see below). The base was abandoned in 1983 with the termination of the *Kvanefjeld Uranium Project*. This reduced the logistic support of field work in the complex, but minor operations have nevertheless con-

tinued. One example is mineralogical studies including examination of material from the tunnelling and drilling of the Kvanefjeld uranium deposit mentioned below. This resulted in the discovery of the new minerals vitusite, kvanefjeldite and tuperssuatsiaite. A progress report bringing results of field and laboratory studies up to 1980 was published in 1981 (Bailey *et al.* 1981).

The Kvanefield Uranium Project was carried out from 1978 to 1983 with the aim of examining the economic potential of the Kvanefjeld uranium deposit. In 1977, this project was preceded by a drilling programme comprising 27 holes with a total core length of 5103 m in the lujavrites in the northern part of the Kvanefjeld plateau and to the east of Kvanefjeld (Nyegaard et al. 1977). It was found that the method of sulphatising roasting applied to the steenstrupinebearing lujavrites in the northern part of the Kvanefjeld plateau gave a low recovery of uranium, whereas pressurised carbonate leaching gave a satisfactory recovery. In order to test this method in a pilot plant established at Risø National Laboratory, 20 000 metric t of ore were extracted from a 960 m long horizontal adit driven through the deposit (Nyegaard 1980). The opening of the adit was in the slope above the Narsaq Elv valley 100-150 m below the surface of the plateau (Nyegaard 1979). In total 4700 metric t of ore were shipped to Risø and treated in the pilot plant. It was found that the method gave a recovery of more than 80% of the uranium content of the different varieties of ore (Forsøgsanlæg Risø 1984; Sørensen & Jensen 1985; Sørensen et al. 1990). The reasonably assured resources were estimated to 20 440 metric t U in ore with an average concentration of 365 ppm U (Forsøgsanlæg Risø 1984). Detailed mineralogical studies of the uranium ore were carried out in connection with this project (Makovicky et al. 1980).

The *Kvanefjeld Uranium Project* also presented proposals for the planning of the mine and the ore dressing facilities and energy supply, as well as studies of radiation exposure and the environmental impact of the mining activity, including the effects of leaching of tailing products (Pilegaard 1990). Overviews of the many internal reports of this project are found in the report on the project (Forsøgsanlæg Risø 1984) and in Bondam (1995).

In the years 1979–1982, the *Syduran Project* carried out a regional exploration for uranium in South Greenland by means of airborne radiometric surveying and stream sediment geochemistry. This work confirmed the anomalous character of the complex and its surroundings (Armour-Brown *et al.* 1983, 1984; Thorning *et al.* 1994; Schjøth *et al.* 2000).

Exploration of the zirconium-rich kakortokites continued in 1985, when the Danish company A/S Carl Nielsen obtained an exclusive licence to carry out exploration centred around the exposed kakortokites and the adjacent marginal pegmatite in the southern part of the complex. The thickest layer of red kakortokite, layer +16, was examined in two drill holes in 1986. During 1987, potentially economic eudialyterich parts of the marginal pegmatite, kakortokites and naujaites within the concession area were mapped and sampled, and samples of the marginal pegmatite were metallurgically tested.

In 1987, the Canadian company Highwood Resources Ltd. obtained permission to explore areas between the fjords Tunulliarfik and Kangerluarsuk and carried out bulk sampling and drilling in order to test the feasibility of exploitation of eudialyte-rich rocks. This company was joined by Platinova Resources Ltd. and Aber Resources Ltd. In 1988 this group and A/S Carl Nielsen formed a joint venture, combining their mineral licences. The main target was the exposed kakortokites, minor targets were the marginal pegmatites in the southern part of the complex. The joint venture co-operation was continued in 1990 with an extensive drilling programme and metallurgical testing of potential ores from the southern part of the complex. At the end of this activity the Canadian partners and the Danish participants went through a period of restructuring resulting in Highwood Resources taking over all interests in the prospect at the end of 1992.

In 1992 the Danish company Mineral Development International A/S (MDI) obtained the exclusive right to explore the sodalite-rich naujaites in the northern part of the complex. The aim was to investigate the possibilities of using sodalite as raw material for the production of synthetic zeolites.

None of the above-mentioned activities have so far been able to demonstrate with certainty that beneficiation of eudialyte and sodalite can be economically viable.

A number of research projects involving colleagues from other countries have been supported by various foundations. The Danish Natural Science Research Council supported a Canadian–Danish project aiming at a comparison of the mineralogy of Mont Saint-Hilaire, Quebec, with the Narssârssuk mineral occurrence associated with the Igaliko Complex, South Greenland, and the Ilímaussaq complex. Fig. 12. Destruction of one of the tugtupite localities by mineral collectors' excessive use of blasting. The Kvanefjeld plateau July 1974. Scale: Professor Brian Mason, Washington DC, USA, one of the many scientists who have visited the Ilímaussaq complex. Since the photograph was taken this site has been transformed into a veritable crater, about 20 m in diameter and a few metres deep, as a result of the hunt for deeper-lying tugtupite veins.



The Danish company First Development International A/S in 1993 supported a Danish–Russian project consisting of an examination of the drill cores from the 1977 drilling programme kept at the Risø National Laboratory. The aim was to find some of the water soluble minerals discovered in the Khibina and Lovozero complexes (Khomyakov 1995). The drill cores are rich in villiaumite, but holes in the samples indicate that other water soluble minerals have been dissolved during and after drilling. Only one of the Kola minerals was discovered, natrophosphate (Petersen *et al.* 2001, this volume).

In 1994–1997 INTAS (International Association for the Promotion of Co-operation with Scientists from the Independent States of the Former Soviet Union) supported a Danish–French–Russian–Spanish research co-operation with the purpose of promoting comparative studies of the mineralogy of agpaitic nepheline syenites in Ilímaussaq, the Khibina and Lovozero complexes of the Kola Peninsula, and the Tamazeght complex, Morocco. Field work was carried out in Ilímaussaq in 1994, in Khibina and Lovozero in 1997 and in Tamazeght in 1999. One of the outcomes of this work is the paper on hiortdahlite in this volume (Robles *et al.* 2001).

The Danish Natural Science Research Council in 1997 supported an Austrian–Danish research project with the purpose of studying pegmatites and hydrothermal veins and the relations to their country rocks in the Ilímaussaq complex and at the Narssârssuk mineral locality associated with the Igaliko Complex in South Greenland.

A number of excursions, workshops and summer

schools have taken place in the Ilímaussaq complex since 1981:

- 1981, excursion arranged for the directors of the European geological surveys.
- 1982, excursion for Société Minéralogique de la France.
- 1984, summer school on environmental geology sponsored by the Nordic Council of Ministers (Nordisk Ministerråd 1984).
- 1986, NATO Advanced Research Workshop on Igneous Layering (Parsons 1987).
- 1989, excursion for colleagues from Naturhistorisches Museum and Österreichisches Mineralogische Gesellschaft.
- 1990, Nordic summer school on igneous petrology.
- 1992, ABC Mines from École des Mines, Paris, an excursion to Iceland and the Igaliko and Ilímaussaq complexes.

A number of mineral collectors and societies of mineral collectors have visited the complex resulting in the discovery of the minerals bavenite, dorfmannite, fersmite, nacareniobsite-(Ce) and turkestanite. This activity has put heavy pressure on many of the mineral localities in the complex resulting in transformation of many of them into heaps of boulders (Fig. 12).

A significant part of the research carried out in the complex has resulted in the awarding of academic degrees in mineralogy and geology by universities in Denmark and abroad. The Danish awards comprise: three degrees of dr.scient., six lic.scient. and Ph.D. degrees, 18 cand.scient. degrees in geology, five cand. scient. and four Ph.D. degrees in biology and ecology as a spin-off of the *Narsaq Project*; three scientific papers have been awarded the gold medal of the University of Copenhagen. The number of degrees awarded in other countries is not known with certainty.

Concluding remarks

An impressive number of papers have been published on the geology, mineralogy, petrology and geochemistry of the Ilímaussaq alkaline complex (Rose-Hansen *et al.* 2001). Major exploration programmes have investigated the economic potential of rocks rich in uranium, zirconium, niobium and beryllium and the technical use of sodalite. Much remains, however, to be investigated and published.

The southern half of the complex has been mapped in the scale of 1:20 000; the northern half should be mapped in the same detail.

In order to gain a fuller understanding of the petrogenesis of the complex a number of drill holes are required, first of all in the deepest part of the kakortokites to explore the hidden layered floor series, and through the roof series to give access to the sheets of augite syenite, alkali granite, etc. occurring in a topography which makes access difficult. Many aspects of the geology of the complex have not yet been studied in detail, this applies for instance to the spectacular layering of some of the arfvedsonite lujavrites. Future drilling programmes and quarrying activities should take special measures to safeguard the watersoluble minerals because these must be collected immediately on exposure to the atmosphere.

The agpaitic nepheline syenites are among the most evolved igneous rocks known. Petrological studies of the rocks of the complex can therefore bring important knowledge about many natural petrological processes.

The Ilímaussaq complex contains a treasure of rare elements and minerals. Future developments in material sciences and the need for rare elements in new applications should therefore be followed closely in order to be ready when new opportunities become apparent for use of elements abundant in the complex.

The Ilímaussaq complex is vulnerable if exposed to invasions of mineral collectors, local as well as foreign visitors (Fig. 12). Some mineral occurrences have already been exhausted, others destroyed; an example is the tugtupite occurrence in the south-western part of the Kvanefjeld plateau (Sørensen 1997b). It may be necessary to regulate the collection of minerals in the complex. On the other hand, the complex elucidates many geological processes in a very clear and informative way and should therefore be open for excursions, summer schools, etc. and be a show window for the geological sciences.

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