# RECONNAISSANCE STUDIES ON THE SYNTHESIS AND STABILITY OF STEENSTRUPINE

## Contribution to the mineralogy of Ilimaussaq no. 75

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SØRENSEN, HENNING; MAKOVICKY, MILOTA and ROSE-HANSEN, JOHN, 1985: Reconnaissance studies on the synthesis and stability of steenstrupine. Contribution to the mineralogy of Ilimaussaq no. 75. *Bull. Geol. Soc. Finland 57, Part 1—2, 103—112.*

Steenstrupine, a complex silicophosphate of sodium, the lanthanides, calcium, manganese, iron, zirconium and thorium, is the principal uranium-bearing mineral (about 0.1 % U) in the Kvanefjeld uranium deposit in the northern part of the Ilímaussaq alkaline complex, South Greenland. The steenstrupine occurs in a number of varieties in lujavrites, in fenitized country rocks and in pegmatites and hydrothermal veins, thus spanning a range of conditions of formation. Its geological occurrence indicates that steenstrupine is formed only in sodium-rich rocks containing albite, analcime and other Na-rich minerals.

The stability conditions for steenstrupine have been investigated in a series of hydrothermal runs at 1000 atm and 400°—700°C using »synthetic» steenstrupine compositions produced from pure chemicals and gel charges obtained by decomposition of steenstrupine as starting materials. The hydrothermal solutions were aqueous solutions of NaOH and NaCl of various compositions.

It was found that steenstrupine at 1000 atm may form in the whole temperature range investigated, 400°—700°C, its formation being determined by the chemical composition of the initial solid phases and of the solutions. It formed only when the starting material was sodium-rich and in the case of »synthetic» compositions only when the starting material was rich in Zr.

Other minerals produced in the experiments were: monazite, lessingite and britholite.

The results of the experiments are in accordance with the field observations and petrological evidence for the occurrence of steenstrupine and monazite. Britholite has been observed rarely in the rocks of Ilímaussaq, lessingite has not been indentified to date in this locality.

Key words: steenstrupine, Ilímaussaq complex, uranium deposit.

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## Introduction

Steenstrupine, a complex silicophosphate of sodium, the lanthanides, calcium, manganese, iron, zirconium and thorium, represents the main uranium mineral in the rocks of the Kvanefjeld uranium deposit in the northern part of the Ilímaussaq alkaline complex (Sørensen 1962; Sørensen *et al.* 1974; Makovicky *et al.* 1980).

The Ilímaussaq alkaline complex is one of a number of intrusive complexes of the Gardar rift system in South Greenland, which was active 1300 to 1100 m.y. ago. The complex is made up of a variety of alkaline rocks and predominantly of agpaitic nepheline syenites (see Bailey *et al.* 1981a for a recent review). The Kvanefjeld uranium deposit and other uranium occurrences in the complex are associated with lujavritic rocks enriched in a number of rare elements (Sørensen *et al.* 1974).

Steenstrupine occurs in a number of forms. For the purpose of the present paper, it is convenient to distinguish three main types.

1. Type A occurs as small mm-sized crystals in the arfvedsonite and naujakasite lujavrites.

2. Type B steenstrupine occurs as large (often >1 cm), irregular poikilitic grains and is especially common in medium- to coarse-grained lujavrites and their surrounding rocks.

3. Late veins of analcime, albite, etc. contain a third variety of steenstrupine that often forms large well developed crystals, several cm across. This variety is termed type C.

These variable occurrences imply a range in the conditions of formation of steenstrupine. This, together with the potential practical importance of the mineral, makes it interesting to attempt its synthesis and to study the pressure, temperature and compositional conditions for its stability.

The observed broad compositional variability of steenstrupine is still being studied. However, reliable compositional data on steenstrupine from a number of localities have been obtained by Makovicky *et al.* (1980) and Makovicky & Karup-Møller (1981).

Very few data on the P—T conditions for the crystallization of the Ilímaussaq rocks were available when the present study was initiated (Piotrowski and Edgar 1970; Sood and Edgar, 1970). Although general conditions of formation for the Ilímaussaq intrusion were proposed by Sørensen in 1962, only recently have more precise data become available (Konnerup-Madsen *et al.* 1979, 1981a).

The association between steenstrupine and the youngest Ilímaussaq agpaitic rocks with augmented U and Th contents has long been recognized (see Bailey *et al.* 1981b for references). The importance of an especially high Na concentration for its formation has been demonstrated experimentally by Konnerup-Madsen *et al.* (1981a) and has been discussed by Makovicky and Karup-Møller (1981) and others.

The three types of steenstrupine mentioned above illustrate the range of conditions of formation of this mineral.

Type A steenstrupine, which forms well-developed crystals of the same size and mode of occurrence as the much more wide-spread rare metal silicate eudialyte, is a mineral of primary magmatic origin (Buchwald and Sørensen 1961; Sørensen 1962 and Engell 1973). Eudialyte is the stable primary lanthanide — zirconium mineral in the earliest aegirine lujavrites and the early arfvedsonite lujavrites. In the late-stage arfvedsonite and naujakasite lujavrite, steenstrupine crystals appear instead.

Microcline and nepheline are the predominant salic minerals in the early lujavrites, albite and analcime are predominant in the late ones. The early lujavrites may have minor steenstrupine, often along the margins of eudialyte crystals. The late steenstrupine lujavrites may have scarce grains of eudialyte and often exhibit small pigmentary clusters that may be pseudomorphs after eudialyte (Sørensen 1962). A similar relation is seen in naujaite pegmatites in which primary crystals of eudialyte may have

rims of steenstrupine, especially in albitized parts of the pegmatites.

These features indicate that eudialyte is a stable liquidus mineral at higher temperatures, whereas steenstrupine crystallizes at lower temperatures from residual magmas enriched in the lanthanides, uranium, thorium, phosphorus, etc.

Type B steenstrupine has been described by Makovicky et al. (1980) from the Kvanefjeld area, where it occurs as large poikilitic grains in the latest lujavrite phase of the whole intrusion, the medium- to coarse-grained lujavrites, and in their aureoles of fenitized volcanic rocks and earlier fine-grained lujavrites. It appears to be connected with the pegmatitic and metasomatic processes that produced these lujavrites and their aureole. This type of steenstrupine was also described by Sørensen (1962) from green and brown lujavrites of the north coast of Tunugdliarfik and from the contact zones between lujavrites and their naujaite xenoliths. These poikilitic types of steenstrupine are especially common in lujavrites in which analcime is the predominant salic mineral. Type B steenstrupine may thus be regarded as a result of extended crystallization of steenstrupine with decreasing temperature and increasing partial pressure of volatiles.

The poikilitic grains of steenstrupine are often altered into an »aggregate» of monazite, thorite, neptunite, pigmentary material and a number of other minerals (see Sørensen 1962 and Sørensen *et al.* 1974; Makovicky *et al.* 1980). The present study sheds new light on the formation of these aggregates.

Type C steenstrupine forms large crystals in late veins of albite, ussingite, sodalite, analcime, etc. These grains may be replaced by acmite, uranothorite, monazite and unidentified fine-grained alteration products.

In order to specify more closely the range of stability conditions for steenstrupine a series of hydrothermal runs using starting compositions close to or identical with that of steenstrupine

were conducted under conditions approximating those given by the authors cited above.

## Experimental methods and materials

### Procedures for hydrothermal runs

Hydrothermal runs were done in 12-inch »cold-seal» bombs (Tuttle 1949) heated externally in vertical resistance furnaces. Cooling by compressed air and immersion in water resulted in a quenching time to room temperature of less than two minutes. Temperatures were measured with calibrated internal thermocouples and are considered accurate to within $\pm 2°C$ of the recorded value. Measurements indicate that the thermal gradient in the sample region did not exceed $10°C$. Pressure was measured with a transducer and is considered accurate to $\pm 10$ bars.

Charges consisted of finely ground ($< 74 \mu m$) material of steenstrupine composition (ca. 40 mg) and 30—40 mg of reaction solution. The charges were sealed in welded gold capsules.

### Steenstrupine starting compositions

Two types of starting material of steenstrupine composition were prepared for the hydrothermal runs: 1) melted charges prepared from pure chemicals, and 2) gel charges obtained from decomposition of natural steenstrupine.

### The synthetic, melted preparations

The first type of starting material was prepared by melting mixtures of chemicals weighed in the proportions observed in natural steenstrupine.

The model chemical composition of the starting material was that of steenstrupine sample GGU no 199104 (Makovicky and Karup-Møller, 1981) with the empirical formula $H_{6.96}$ $Na_{3.41}$ $Ca_{1.04}$ ($La_{1.98}$ $Ce_{2.88}$ $Pr_{0.23}$ $Nd_{0.71}$ $Gd_{0.02}$ $Sm_{0.02}$ $Er_{0.01}$

Table 1. Synthetic starting materials I.

| Composition 7 I | | Composition 7 II | |
|---|---|---|---|
| Atomic ratio | Molar ratio | Atomic ratio | Molar ratio |
| Si 11.3 | 5.65 $SiO_2$ | Si 11.3 | 5.65 $SiO_2$ |
| P 6.7 | 3.35 $H_3PO_4$ | P 6.7 | 3.35 $H_3PO_4$ |
| Na 12.8 | 3.20 $Na_2CO_3$ | Na 1.9 | 0.48 $Na_2CO_3$ |
| Ca 1.0 | 0.5 $CaCl_2 \cdot 2H_2O$ | | |
| Mn 1.8 | 0.9 Mn | | Remaining elements as in 7 I |
| La 2.44 | 0.61 $La_2O_3$ | | |
| Ce 3.56 | 0.89 $Ce_2O_3$ | | |
| Zr 1.00 | 0.5 $ZrO_2$ | | |

Procedure: $H_3PO_4$ was slowly added to the wet mixtures of $SiO_2$ and $Na_2CO_3$. The resulting mixture was slowly dehydrated at fairly strong heat. After adding the remaining components the product was melted in an oven at 1150°C.

Table 2. Synthetic starting materials II.

| Composition 9 I | | Composition 9 II | |
|---|---|---|---|
| Atomic ratio | Molar ratio | Atomic ratio | Molar ratio |
| Si 11.30 | 5.65 $SiO_2$ | Si 11.30 | 5.65 $SiO_2$ |
| P 6.70 | 3.35 $H_3PO_4$ | P 6.70 | 3.35 $H_3PO_4$ |
| Na 12.80 | 3.20 $Na_2CO_3$ | Na 1.90 | 0.48 $Na_2CO_3$ |
| Ca 1.00 | 0.5 $CaCl_2 \cdot 2H_2O$ | Ca 1.00 | 0.5 $CaCl_2 \cdot 2H_2O$ |
| Mn 1.80 | 0.9 Mn | Mn 1.80 | 0.9 Mn |
| La 2.44 | 0.61 $La_2O_3$ | La 2.44 | 0.61 $La_2O_3$ |
| Ce 3.56 | 1.78 $CeCl_3 \cdot 7H_2O$ | Ce 3.56 | 1.78 $CeCl_3 \cdot 7H_2O$ |
| Zr 1.00 | 0.5 $ZrO_2$ | Zr 1.00 | 0.5 $ZrO_2$ |

The mixture of above chemicals was melted in an oven at 1400°C for 8 hours.

$Y_{0.19}$) $Th_{0.48}$ $U_{0.05}$ ($Zr_{0.41}$ $Mn_{1.80}$ $Fe_{1.69}$ $Al_{0.07}$ $Ti_{0.10}$) ($Si_{13.39}$ $P_{4.61}$) $O_{60}$ (F, OH) $\cdot$ 14.6$H_2O$.

The Na content was varied in accordance with the range reported by Makovicky *et al.* (1980), that is, between 1 and 12 atoms in the above formula.

In the initial runs, the starting compositions were simplified as follows: the total minor REE contents were proportionally distributed between La and Ce; octahedrally coordinated metals were simplified to either Mn and Fe or Mn, Fe and Zr or Mn and Zr. Th was omitted and the amounts of Na, as well as the ratio of the tetrahedrally coordinated elements Si and P were adjusted to maintain the charge balance (Tables 1 and 2).

Table 3. Synthetic starting materials III.

| Composition 20 | | |
|---|---|---|
| Atomic Proportions | Weighed amounts | |
| 13.386 Si | 0.8043 g | $SiO_2$ |
| 4.614 P | 0.6598 g | $Na_2HPO_4$ |
| 1.386 Na | 0.1469 g | $Na_2CO_3$ |
| 1.044 Ca | 0.0892 g | $CaCl_2 \cdot 2H_2O$ |
| 0.437 F | 0.0341 g | $CaF_2$ |
| 4.064 Ce | 0.7167 g | $CeO_2$ |
| 0.535 Th | 0.1470 g | $ThO_2$ |
| 1.752 Fe | 0.0979 g | Fe |
| 1.976 La | 0.6438 g | $La_2O_3$ |
| 0.508 Zr | 0.0626 g | $ZrO_2$ |
| 1.802 Mn | 0.0990 g | Mn |

Annealed at 1100°C for 20 hours. After cooling annealed at 1450°C for 10 hours. Quenched in the $Hg/H_2O$ quenching arrangement and pulverized.

In subsequent experiments, a composition close to that of the model natural steenstrupine was used. However, the minor lanthanides were replaced by Ce, and U was replaced by Th (Table 3).

The desired proportions of the compositions were weighed and thoroughly mixed. One or two fusions of several hours duration at temperatures varying from 1150° to 1450°C were conducted in platinum crucibles. Quenching in a few seconds was obtained by dropping the platinum crucibles straight into a water-ice mixture. Although the mixing and melting procedures should have produced homogeneous glass of a simplified steenstrupine composition, crystalline phases were always present after quenching. Further details concerning the different glass compositions prepared are given in the explanations to Tables 1 to 3.

## The gel/salt charges obtained by decomposition of steenstrupine

Relatively large amounts of type C steenstrupine (GGU 199104) from albititic veins in Tunugdliarfik also provided starting material for steenstrupine synthesis.

A steenstrupine fraction with a density greater than 3.16 $g/cm^3$ was separated and purified. Decomposition was achieved at moderate temperatures by repeated addition of 6N HCl. After complete decomposition, the resulting gel-like substance and solution were diluted with water and neutralized to pH 7 with NaOH. This solution was evaporated slowly by extended heating at 60°C. The resulting precipitate was then heated at 100°C to remove most of the water.

Table 4. Runs based on synthetic mixtures.

| Run no. | Solid starting composition | Solution (wt%) | Conditions (p = 1000 atm) | Final products |
|---|---|---|---|---|
| M 3A | low-Na, Mn-Fe-Zr | $H_2O$ | 700°C, 7 days | Mon + $CeO_2$ + NaCl + ? |
| M 4A | high Na, Mn-Fe-Zr | $H_2O$ | 700°C, 7 days | Mon + $CeO_2$ + NaCl + Steen |
| M 5A | low-Na, Mn-Fe | $H_2O$ | 700°C, 7 days, | Mon + $CeO_2$ + NaCl + ? |
| M 6A | high Na, Mn-Fe | $H_2O$ | 800°C, 7 days | Mon + $CeO_2$ + NaCl |
| S 7A | Na-rich, Mn-Zr compos. 7I | $H_2O$ | 500°C, 8 days | Mon + Steen + $CeO_2$ |
| S 7B | Na-poor, Mn-Zr compos. 7II | 50 % NaCl | 500°C, 8 days | Mon + Steen + $CeO_2$ + ? Less |
| S 9I | Na-rich, Mn-Zr compos. 9I | 43 % NaCl + 14 % NaOH | 500°C, 14 days pH = 7.5—8 | Steen + Mon + $CeO_2$ + ? |
| S 9II | Na-rich, Mn-Zr compos. 9I | 43 % NaCl + 14 % NaOH | 700°C, 14 days pH = 9.5 | Steen + Mon + $CeO_2$ + ? |
| S 9III | Na-poor, Mn-Zr compos. 9II | 43 % NaCl + 14 % NaOH | 500°C, 14 days | Mon ± $CeO_2$ |
| S 14II | Na-rich, Mn-Zr compos. 7I | 46 % NaCl | 500°C, 18 days | Mon + Steen + $CeO_2$ |
| S 21-1 | full compos. 20 with Th (Less + $CeO_2$ + ?) | 2 % NaOH 2 % NaCl | 500°C, 23 days | Less + Steen + $CeO_2$ + NaCl + (Mon) |
| S 21-2 | full compos. 20 with Th (Less + $CeO_2$ + ?) | 2 % NaOH 2 % NaCl | 400°C, 23 days | Less + Steen + $CeO_2$ + NaCl + Mon |

Abbreviations: Mon = monazite, Steen = steenstrupine, Less = lessingite

## Compositions of experimental solutions

In addition to the solid material of steenstrupine composition, the charges consisted of added distilled water or aqueous solutions of NaOH, NaCl and mixtures of these. The nature of the added solution is given in Tables 4 and 5 together with the experimental conditions. The pH of the experimental solutions indicated in the tables was measured immediately after the post-run opening of the capsules.

## Identification of run products

The very fine-grained nature of the run products prevented ordinary microscopic examination. The lack of good crystals also prevented quantitative electron microprobe analysis. Powder diffraction analysis was therefore the principal method used in the identification of the run products.

The Guinier patterns were obtained with Guinier-Hägg cameras at the Institute for Mineralogy, University of Copenhagen. Cu $K_\alpha$ radiation and in most cases quartz internal standard were used.

Because of the large number of potential phases and considerable line overlap, the deciphering of the diffractograms is difficult and sometimes ambiguous. Attention was therefore concentrated on the REE/Zr/Th-bearing phases. The range of Na-silicates and Mn and Fe silicates (e.g. acmite) is too large to allow their determination from the few remaining lines not overlapped by the »major» REE/Zr/Th phases.

Among the *REE-rich phases,* monazite can only be determined unambiguously when it is a major phase. Its presence becomes questionable when it is a minor phase with steenstrupine and lessingite. Minor amounts of lessingite and britholite are not recognizable if large amounts of monazite and steenstrupine are present. Lessingite and britholite showed variation in their $d$ values, suggesting variations in chemical composition (Strunz 1944, Ito 1968).

Steenstrupine is the most difficult phase to identify. The powder patterns of natural steenstrupine materials are generally very weak and unreliable because of extensive metamictization. Indexed patterns from single crystal photographs published by Makovicky and Karup-Møller (1981) may contain traces of acmite (per-

Table 5. Runs based on the steenstrupine decomposition products.

| Run No. | Medium (wt %) | Conditions (p = 1000 atm) | Solid run products |
|---|---|---|---|
| S 8I | $H_2O$ | 500°C, 5 days, pH = 1.5 | Mon + NaCl |
| S 8II | $H_2O$ | 700°C, 5 days, pH = 1.5 | Mon + NaCl |
| S 8III | $H_2O$ | 700°C, 7 days, pH = 1.5 | Mon + NaCl |
| S 8V | $H_2O$ | 400°C, 7 days, pH = 1.5—2 | Mon + NaCl |
| S 8VI | $H_2O$ | 600°C, 7 days, pH = 2—3 | Mon + $FeCl_2 \cdot 4H_2O$ + NaCl |
| S 10I | 36 % NaOH sol.[1] | 500°C, 12 days, pH = 7.5—8 | Mon + Steen + ? Less + NaCl + ? $CeO_2$ |
| S 10II | 39 % NaOH sol.[1] | 700°C, 12 days | Mon + Steen + NaCl |
| S 15I | NaOH/charge ≐ 1.4:1 | 500°C, 14 days, pH = 9 | Mon + Steen + $CeO_2$ + NaCl + ? |
| S 15II | NaOH/charge = 0.8:1 + $CaF_2$ | 500°C, 15 days, pH = 9 | Brith + $CaF_2$ + NaCl |

[1]  NaOH/charge = 0.45:1

Abbreviations: Mon = monazite, Steen = steenstrupine, Less = lessingite, Brith = britholite

sonal communication). The 15 Å line of the synthetic steenstrupine is very weak and was identified positively in only one case. Thus, steenstrupine identification in this work is based on sets of $d$ values in agreement with the $d$ values given by Makovicky and Karup-Møller (1981) (Tables 4 and 5). In particular, both the 7 Å line and several other lines with high $d$ values recognized in natural steenstrupine must be present.

## Results of experimental study

The starting materials used permitted the variation of both physical (temperature) and chemical (starting compositions, types of solution) parameters in the hydrothermal runs. All runs were made at a pressure of 1000 atm, the value considered appropriate for the final solidification of the late agpaitic rocks of the Ilímaussaq intrusion.

### Results of steenstrupine syntheses with synthetic, melted compositions

The chemical variability of the glass charges prepared allowed both the possible influence of steenstrupine composition and solution composition to be evaluated. The solid starting compositions, the solution composition and the experimental parameters are given in Table 4 together with the identified run products.

The Mn-Fe-Zr and the Mn-Fe compositions (with Zr replaced by Fe) were used in the runs with $H_2O$ as the reaction medium (runs M3-M6, Table 4). Low-Na and high-Na compositions were studied in both of the above compositional groups; they were all run at 700 or 800°C. In all four runs, monazite plus $CeO_2$ were the main solid products. Steenstrupine was only positively identified in the high-Na, Fe-Mn-Zr charge, suggesting that both Na and Zr are important for its formation (run M4, Table 4).

For compositions with high-Na and Mn + Zr

(nos. 7I and 9I, Tables 1 and 2) the following conditions were examined: (1) runs with distilled water as reaction medium at 500°C (run S7A, Table 4), and (2) runs with a 43 wt.% NaCl + 14 wt. % NaOH solution as reaction medium at 500°C and 700°C (runs S9 I — S9 II). Monazite, steenstrupine and $CeO_2$ were produced in all the experiments. The highest steenstrupine yield was obtained at 700°C.

Two runs with Na-poor Mn-Zr composition (glass composition 7 II and 9 II; Tables 1 and 2, respectively) in NaCl-rich solution (runs S7B and S9 III) gave different products. Both are rich in monazite and $CeO_2$, but in run S7B, steenstrupine and perhaps lessingite were present.

The starting glass approximating the composition of the model natural steenstrupine (composition 20, Table 3) contained microlites of lessingite and $CeO_2$. Two hydrothermal experiments (S21-1 and S21-2) at 500°C produced solid products consisting of steenstrupine, lessingite and minor amounts of $CeO_2$ and monazite.

### Results of steenstrupine syntheses with the gel/salt starting mixture

In order to study the formation of steenstrupine as a function of the concentration of the $Na^+$ available, NaOH was added to the gel/salt charges. Runs were made with weight ratios of NaOH to solid steenstrupine of 0, 0.45 and 1.4 respectively (Table 5). Final solution pH values were 1.5—3, 7.5—8 and 9, respectively.

The only phase detectable in the solid products of runs S8 I to S8 VI (Table 5) under acid reaction conditions was siliceous monazite. In addition, amorphous silica (or Na silicate) was detected by microprobe analysis. Identical results were obtained in the runs at 400°C, 500°C, 600°C and 700°C.

In runs with a weight ratio of NaOH to starting steenstrupine material equal to 0.45 (runs

S10 I and S10 II), only monazite and steenstrupine (identified by X-ray diffraction) were present in the solid products at 700°C. Lessingite and $CeO_2$ were tentatively identified in the 500°C experiment.

With a ratio of 1.4 (run S15 I), and consequently a highly alkaline residual medium, steenstrupine prevailed over monazite. $CeO_2$ and poorly developed lessingite were also present (Table 5).

Finally, in one run (S15 II) at 500°C with a NaOH/starting material ratio of 0.84, excess $CaF_2$ was added to the charge. Britholite and recrystallized fluorite were the solid run products.

## General conclusions with regard to the stability of steenstrupine

The present study permits the following general conclusions to be drawn as to the stability relations of steenstrupine:

1. At a pressure of 1000 atm, steenstrupine may form under hydrothermal conditions at all the examined temperatures from 400°C to 700°C.

Its formation depends on both the composition of the initial solid phases and the composition of the associated solution.

2. With a gel/salt mixture of natural steenstrupine composition, steenstrupine only forms in neutral to alkaline solutions. In acid solutions, monazite and colloidal silica are formed, the remaining elements staying in the residual solution. Addition of $CaF_2$ to an alkaline solution results in formation of britholite and fluorite, not steenstrupine.

3. With synthetic glass as a starting material, steenstrupine only forms from Zr-bearing compositions rich in Na, irrespective of the character of the hydrothermal solution. Low-Na, Zr-rich glasses do not yield steenstrupine, not even in Na-rich solution. Glass compositions without Zr do not yield steenstrupine. Thus

high contents of Na and Zr are needed for the hydrothermal formation of steenstrupine.

These general conclusions are in accordance with field observations and petrological evidence for the occurrence of steenstrupine.

With respect to its chemical composition, steenstrupine can be expected to crystallize from melts with higher relative contents of lanthanides than eudialyte, especially when enough Th is present to compete with Zr. Indeed steenstrupine appears to succeed eudialyte in the lujavrites and in the naujaite pegmatites when they become impoverished in Zr and Ca and enriched in Th and U (Engell 1973).

To date lessingite has so far not been identified in the rocks of the Ilímaussaq complex, whereas monazite is of widespread occurrence, especially in the lujavrites.

The presence of highly saline, NaCl-dominated aqueous fluid inclusions in minerals from these rock types indicates high Na-contents of an associated solution. The results obtained are consistent with the general observation that the common U-Th-REE minerals in granites and non-agpaitic nepheline syenites are the corresponding oxides, whereas U-Th-REE silicophosphates such as steenstrupine are limited to agpaitic nepheline syenites. Granites and non-agpaitic nepheline syenites may have high Zr-levels in late-stage fractionates. The lack of complex minerals such as steenstrupine in these rocks might be due to excessively low Na concentrations.

Finally, our experiments suggest reasons for the above mentioned replacement of steenstrupine by well-crystallized monazite associated with uranothorite and other products in some parts of the Ilímaussaq massif. These reactions were apparently connected with decreasing pH values and/or Na contents of the fluids that permeated the rocks in the late stages of their formation (see also Konnerup-Madsen *et al.* 1981).

The frequent presence in the Ilímaussaq lujavrites of »pseudomorphs» after eudialyte (?)

made up of aggregates of small crystals of monazite having very low contents of U and Th (Danø and Sørensen 1959) may also be explained in the light of the experimental data as a result of late magmatic reactions in an environment enriched in rare earth elements, poor in U and Th, and relatively poor in Na. It should be emphasized that the formation of eudialyte requires a sodium-rich environment (Cristophe-Michel Levy 1961 and Ahmed and MacKenzie 1978), in which eudialyte is substituted by catapleiite with decreasing temperature.

The presence or absence of minerals such as eudialyte, monazite, steenstrupine and catapleiite thus yields information about the physico-chemical conditions of consolidation of agpaitic nepheline syenite.

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