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*Article*

# Mineralogical and Geochemical Constraints on the Origin and Late-Stage Crystallization History of the Breivikbotn Silicocarbonatite, Seiland Igneous Province in Northern Norway: Prerequisites for Zeolite Deposits in Carbonatite Complexes

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**Abstract:** The present work reports new mineralogical and whole rock geochemical data from the Breivikbotn silicocarbonatite (Seiland igneous province, North Norway), allowing conclusions to be drawn concerning its origin and the role of late fluid alteration. The rock shows a rare mineral association: calcite + pyroxene + amphibole + zeolite group minerals + garnet + titanite, with apatite, allanite, magnetite and zircon as minor and accessory minerals, and it is classified as silicocarbonatite. Calcite, titanite and pyroxene ($Di_{36-46}$ $Acm_{22-37}$ $Hd_{14-21}$) are primarily magmatic minerals. Amphibole of hastingsitic composition has formed after pyroxene at a late-magmatic stage. Zeolite group minerals (natrolite, gonnardite, Sr-rich thomsonite-(Ca)) were formed during hydrothermal alteration of primary nepheline by fluids/solutions with high Si-Al-Ca activities. Poikilitic garnet (Ti-bearing andradite) has inclusions of all primary minerals, amphibole and zeolites, and presumably crystallized metasomatically during a late metamorphic event (Caledonian orogeny). Whole rock chemical compositions of the silicocarbonatite differs from the global average of calciocarbonatites by elevated silica, aluminium, sodium and iron, but show comparable contents of trace elements (REE, Sr, Ba). Trace element distributions indicate within-plate tectonic setting of the carbonatite. The spatial proximity of carbonatite and alkaline ultramafic rock (melteigite), the presence of “primary nepheline” in carbonatite together with the trace element distributions indicate that the carbonatite was derived from crystal fractionation of a parental carbonated foidite magma. The main prerequisites for the extensive formation of zeolite group minerals in silicocarbonatite are revealed.

**Keywords:** silicocarbonatite; melteigite; calcite; nepheline; zeolite group minerals; garnet; crystal fractionation; Breivikbotn; Northern Norway.

## 1. Introduction

Alkaline rocks and carbonatites represent less than 1% of all igneous rocks of the Earth’s crust. However, the petrogenesis of these rocks is particularly interesting, in part due to their great variability and in part because they are economically important, containing most of the global reserves of, for example, the rare earth elements (REE), zirconium, niobium and phosphorus (apatite). Moreover, alkaline igneous rocks and carbonatites have great petrological and geodynamical significances. Most common point of view for their generation is low-volume partial melting of mantle domains enriched in trace elements and volatiles. Owing to their deep source and

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rapid ascent to the crust the rocks of this clan bear information on the composition of the deep Earth. Combining observations on the geochemical characteristics of natural rocks with experimental results may provide insight into processes of mantle metasomatism and melt generation under high-pressure conditions. Metasomatism by carbonatite melts has been recognized as an important mechanism for enrichment of mantle domains.

Although it has long been recognized that alkaline rocks occur in various tectonic settings worldwide, study of these rocks, particularly carbonatites, has focused mainly on within-plate continental rift environments. Due to the typically low volume and high reactivity of these magmas, it may nonetheless be assumed that an extensional tectonic environment is prerequisite to their emplacement into the upper crust. In post-collisional extensional settings, alkaline rocks and carbonatites have the potential to provide information about the effects of convergent tectonic processes on the geochemical evolution of the upper mantle. Although the majority of known carbonatites are found in rift or near-rift settings, they may rarely occur in off-craton settings where extension may be localized in back-arc regimes or occur from widespread orogenic collapse [1]. Nevertheless, recently the carbonatites in pure collisional and subduction tectonic settings were reported from several localities worldwide [2-6]. Enrichment in high field strength elements (HFSE) such as niobium and zirconium, once considered an essential characteristic of carbonatite, is conspicuously absent from carbonatites in collisional and subduction tectonic settings. Furthermore, carbonatite melts are highly reactive and cause alkaline metasomatic alteration (fenitization) of the surrounding rocks [7].

In this paper new geochemical and mineralogical data on the Breivikbotn carbonatite occurrence are presented and its possible origin and late-, post-crystallization processes are proposed. The occurrence is remarkable for the high zeolite content, and the main prerequisites for zeolite formation in carbonatite complexes are substantiated.

## 2. Geological Setting

### *2.1. Seiland Igneous Province*

The Seiland Igneous Province (SIP) consists of contemporaneous mafic, ultramafic, intermediate, granitic and alkaline intrusions emplaced into a 50 km by 100 km area, 570-560 Ma ago. These intrusions are constrained to a single nappe within the Kalak Nappe complex of Northern Norway. This nappe complex has been generally assumed to be a parautochthonous terrane within the 420 Ma Norwegian Caledonides [8], but more recent work has indicated that the terrane may be exotic and allochthonous [9]. The largest component of the SIP consists of numerous mafic plutons, commonly layered, which comprise at least 50-60% of the province. Large ultramafic complexes comprise a further 25-35% of the complex, while intermediate rock types such as monzonite and diorite make up 10%. Alkaline intrusions occur throughout the province, covering about 5% of the area. Granitic rocks are restricted to a few small, insignificant bodies on Øksfjord and Sørøy.

Sturt et al [10] and Ramsay & Sturt [11] suggested that the magmatic activity in the SIP was synorogenic and related to the "Finnmarkian Orogeny", an early phase of the Caledonian Orogeny. This was based on field observations suggesting that the foliated igneous rocks were cut by younger intrusions that in turn were overprinted by a later metamorphic fabric. According to Sturt & Ramsay [12] and Sturt et al [10], the late stage alkaline rocks in the province were both intruded and deformed during the youngest phase of the Finnmarkian deformation in the Sørøy Nappe. Krill and Zwaan [13] re-evaluated the field relationships in Sørøy and suggested that the igneous rocks of SIP were preorogenic, rather than synorogenic. They proposed that the magmatic activity was related to crustal attenuation and rift formation of the margin of Fennoscandia. Based on new U-Pb zircon ages from SIP, Roberts et al [14] proposed an extensional setting for SIP and interpreted the younger ages (460-420 Ma) as evidence for imposed metamorphism during the main stages of the Caledonian orogeny.

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*2.2. Breivikbotn occurrence*

This complex consists of carbonatite, malignite (named shonkinite by [12]), nepheline syenite, aplitic syenite and pyroxenite. It occurs as a deformed, 2 km long and 500 m wide sill hosted within the Klubben Psammite, a metasedimentary unit of the Kalak Nappe Complex (Jonassen, 1996). At the northern side of the bay at Haraldseng, the carbonatite has intruded one limb of a north–south fold, and stands out clearly in contrast to the layered sediments hosting it. Single layers of the intrusion run north–south, with the eastern edge marked by a thin (<10 m) aegirine–augite pyroxenite, which appears to lie conformably on the steeply dipping psammites. Some shearing has taken place along this contact, but there is nothing to contradict the conclusion that this is the bottom contact of the intrusion [12]. The pyroxenite, like the rest of the complex, is intruded by numerous centimeter thick nepheline syenite and dolerite dykes, and is extremely variable in appearance.

Overlying the pyroxenite, across a thin band of carbonatitic breccia, there is a coarse-grained malignite, dominated by feldspar, but also containing pyroxene (aegirine–augite) and amphibole. Some localities are rich in melanitic garnet [15]. Malignite from the Breivikbotn complex can be divided in two varieties based on the dominating mafic mineral. The most common type is rich in melanite. The other type is rich in clinopyroxene. Both types occur within the central parts of the Breivikbotn complex. Also present are zeolites (pseudomorphosed after nepheline; Jonassen 1996), calcite, titanite and zircon. Melanite (up 30-70 modal %), pyroxene (40-60 modal %), feldspar (10 – 40 modal %, in parts is absent) and "nepheline" are obviously magmatic minerals. For less deformed malignite, melanite shows oscillatory zoned crystals. The zoning is characterized by an alternation of light-brown and dark-brow zones, probably reflecting variations in the Ti-content of garnet (overall, the $TiO_2$ content in garnet varies from 2.1 to 4.1 wt % [15]). Commonly, melanite shows dark central parts and lighter marginal parts. The crystals are euhedral, and might be overgrown by later garnet. Characteristic for melanite is the presence of inclusions of pseudomorphs of zeolite aggregates. Furthermore, melanite has been overgrown by clinopyroxene, which in turn has been overgrown by amphibole. Clinopyroxene occurs as prismatic, subhedrel to euhedral crystals. The size of the crystal varies; they can be up to 3 cm long. They show inclusions of an opaque mineral. In addition, grains of titanite and calcite are observed. Pyroxene is often overgrown by amphibole and light yellow garnet. In places, clinopyroxene has both inclusions, and a rim of amphibole. Pyroxene is occasionally observed as inclusions in melanite. In one case, clinopyroxene is partly overgrown by "nepheline". Based on its optical properties, in addition to mineral chemical analyses [15], it is assumed that the mineral is aegirine-augite. From petrography (particularly, the presence of garnet and disproportion between nepheline and alkali feldspar) this rock does not fit well to *s.s.* malignite and below the term "malignite" is used.

The "malignite" has a banded appearance, caused by changes in grain-size and mineralogy. The underlying carbonatitic breccia comprises a network of thin carbonatite veins enclosing numerous large angular fragments of both "malignite" and pyroxenite, and generally sheared along the edges.

The "malignite" can be observed intruding alkali pyroxenite. Alkali pyroxenite can also be observed as inclusions in the «malignite». The "malignite" has been carbonatized, and it is cross-cut by 4-5 cm thick carbonatite dykes. "Malignite" also occurs as xenoliths in the carbonatite. Furthermore, the rock has been intruded by nepheline syenite, pegmatites and aplitic syenites. The «malignite» grades into carbonatite, with no clear boundary between the two rock types. The carbonatite is very variable in both texture and composition, and generally occurs as sheets. In some layers, particularly at the top of the intrusion, the carbonatite contains many fragments of country rock, and is referred to as xenolithic carbonatite [12]. At the top of the intrusion the carbonatite sharply truncates the sedimentary banding in the psammites, but there is a ubiquitous aureole of metasomatic alteration [12, 15].

Nepheline syenite occurs mainly as thin carbonated dykes but seems to develop into a more extensive unit further north [15]. It commonly contains biotite, or locally pyroxene. The nepheline

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syenite is often gneissic and has extremely variable nepheline and carbonate contents that allow interpreting it as a product of alkaline metasomatism (fenitization) of aplitic syenite.

The carbonatite is intruded by numerous dolerite dykes, somewhere folded and boudinaged.

## 3. Analytical Methods

### *3.1. Mineral Composition Analyses and X-Ray Identification*

The chemical compositions of minerals from the carbonatite were carried out at the Geological Institute, Kola Science Center, by means of an electron microprobe Cameca MS-46 (WDS mode, 22 kV, 30-40 nA, with 50 sec counting time). The following calibrating materials (and analytical lines) were used: wollastonite (Si*Kα*, Ca*Kα*), hematite (Fe*Kα*), apatite (P*Kα*), lorenzenite (Na*Kα*), thorite (Th*Mα*), $MnCO_3$ (Mn*Kα*), $Y_3Al_5O_{12}$ (Y*Lα*), (La,Ce)S (La*Lα*), CeS (Ce*Lα*), $Pr_3Al_5O_{12}$ (Pr$L\beta_1$), $LiNd(MoO_4)_2$ (Nd*Lα*), $SmFeO_3$ (Sm*Lα*), $EuFeO_3$ (Eu*Lα*), GdS (Gd*Lα*), $TbPO_4$ (Tb*Lα*), $Dy_3Al_5O_{12}$ (Dy*Lα*), $Ho_3Ga_5O_{12}$ (Ho$L\beta_1$), $ErPO_4$ (Er*Lα*), $Tm_3Al_5O_{12}$ (Tm*Lα*), $Yb_3Al_5O_{12}$ (Yb*Lα*), and $Y_{2.8}Lu_{0.2}Al_5O_{12}$ (Lu*Lα*). Detection limits for Fe, Mn are 0.01%; Si, Al, Cl, Ca, K, Cl – 0.02%; P, Na, Y, Sr, La, Ce, Nd – 0.03%; Ba – 0.05%; Nb, Zr – 0.1%. Representative electron microprobe data for minerals are given in Tables 1, 2, 3 and 4.

The accessory mineral identification was performed using a LEO-1450 SEM (scanning electron microscope) equipped with XFlash-5010 Bruker Nano GmbH EDS (energy-dispersive Xray spectroscopy). The system was operated at 20 kV acceleration voltage, 0.5 nA beam current, with 200 s accumulation time.

Materials from small areas of zeolite group minerals close to points analyzed by microprobe were examined by the X-ray powder diffraction (XRPD) method (Debye-Scherer) by means of an URS-1 operated at 40 kV and 16 mA with RKU-114.7 mm camera and Fe*Kα*-radiation.

### *3.2. Whole Rock Analyses*

Whole rock composition data for carbonatite-like rock were obtained at the Kola Science Center in Apatity, Russia. Most of the major elements were determined by atomic absorption spectrophotometry; $TiO_2$ by colorimetry; $K_2O$ and $Na_2O$ by flame photometry; FeO and $CO_2$ by titration (volumetric analysis); and F and Cl by potentiometry using an ion-selective electrode (for a description of the methods, see [16]). Trace elements were determined by Inductively Coupled Plasma-Mass Spectrometry.

The additional whole rock composition data for the carbonatite-like and alkaline rocks from the occurrence were obtained at the Department of Biology and Geology, University of Tromsø. Two parallels of each sample were analyzed for major, minor and trace elements by X-ray fluoresence (XRF) on a Philips PW 1400 instrument. For major and minor elements analyses, fused pellets containing a mixture of rock powder and lithium tetraborate flux were used (mixed in ratio of 1:6). Trace element analyses were carried out on pressed powder pellets. The calibration of the analytical instrument was checked against the international standards GH, GM and NIM-S [17].

Major and minor element compositions of selected samples are presented in Table 5, and trace elements in Table 6. The whole dataset is presented in the Supplementary Data Table S1.

## 4. Petrography and Mineral Compositions

The Breivikbotn carbonatite is a massive rock of porphyritic texture with hypidiomorphic, lesser idiomorphic phenocrysts of garnet and pyroxene. The groundmass is medium and coarse grained and composed of carbonate, pyroxene, amphibole and zeolite. The textural relationships of the studied samples are shown on Figure 1. The mineral content is variable and consists of carbonate (20-50 vol. %), amphibole (5-20 vol. %), pyroxene (5-20 vol. %), zeolite group minerals (0-20 vol. %), garnet (0-30 vol. %). Minor and accessory minerals are apatite (1-3 vol. %), titanite (1-5 vol. %), allanite, magnetite, zircon, pyrite, pyrrhotite, chalcopyrite, scheelite, celestine, barite, and baddeleyite.

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In several thin sections, single grains of quartz were observed. Its interstitial character points on deuteric nature.

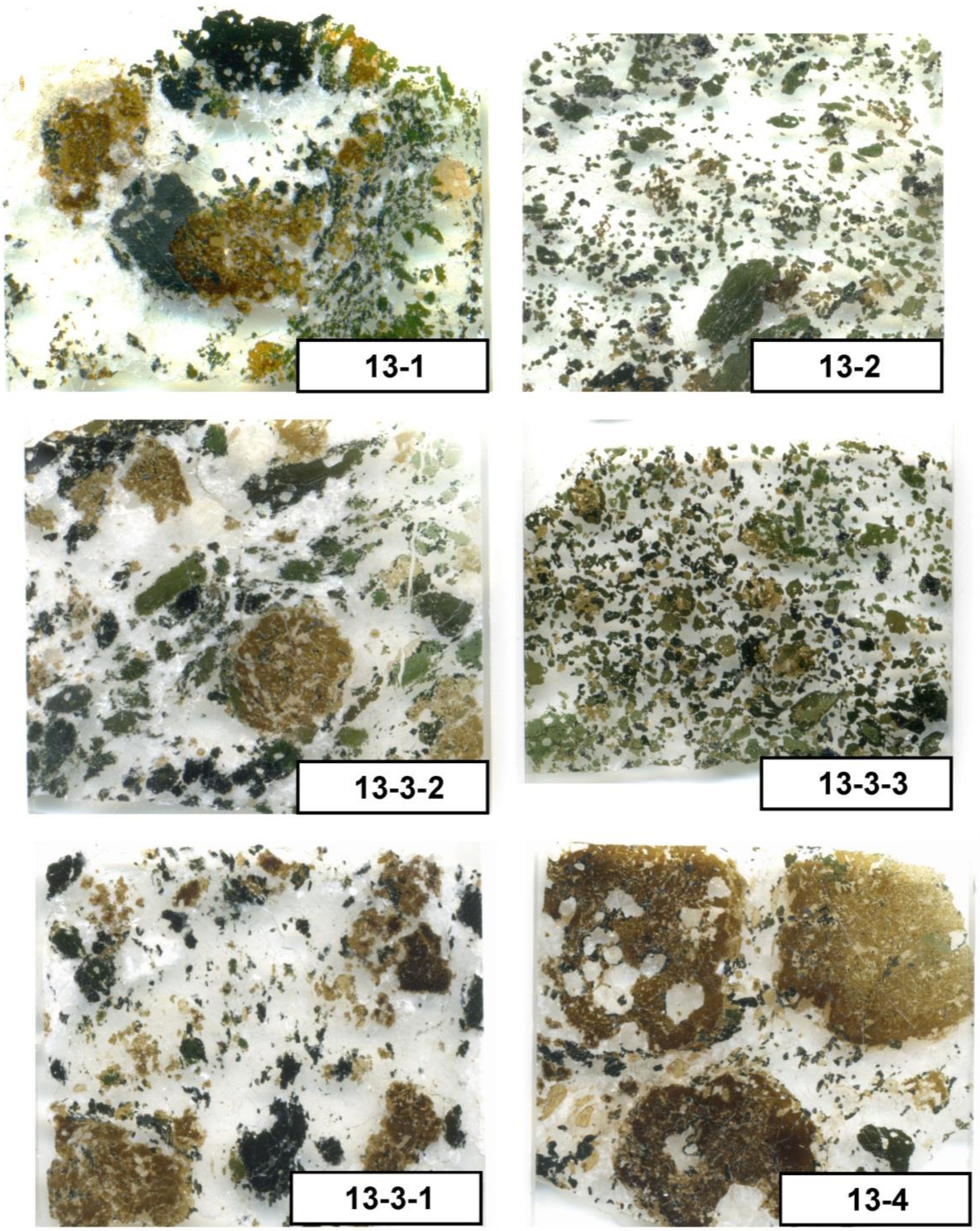


**Figure 1.** Scans of the selected samples of the Breivikbotn carbonatite, showing extremely heterogeneous textures. Carbonate and zeolite group minerals (white and transparent) form the groundmass; garnet (dark brown) forms rounded poikilitic grains; titanite (yellow) forms small angular and elongated euhedral crystals, pyroxene (greenish) forms subhedral and euhedral crystals, amphibole (bluish, dark-green, and indigo) forms anhedral grains and large laths. Width of all of the scans is about 2.5 cm.

***Carbonate*** is clearly a primary mineral as it forms euhedral and subhedral crystals of 0.15 – 5 mm size with triple junctions between grains (Fig. 2c). The carbonate is calcite (average formulae $((Ca_{0.953}\ Sr_{0.014}\ Mn_{0.003}\ Fe_{0.002}\ Mg_{0.001})_{0.973}\ CO_3)$ with negligible contents of Mg, Fe, and Mn (Table 1).

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Calcite contains elevated SrO (up to 2.15 wt.%) that is characteristic for magmatic calcite from carbonatites.

***Pyroxene*** occurs as subhedral grains of 0.3-4 mm, rarely as phenocrysts up to 1 cm size. Pyroxene rims are often resorbed, and amphibole growth along the rims is observed. Pyroxene shows inclusions of calcite grains. Representative compositions of pyroxene are given in Table 2. Compositionally, pyroxene show high content of the diopside component, with essential quantities of acmite and hedenbergite ($Di_{36-46}$ $Acm_{22-37}$ $Hd_{14-21}$). The average formula is $(Ca_{0.75}Na_{0,27})_{1,02}(Mg_{0.42}Fe^{3+}_{0.27}Fe^{2+}_{0.20}Al_{0.07}Ti_{0.02})_{0.98}[Si_{1.9}Al_{0.1}O_6]$. Pyroxene always shows minor content of $TiO_2$ (up to 1 wt.%).

***Amphibole*** occurs as anhedral and subhedral grains of 0.3-5 mm size. The mineral occurs as individual grains and as overgrowths on pyroxene (Fig. 2b). Rarely, amphibole forms up to 1 cm poikilitic grains with inclusions of calcite and pyroxene. Amphibole is hastingsitic with an essential proportion of magnesiohastingsite. The average formula is $(Na_{0.88}\ K_{0.43})_{1.31}\ (Ca_{1.68}\ Na_{0.14}\ Fe^{2+}_{0.18})_2\ (Ti_{0.15}\ Fe^{2+}_{2.73}\ Mg_{1.56}\ Al_{0.43}\ Mn_{0.12})_{4.99}\ [Si_{6.1}\ Al_{1.9}\ O_{23}]$. The mineral shows elevated contents of $K_2O$ (2-2.3 wt.%) and $TiO_2$ (1.2-1.4 wt.%).

***Garnet*** usually occurs as porphyritic subhedral rounded grains of 0.5-1 cm size. The mineral has poikilitic texture and contains inclusions of calcite, pyroxene, amphibole, titanite, zeolite group minerals (Figures 2a, 2d, 2f, 4). Rims of garnet overgrowing amphibole can be observed (Fig. 2c). Garnet is patchily zoned; in BSE images, garnet is generally bright along the rims, with darker central parts, however, patches of bright garnet are also observed within the darker central parts (Fig. 2a). The bright patches appear to reflect elevated Fe content. Garnet texture and morphology suggest porphyroblastic growth of the mineral. Representative chemical compositions of garnet are given in Table 3. Garnet can be classified as Ti-bearing andradite with the average formula $(Ca_{2.90}Na_{0.01}Y_{0.01})_{2.92}(Fe^{3+}_{1.50}Al_{0.31}Ti_{0.15}Mn_{0.10}Mg_{0.03}Zr_{0.02})_{2.11}[Si_{0.91}Al_{0.09}O_{12}]$. The mineral contains V, Zr and Y as minor constituents. The content of $TiO_2$ varies in the range 1.3-3.2 wt.%, which is low compared to titaniferous garnets from carbonatites and alkaline rocks (>5 wt.%, according to [18-22], and even lower than for melanite from the «malignite» from the Breivikbotn occurrence.

***Zeolite group minerals and "altered nepheline".*** Clusters of zeolite group minerals (ZGM) have a stubby rectangular or equant rounded (roughly hexagonal) form, up to 2-3 mm in diameter (Fig. 2e, 3, 4). Most clusters are composed of natrolite and gonnardite; natrolite often occurs in the central parts of gonnardite aggregates, and it is inferred that natrolite is the earliest phase (Fig. 3e, f).

Natrolite forms colorless, white, smooth anhedral grains of 1-2 mm size. The average chemical composition of natrolite is $Na_{1.98}Ca_{0.03}Si_{3.01}Al_{1.98}O_{10}\cdot 2H_2O$ (Table 4), which is very close to the stoichiometric formula $(Na_2(Si_3Al_2)O_{10}\cdot 2H_2O$, IMA-list 09-2017). The Si/(Si+Al) ratio varies from 0.57 to 0.65, while sodium is in the range 1.81-2.22 apfu and calcium does not exceed 0.1 apfu.

Gonnardite occurs as colorless, uneven, cracky aggregates up to 1-3 mm size, the individual grains are anhedral and 100-500 μm in diameter. The average composition of gonnardite is calculated as $(Na_{1.57}Ca_{0.38})_{2.05}(Si_{2.67}Al_{2.38})_{5.05}O_{10}\cdot 3H_2O$, which is close the stoichiometric formula $((Na,Ca)_2(Si,Al)_5O_{10}\cdot 3H_2O$, IMA-list 09-2017). The Si/(Si+Al) ratio varies from 0.52 to 0.54, while Na/(Na+Ca) from 0.76 to 0.85.

Thomsonite-(Ca) forms colorless and white rectangular grains (Fig. 3d). It is irregularly zoned, and in BSE images characterized by brighter and darker zones. The mineral appears as partly fibrous. The average composition is $Na_{1.13}Ca_{1.7}(Al_{4.98}Si_{5.1})O_{20}\cdot 6H_2O$, which is close to ideal formula $(NaCa_2(Al_5Si_5)O_{20}\cdot 6H_2O$, IMA-list 09-2017). The mineral is characterized by elevated Sr content (0.02 – 0.34 apfu, with average 0.09 apfu). The Sr content may vary within a single crystal as indicated by the brighter and darker zones in BSE images. The Si/(Si+Al) ratio varies from 0.49 to 0.52, while Na/(Na+Ca+Sr) varies from 0.34 to 0.42.

Overall, the ZGM of Breivikbotn carbonatite shows successively increasing Ca and Al content from natrolite, via gonnardite to thomsonite-(Ca).

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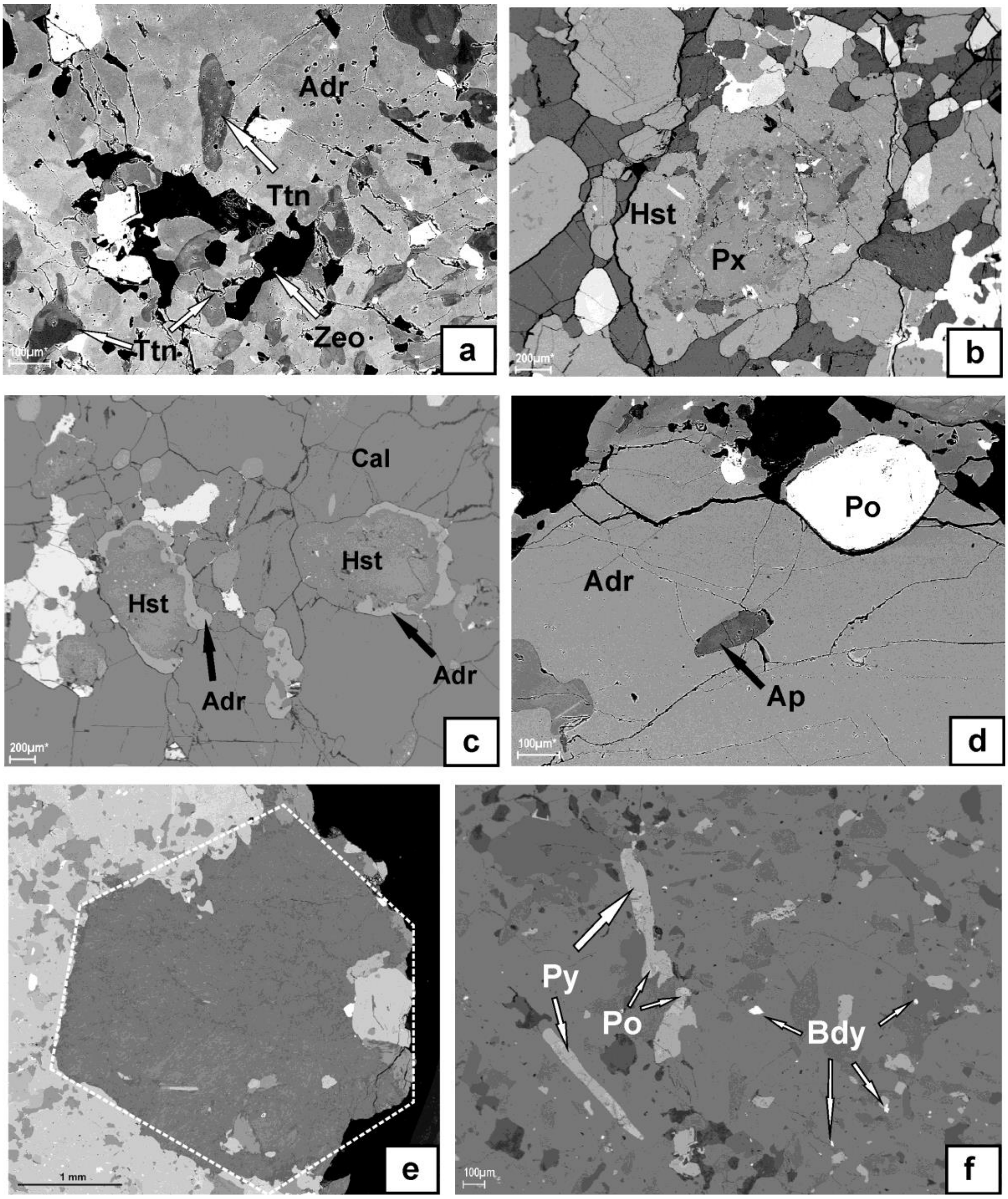


**Figure 2.** Back-scattered-electron (BSE) images showing the relationships between different minerals in the Breivikbotn carbonatite: **(a)** titanite, zeolite and magnetite (brightest) included in andradite; **(b)** hastingsite rimming pyroxene; **(c)** andradite rim around hastingsite, calcite grains show typical triple junctions; **(d)** apatite and pyrrhotite included in andradite; **(e)** roughly hexagonal habit of zeolite aggregate; **(f)** inclusions of accessory pyrite, pyrrhotite and baddeleyite in andradite. Mineral abbreviations are from [23].

In some natrolite-gonnardite clusters, water-absent Na-Al silicates with chemical compositions close to nepheline were found (Table 4). These compositions in combination with the textural appearance of the natrolite-gonnardite aggregates suggest that the aggregates are pseudomorphs after nepheline. Thomsonite-(Ca) can also be inferred as an alteration product of nepheline. The water-absent nepheline-like mineral is characterized by compositions corresponding to $Na_{0.53-0.7}Ca_{0.01-0.16}Al_{1.07-1.24}Si_{1.06-1.22}O_4$, which is somewhat different from the stoichiometric formula of nepheline, with lower Na and higher Ca. We suggest that the mineral initially crystallized as

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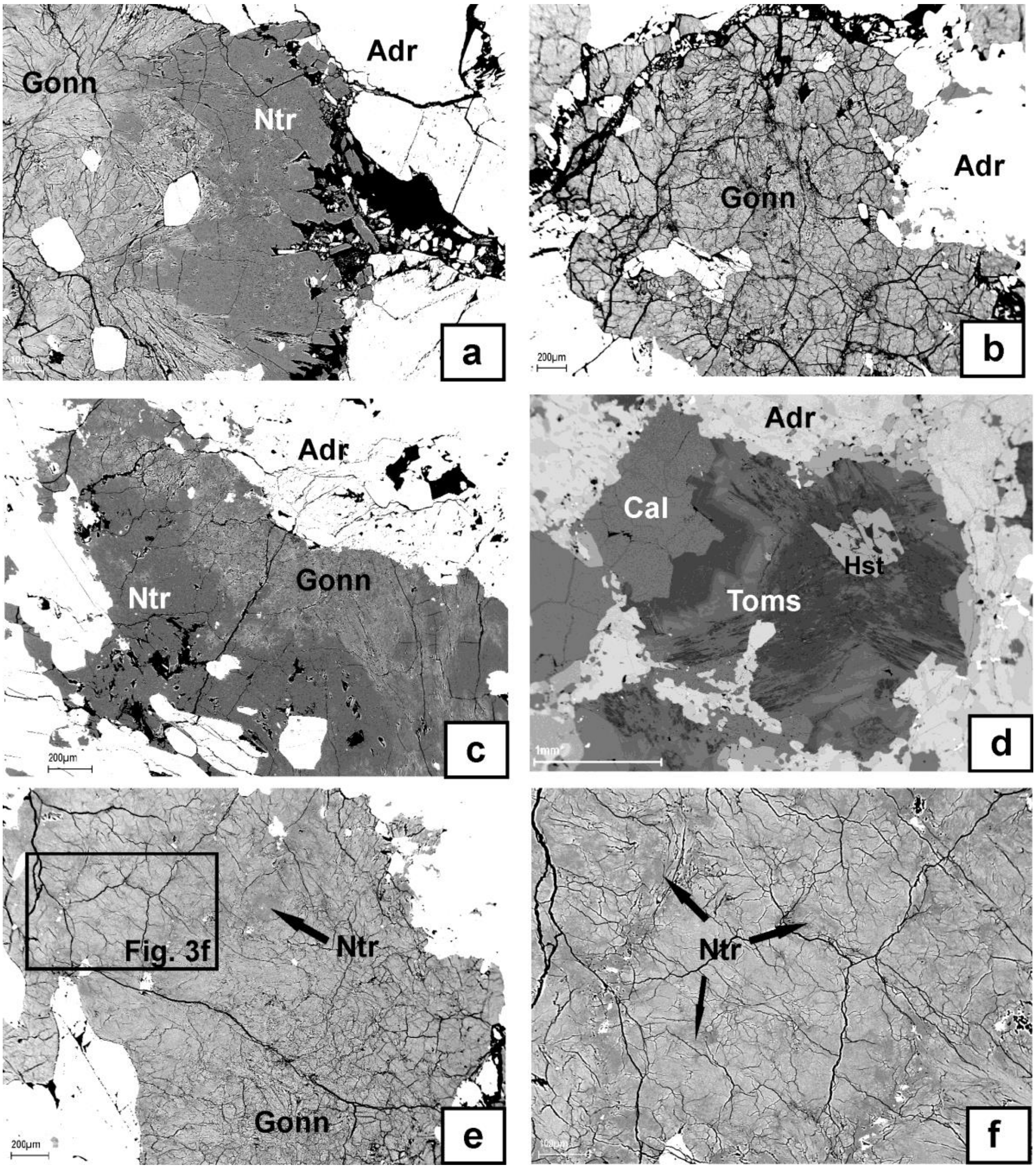


**Figure 3.** BSE images showing the morphology and internal textures of zeolite group minerals: **(a, b, c)** intergrowths and aggregates of natrolite and gonnardite; **(d)** zonal structure of thomsonite-(Ca) with low (dark gray) and high (gray) Sr content; **(e, f)** "shadow"-type domains of natrolite (dark-gray) in gonnardite (light-gray), illustrating the early crystallization of natrolite relative to gonnardite (XRPD of the sample indicated a mix of both minerals). Mineral abbreviations are from [23]. Toms – thomsonite-(Ca), Gonn – gonnardite.

nepheline from a carbonatite magma, and subsequently underwent alteration in a high-Ca environment. Ca-bearing and Ca-rich nephelines have been found in alkaline rocks from the Messum complex, Namibia [24], from the Marangudzi Complex, Zimbabwe [25], and from the Allende meteorite [26, 27].

***Titanite*** occurs as euhedral and subhedral elongated grains. It is often associated with garnet, i.e. included in garnet and occurring adjacent to garnet. The average chemical composition of titanite is $(Ca_{0.98}Y_{0.1}Ce_{0.1})$ $(Ti_{0.94}Fe_{0.05}Zr_{0.01}Nb_{0.01})$ $(Si_{0.98}Al_{0.04}O_5)$. Titanite shows elevated contents of REE, Zr, Nb, Fe and Al. Incorporation of iron and aluminium in the titanite structure

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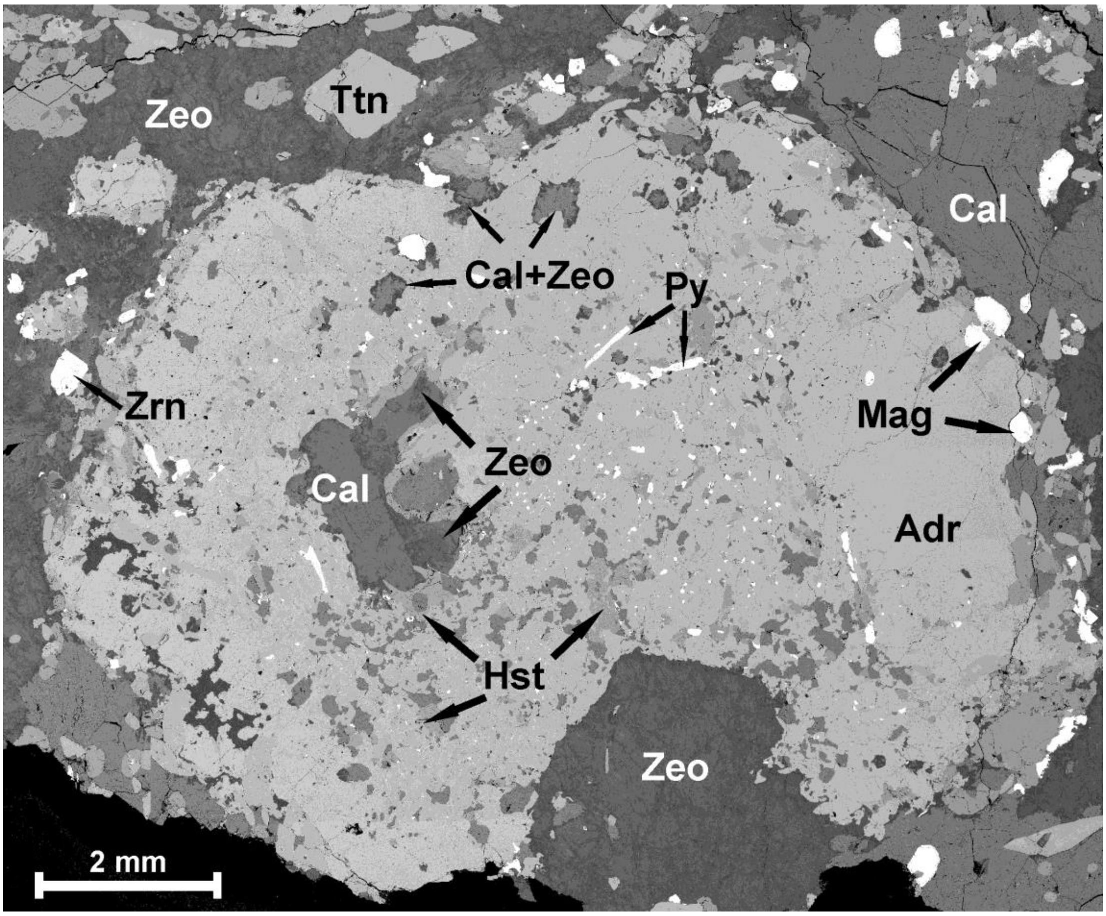


**Figure 4.** BSE image of typical poikilitic andradite with inclusions of calcite, hastingsite, zeolites, magnetite and pyrite. Zeolites often form intergrowths and aggregates of rectangular shape (bottom). A large number of tiny grains of baddeleyite (bright grains, ≤50 μm) occur in the central part of the andradite grain. Mineral abbreviations are from [23]. The image is a mosaic of 160 small 1.2x0.8 mm BSE images

**Table 1.** Representative chemical compositions and mineral formulae of carbonate from the Breivikbotn carbonatite

| **Sample №** | **13-4-1-5-1** | **13-4-1-5-2** | **13-2-1a** |
|---|---|---|---|
| wt. % | | | |
| FeO | 0.16 | 0.00 | 0.16 |
| MnO | 0.25 | 0.22 | 0.27 |
| MgO | 0.07 | 0.00 | |
| CaO | 53.65 | 55.29 | 51.32 |
| SrO | 2.07 | 0.11 | 2.15 |
| Formulae on the basis of 2 cations | | | |
| Fe | 0.002 | | 0.002 |
| Mn | 0.004 | 0.003 | 0.004 |
| Mg | 0.002 | | |
| Ca | 0.957 | 0.986 | 0.972 |
| Sr | 0.020 | 0.001 | 0.022 |

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requires the coupled substitutions: $Ti^{4+} + O^{2-} = (A1,Fe^{3+}) + (F,OH)^{-}$ and takes place at high-P metamorphic conditions [28].

***Celestine*** is the only accessory mineral that was analyzed ($SO_3$ = 43.42 wt%; BaO = 0.35 wt%; CaO = 0.17 wt%; SrO = 56.45 wt%), and its occurrence together with barite and Fe-Cu-sulphides indicate high S fugacity of the system during crystallization of the carbonatite.

***Baddeleyite*** occurs as tiny angular grains of 10-60 μm size as numerous inclusions in garnet (Fig. 2f). Baddeleyite is a characteristic mineral of carbonatite-ultramafic intrusions worldwide, indicative the Si-undersaturated environment during formation of such rocks [29].

Table 2. Chemical analyses of pyroxene and amphibole from Breivikbotn carbonatite.

| **Mineral** | **Pyroxene** | | | | | **Amphibole** | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| **Sample** | **13-4** | **13-2** | **13-3a** | **13-3** | **13-3a** | **13-4** | **13-4** | **13-2** | **13-3** | **13-3** |
| | **wt. %** | | | | | | | | | |
| $SiO_2$ | 49.74 | 50.99 | 48.47 | 51.16 | 51.91 | 37.90 | 37.46 | 39.33 | 39.14 | 39.28 |
| $Al_2O_3$ | 3.18 | 3.44 | 4.97 | 3.70 | 3.26 | 12.32 | 12.95 | 11.72 | 12.98 | 12.76 |
| $TiO_2$ | 0.49 | 0.54 | 1.02 | 0.45 | 0.34 | 1.37 | 1.31 | 1.16 | 1.12 | 1.25 |
| $Fe_2O_3$ | | | | | | | | | | |
| FeO | 17.75 | 14.16 | 14.16 | 13.45 | 12.62 | 25.61 | 24.30 | 21.18 | 19.89 | 19.52 |
| MnO | 0.85 | 0.69 | 0.92 | 0.60 | 0.60 | 1.02 | 0.99 | 0.89 | 1.01 | 0.92 |
| MgO | 6.47 | 7.59 | 7.06 | 7.65 | 8.15 | 5.26 | 4.98 | 7.62 | 7.64 | 7.72 |
| CaO | 17.33 | 18.59 | 20.59 | 17.86 | 18.91 | 9.04 | 9.86 | 9.85 | 10.47 | 10.36 |
| $Na_2O$ | 4.17 | 3.59 | 2.66 | 4.33 | 3.73 | 3.75 | 3.50 | 2.85 | 3.17 | 3.32 |
| $K_2O$ | | | | | | 2.21 | 2.24 | 1.96 | 2.28 | 2.06 |
| ZnO | | | | | | 0.09 | 0.10 | 0.07 | 0.09 | 0.06 |
| Total | 99.98 | 99.59 | 99.85 | 99.20 | 99.51 | 98.57 | 97.66 | 96.62 | 97.80 | 97.25 |
| | apfu (4 cations) | | | | | apfu (23 cations) | | | | |
| Si | 1.882 | 1.923 | 1.838 | 1.921 | 1.946 | 6.031 | 5.993 | 6.215 | 6.100 | 6.135 |
| Al(iv) | 0.118 | 0.077 | 0.162 | 0.079 | 0.054 | 1.969 | 2.007 | 1.785 | 1.900 | 1.865 |
| Al(vi) | 0.024 | 0.076 | 0.060 | 0.084 | 0.090 | 0.342 | 0.434 | 0.398 | 0.485 | 0.484 |
| Al(tot) | 0.142 | 0.153 | 0.222 | 0.164 | 0.144 | 2.311 | 2.442 | 2.183 | 2.385 | 2.349 |
| Ti | 0.014 | 0.015 | 0.029 | 0.013 | 0.010 | 0.164 | 0.158 | 0.138 | 0.132 | 0.146 |
| Fe*** | 0.373 | 0.233 | 0.238 | 0.285 | 0.216 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 |
| Fe** | 0.189 | 0.213 | 0.211 | 0.137 | 0.180 | 3.408 | 3.251 | 2.800 | 2.592 | 2.550 |
| Mn | 0.027 | 0.022 | 0.030 | 0.019 | 0.019 | 0.138 | 0.134 | 0.119 | 0.133 | 0.122 |
| Mg | 0.365 | 0.427 | 0.399 | 0.428 | 0.455 | 1.247 | 1.187 | 1.794 | 1.774 | 1.797 |
| Ca | 0.703 | 0.751 | 0.837 | 0.718 | 0.759 | 1.541 | 1.689 | 1.667 | 1.749 | 1.733 |
| Na | 0.306 | 0.262 | 0.196 | 0.315 | 0.271 | 1.158 | 1.084 | 0.874 | 0.959 | 1.005 |
| K | | | | | | 0.448 | 0.456 | 0.394 | 0.453 | 0.411 |

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Table 3. Chemical analyses of garnet and titanite from Breivikbotn carbonatite.

| **Mineral** | **Garnet** | | | | | | | | | **Titanite** | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **Sample** | **13-4 C** | **13-4 R** | **13-4** | **13-4** | **13-4 C** | **13-4 R** | **13-4 C** | **13-4 R** | **13-1** | **13-4** | **13-4** | **13-1** |
| | **wt. %** | | | | | | | | | | | |
| $SiO_2$ | 34.43 | 34.68 | 33.98 | 35.06 | 34.87 | 34.63 | 35.68 | 34.83 | 36.44 | 28.86 | 29.02 | 30.17 |
| $Al_2O_3$ | 4.65 | 3.39 | 3.65 | 3.56 | 4.43 | 3.52 | 5.62 | 3.51 | 3.19 | 1.11 | 1.19 | 1.11 |
| $TiO_2$ | 3.19 | 2.88 | 2.58 | 1.53 | 2.95 | 2.06 | 1.25 | 2.63 | 2.03 | 37.23 | 38.28 | 37.19 |
| $Fe_2O_3$ | 22.17 | 24.22 | 24.73 | 25.03 | 22.51 | 24.73 | 22.95 | 24.01 | 23.74 | | | |
| FeO | | | | | | | | | | 1.88 | 1.58 | 1.68 |
| MnO | 1.28 | 1.41 | 1.48 | 1.20 | 1.10 | 1.25 | 1.70 | 1.35 | 1.25 | 0.08 | 0.06 | 0.05 |
| MgO | 0.34 | 0.21 | 0.17 | 0.15 | 0.27 | 0.18 | 0.14 | 0.29 | 0.15 | 0.00 | 0.00 | 0.00 |
| CaO | 32.67 | 32.87 | 31.64 | 32.76 | 32.70 | 32.19 | 32.14 | 31.63 | 32.81 | 26.79 | 27.82 | 27.92 |
| $Na_2O$ | 0.16 | | 0.22 | | | 0.11 | | | | 0.00 | 0.00 | 0.00 |
| $K_2O$ | | | | | | | | | | 0.00 | 0.00 | 0.00 |
| ZnO | | | | | | | | | | 0.07 | 0.00 | |
| $Y_2O_3$ | | 0.17 | 0.17 | | | | 0.18 | 0.15 | | 0.91 | 0.00 | |
| $ZrO_2$ | 0.64 | 0.29 | 0.31 | 0.35 | 0.78 | 0.42 | 0.14 | 0.76 | 0.21 | 0.69 | 0.65 | 0.53 |
| $Yb_2O_3$ | | 0.04 | | | | | | | | | | |
| $V_2O_5$ | 0.06 | 0.06 | 0.06 | 0.06 | | 0.06 | 0.04 | 0.04 | 0.10 | 0.00 | 0.00 | 0.17 |
| $Nb_2O_5$ | | | | | | | | | | 0.37 | 0.10 | 0.63 |
| $La_2O_3$ | | | | | | | | | | 0.12 | 0.06 | 0.26 |
| $Ce_2O_3$ | | | | | | | | | | 0.30 | 0.26 | 0.71 |
| $Nd_2O_3$ | | | | | | | | | | 0.45 | 0.33 | |
| $Sm_2O_3$ | | | | | | | | | | 0.24 | 0.12 | |
| $Gd_2O_3$ | | | | | | | | | | 0.36 | 0.00 | |
| Total | 99.59 | 100.23 | 98.97 | 99.70 | 99.61 | 99.16 | 99.83 | 99.19 | 99.92 | 99.45 | 99.45 | 100.40 |
| | apfu (8 cations) | | | | | | | | | apfu (3 cations) | | |
| Si | 2.872 | 2.896 | 2.872 | 2.935 | 2.918 | 2.916 | 2.955 | 2.950 | 3.036 | 0.971 | 0.967 | 0.996 |
| Al(iv) | 0.128 | 0.104 | 0.128 | 0.065 | 0.082 | 0.084 | 0.045 | 0.050 | 0.000 | | | |
| Al(vi) | 0.330 | 0.230 | 0.235 | 0.286 | 0.354 | 0.266 | 0.503 | 0.300 | 0.313 | | | |
| Al(tot) | 0.457 | 0.334 | 0.363 | 0.351 | 0.436 | 0.350 | 0.549 | 0.351 | 0.313 | 0.044 | 0.047 | 0.043 |
| Ti | 0.200 | 0.181 | 0.164 | 0.096 | 0.186 | 0.131 | 0.078 | 0.168 | 0.127 | 0.942 | 0.959 | 0.923 |
| Fe*** | 1.392 | 1.522 | 1.573 | 1.576 | 1.417 | 1.568 | 1.430 | 1.530 | 1.488 | | | |
| Fe** | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.000 | 0.053 | 0.044 | 0.046 |
| Mn | 0.090 | 0.100 | 0.106 | 0.085 | 0.078 | 0.089 | 0.119 | 0.096 | 0.088 | 0.002 | 0.002 | 0.001 |
| Mg | 0.042 | 0.026 | 0.021 | 0.019 | 0.034 | 0.023 | 0.018 | 0.036 | 0.019 | | | |
| Ca | 2.920 | 2.941 | 2.865 | 2.937 | 2.931 | 2.905 | 2.852 | 2.869 | 2.929 | 0.965 | 0.993 | 0.987 |
| Na | 0.026 | 0.000 | 0.036 | 0.000 | 0.000 | 0.018 | 0.000 | 0.000 | 0.000 | | | |
| K | | | | | | | | | | | | |
| Y | | 0.008 | 0.007 | | | | 0.008 | 0.006 | | 0.016 | 0 | |
| Zr | 0.026 | 0.012 | 0.013 | 0.014 | 0.032 | 0.017 | 0.006 | 0.031 | | 0.011 | 0.011 | 0.008 |
| Nb | | | | | | | | | | 0.006 | 0.002 | 0.009 |
| Ce | | | | | | | | | | 0.004 | 0.003 | 0.009 |

Note. C – core; R – rim.

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Table 4. Chemical analyses of zeolite group minerals and "altered nepheline" from Breivikbotn carbonatite.

| Mineral | Natrolite | | | | Gonnardite | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Sample | 13-1 | 13-3 * | 13-3 * | 13-3 * | 13-3 * | 13-3 * | 13-3 * | 13-3 * | 13-3 * |
| | wt. % | | | | | | | | |
| $SiO_2$ | 45.22 | 46.05 | 46.35 | 45.04 | 41.28 | 39.47 | 38.85 | 40.44 | 38.40 |
| $Al_2O_3$ | 20.38 | 26.46 | 27.33 | 28.36 | 30.20 | 30.50 | 30.56 | 28.89 | 30.16 |
| CaO | 0.08 | 0.12 | 0.31 | 1.48 | 4.17 | 5.12 | 6.43 | 4.34 | 6.25 |
| $Na_2O$ | 16.61 | 15.67 | 15.13 | 14.41 | 12.67 | 12.96 | 11.41 | 11.80 | 11.62 |
| $K_2O$ | | 0.02 | 0.03 | 0.02 | 0.03 | 0.04 | 0.02 | 0.00 | 0.02 |
| SrO | | | | | | | | | |
| Total | 82.29 | 88.32 | 89.15 | 89.31 | 88.36 | 88.09 | 87.28 | 85.47 | 86.44 |
| | apfu (7 cations) | | | | apfu (7 cations) | | | | |
| Si | 3.118 | 2.991 | 2.997 | 2.919 | 2.727 | 2.605 | 2.617 | 2.775 | 2.605 |
| Al | 1.656 | 2.026 | 2.083 | 2.166 | 2.352 | 2.372 | 2.426 | 2.336 | 2.411 |
| Ca | 0.006 | 0.008 | 0.021 | 0.103 | 0.295 | 0.362 | 0.464 | 0.319 | 0.454 |
| Na | 2.220 | 1.974 | 1.897 | 1.810 | 1.622 | 1.658 | 1.490 | 1.570 | 1.528 |
| K | | 0.002 | 0.002 | 0.002 | 0.003 | 0.003 | 0.002 | 0.000 | 0.002 |
| Sr | | | | | | | | | |

Note. * - mineral species were confirmed by XRPD.

Table 4. Contd.

| Mineral | Thomsonite-(Ca) | | | | | | | | Altered nepheline | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Sample | 13-4a | 13-4 | 13-4 | 14-3 * | 14-3 * | 14-3 * | 14-3 * | 14-3 * | 13-1 | 13-1 | 13-1 |
| | wt. % | | | | | | | | | | |
| $SiO_2$ | 36.34 | 35.58 | 34.93 | 37.28 | 37.07 | 36.84 | 37.59 | 37.54 | 49.35 | 42.87 | 41.36 |
| $Al_2O_3$ | 32.06 | 31.56 | 30.36 | 29.57 | 30.23 | 30.04 | 29.54 | 29.47 | 36.67 | 42.32 | 41.09 |
| CaO | 13.16 | 12.19 | 7.08 | 11.36 | 12.61 | 12.08 | 12.06 | 11.14 | 0.24 | 6.05 | 5.97 |
| $Na_2O$ | 3.82 | 3.75 | 3.68 | 4.09 | 4.38 | 4.35 | 5.07 | 4.57 | 14.57 | 10.86 | 10.80 |
| $K_2O$ | | | | | | | | | | | |
| SrO | 0.73 | 2.69 | 11.76 | 3.21 | 1.75 | 2.14 | 1.80 | 1.71 | | | |
| Total | 86.12 | 85.78 | 87.81 | 85.51 | 86.05 | 85.45 | 86.07 | 84.43 | 100.83 | 102.09 | 99.22 |
| | apfu (13 cations) | | | | | | | | apfu (3 cations) | | |
| Si | 4.932 | 4.940 | 5.176 | 5.220 | 5.070 | 5.092 | 5.116 | 5.225 | 1.223 | 1.069 | 1.059 |
| Al | 5.129 | 5.164 | 5.302 | 4.878 | 4.874 | 4.894 | 4.739 | 4.835 | 1.071 | 1.244 | 1.240 |
| Ca | 1.914 | 1.813 | 1.123 | 1.705 | 1.848 | 1.790 | 1.759 | 1.662 | 0.006 | 0.162 | 0.164 |
| Na | 1.005 | 1.009 | 1.058 | 1.109 | 1.161 | 1.166 | 1.338 | 1.232 | 0.700 | 0.525 | 0.537 |
| K | | | | | | | | | | | |
| Sr | 0.019 | 0.073 | 0.341 | 0.088 | 0.047 | 0.058 | 0.048 | 0.047 | | | |

Note. * - mineral species were confirmed by XRPD.

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## 5. Whole Rock Compositions

### *5.1. Major Elements*

Major element concentrations of 28 rock samples (17 carbonatitic and 11 "malignite" samples) have been analyzed. Representative analyses are given in Table 5, while all analyses and CIPW norms are available in the Supplementary Data Table S1.

*Carbonatite* has $SiO_2$ contents in the range of 20-36 wt % (average 31 wt %), $Al_2O_3$ = 2.5-15 wt %, MgO = 1.1-4.2 wt %, CaO = 17-34 wt %, $TiO_2$ = 1.0-2.3 wt %, $Na_2O$ = 1.8-6.5 wt %, $CO_2$ = 6-15.7 wt % and $P_2O_5$ = 0.27-1.44 wt %. Relatively large variations in aluminium, sodium, phosphorous and carbon oxide reflect variable modal contents of zeolite group minerals, apatite and calcite. The high contents of $Fe_2O_3$ (4-9.7 wt %) and FeO (3-7 wt %) can be explained by elevated contents of andradite, magnetite and possibly pyroxene. The content of $K_2O$ is low and varies from 0.27 to 1.1 wt % (average 0.54 wt %). The Mg# ranges from 20-56. The CIPW composition of carbonatite is characterized by the prevalence of *calcite* (15 – 36 wt %), *nepheline* (6-20 wt %), *diopside* (6-29 wt %), *hedenbergite* (5-16 wt %), *magnetite* (6-14 wt %) and appearance of *acmite* (up to 3.3 wt %).

*"Malignite"* is characterized by $SiO_2$ contents in the range of 35-39 wt %, which is much lower than in true malignite worldwide, and reflects its melanocratic features. $Al_2O_3$ varies in the range 10-17 wt %, MgO = 0.2-1.1 wt %, CaO = 17-26 wt %, $TiO_2$ = 0.8-2.1 wt %, $Na_2O$ = 2-6 wt %, $K_2O$ = 0.26-1.38 wt %, $CO_2$ = 0.8-3.35 wt % and $P_2O_5$ = 0.17-0.26 wt %. "Malignite" also shows elevated contents of $Fe_2O_3$ (6.7-13.6 wt %) and FeO (1.2-3.5 wt %). The Mg# ranges from 10-20. Compared to carbonatite, "malignite" has higher silica, iron, potassium and lower phosphorus. The CIPW norms of rock are characterized by the appearance of *nepheline* (9-21 wt %), *wollastonite* (20-41 wt %), *diopside* (1.3-8.7 wt %), *magnetite* (3-10 wt %), *hematite* (2-9 wt %) and *orthoclase* (up to 8.2 wt %).

### *5.2. Trace Elements*

Trace element analyses of representative samples are given in Table 6 (the complete data set is available in the Supplementary Data Table S1). Five samples of carbonatite were analyzed by ICP-MS for a broad range of elements, while the Rb, Sr, Y, Zr, Nb analyses are available for the rest of the carbonatite samples and the "malignite" samples.

The *carbonatite* is strongly enriched in large-ion lithophile elements (LILE), particularly LREE (880-1900 ppm), Sr (2700-8900 ppm) and Ba (200-1000 ppm) (Fig. 5), as compared to the primitive mantle [30]. Mantle-normalized patterns show strong to moderate negative anomalies of K, Pb, P and and Ti (Fig. 5). Compared to average calico-carbonatite, the Breivikbotn carbonatite has the lower contents of most incompatible elements, except of K, Zr and Hf. Chondrite-normalized REE patterns (Fig. 6) show negative slopes ($(La/Yb)_n$ = 6-70), but not as steep as in "average" carbonatite. The REE patterns and the large variations in the REE content of the rocks reflect variations in the modal content of garnet, which is responsible for the accumulation of HREE. The carbonatite does not show any Eu anomalies (Eu/Eu* = 0.9-1.1).

The *"malignite"* shows elevated concentrations of Sr (650-3900 ppm, average 1580), Zr (1050-1350 ppm, average 1230) and Nb (35-125 ppm, average 80). Compared to the carbonatite, the "malignite" is characterized by higher Zr, but lower Nb, Y and Sr.

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Table 5. Representative major and minor element analyses of the Breivikbotn carbonatite and alkaline rocks (wt. %).

| **Sample** | **13_1** | **13_2** | **13_3** | **13_4** | **13_5** | **B9.5** | **B19.4** | **B21.4** | **B11.1** | **H6.2** |
|---|---|---|---|---|---|---|---|---|---|---|
| Rock | carbonatite | | | | | | | | | |
| $SiO_2$ | 31.74 | 26.4 | 32.1 | 28.97 | 26.51 | 31.54 | 28.67 | 28.96 | 29.76 | 20.47 |
| $TiO_2$ | 1.73 | 1.1 | 1.86 | 1.89 | 1.51 | 0.87 | 0.99 | 0.98 | 1.62 | 1.21 |
| $Al_2O_3$ | 11.7 | 3.35 | 11.27 | 14.9 | 2.46 | 13.11 | 13.82 | 13.63 | 11.71 | 6.87 |
| $Fe_2O_3$ | 6.12 | 8.92 | 6.93 | 9.69 | 5.25 | 5.46 | 4.41 | 4.37 | 5.19 | 9.62 |
| FeO | 7 | 6.19 | 5.74 | 5.48 | 3.63 | 4.62 | 3.46 | 3.43 | 3.64 | 6.39 |
| MnO | 0.52 | 0.57 | 0.56 | 0.45 | 0.36 | 0.5 | 0.43 | 0.44 | 0.47 | 0.69 |
| MgO | 2.22 | 3.98 | 2.17 | 1.1 | 4.22 | 2.33 | 1.99 | 2.13 | 2.08 | 1.49 |
| CaO | 20.46 | 30.22 | 21.49 | 19.98 | 33.6 | 21.32 | 22.54 | 22.5 | 24.16 | 30.41 |
| $Na_2O$ | 4.08 | 1.8 | 4.52 | 3.1 | 1.78 | 4.77 | 5.82 | 5.75 | 5.13 | 2.44 |
| $K_2O$ | 1.1 | 0.41 | 0.67 | 0.41 | 0.27 | 0.56 | 0.33 | 0.33 | 0.29 | 0.27 |
| $H_2O$ | 0.89 | 0.46 | 0.91 | 0.64 | 0.4 | | | | | |
| LOI | 2.94 | 0.26 | 3.83 | 5.35 | 2.06 | | | | | |
| $P_2O_5$ | 0.45 | 0.95 | 0.52 | 0.27 | 1.44 | 0.76 | 0.78 | 0.79 | 0.63 | 0.68 |
| F | 0.068 | 0.086 | 0.067 | 0.042 | 0.1 | | | | | |
| Cl | 0.011 | 0.011 | 0.029 | 0.014 | 0.02 | | | | | |
| $CO_2$ | 7.91 | 13.17 | 6.57 | 6.07 | 15.35 | 9.65 | 10.72 | 10.57 | 10.41 | 15.74 |
| Total | 98.94 | 97.88 | 99.24 | 98.36 | 98.96 | 95.49 | 93.96 | 93.88 | 95.09 | 96.28 |

Table 5. Contd.

| **Sample** | **H40.3** | **H10.7** | **H41.3** | **H11.6** | **H15.5** | **H31.3** | **H48.2** |
|---|---|---|---|---|---|---|---|
| Rock | "malignite" | | | | | | |
| $SiO_2$ | 34.95 | 37.1 | 37.46 | 37.53 | 37.71 | 39.18 | 39.43 |
| $TiO_2$ | 2.01 | 1.94 | 1.59 | 1.61 | 1.39 | 1.49 | 0.83 |
| $Al_2O_3$ | 9.72 | 11.89 | 12.91 | 12.66 | 13.74 | 13.05 | 17.35 |
| $Fe_2O_3$ | 13.59 | 12.39 | 10.85 | 10.39 | 10.23 | 10.27 | 6.73 |
| FeO | 2.79 | 2.55 | 2.96 | 3.5 | 2.86 | 2.89 | 1.16 |
| MnO | 1.19 | 0.99 | 0.91 | 0.91 | 0.84 | 0.87 | 0.55 |
| MgO | 0.77 | 0.49 | 1.02 | 1.08 | 0.82 | 0.92 | 0.24 |
| CaO | 26.42 | 23.08 | 21.28 | 21.41 | 20.41 | 20.16 | 17.12 |
| $Na_2O$ | 2.04 | 2.94 | 3.82 | 3.53 | 3.97 | 3.74 | 5.97 |
| $K_2O$ | 0.45 | 0.77 | 0.66 | 0.77 | 0.77 | 1.16 | 1.15 |
| $P_2O_5$ | 0.23 | 0.20 | 0.26 | 0.26 | 0.33 | 0.25 | 0.17 |
| $CO_2$ | 1.39 | 0.84 | 1.49 | 1.59 | 1.6 | 1.42 | 2.96 |
| Total | 95.55 | 95.18 | 95.21 | 95.24 | 94.67 | 95.40 | 93.66 |

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Table 6. Representative trace elements and REE analyses of the Breivikbotn carbonatite and alkaline rocks (ppm).

| Sample | 13_1 | 13_2 | 13_3 | 13_4 | 13_5 | B19.4 | B21.4 | B11.1 | H6.2 | H40.3 | H10.7 | H41.3 | H11.6 | H31.3 | H48.2 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Rock | carbonatite | | | | | | | | | "malignite" | | | | | |
| La | 214.4 | 265.7 | 190.5 | 150.4 | 470.2 | | | | | | | | | | |
| Ce | 449.7 | 516.8 | 421.8 | 341.1 | 930.9 | | | | | | | | | | |
| Pr | 49.2 | 51.8 | 47.6 | 3   9 | 96.4 | | | | | | | | | | |
| Nd | 189.6 | 174.4 | 209.4 | 169.7 | 324.2 | | | | | | | | | | |
| Sm | 40 | 25.3 | 49.4 | 42.4 | 56.2 | | | | | | | | | | |
| Eu | 12.1 | 6.66 | 1   6 | 13.9 | 15.1 | | | | | | | | | | |
| Gd | 29.7 | 19.8 | 35.7 | 31.1 | 42.5 | | | | | | | | | | |
| Tb | 4.86 | 2 . 2 | 6.87 | 6.06 | 5.77 | | | | | | | | | | |
| Dy | 24.5 | 7.83 | 35.4 | 31.9 | 23.7 | | | | | | | | | | |
| Ho | 4.57 | 1.26 | 6.79 | 6.32 | 4.12 | | | | | | | | | | |
| Er | 12.5 | 2.95 | 18.9 | 17.2 | 9.17 | | | | | | | | | | |
| Tm | 1.84 | 0.37 | 2 . 8 | 2.58 | 1.14 | | | | | | | | | | |
| Yb | 12.7 | 2.59 | 19.2 | 18.3 | 7.14 | | | | | | | | | | |
| Lu | 1.87 | 0.43 | 2.88 | 2.72 | 0.97 | | | | | | | | | | |
| Y | 111.7 | 30.8 | 149.8 | 148.3 | 84 | 30 | 32 | 124 | 28 | 43 | 60 | 46 | 44 | 42 | 14 |
| Ta | 3.23 | 0 . 8 | 3.27 | 4.19 | 18 | | | | | | | | | | |
| Nb | 111.2 | 20.7 | 106.3 | 1 2 4 | 231.4 | 188 | 187 | 136 | 42 | 88 | 103 | 78 | 85 | 92 | 34 |
| Hf | 13.7 | 8.05 | 19.9 | 17.9 | 9.84 | | | | | | | | | | |
| Zr | 492.5 | 363.2 | 696.7 | 7 1 6 | 473.1 | 493 | 505 | 840 | 616 | 1359 | 1350 | 1133 | 1137 | 1145 | 1043 |
| Sr | 4735 | 5803 | 3290 | 8856 | 4656 | 6169 | 6173 | 3780 | 8985 | 645 | 958 | 1211 | 1235 | 1505 | 3993 |
| Rb | 16.4 | 5.82 | 1   0 | 5.23 | 6.41 | bd | bd | bd | bd | 8 | 10 | 6 | 8 | 12 | 9 |
| Ba | 618.5 | 638.4 | 204.7 | 178.2 | 1003 | | | | | | | | | | |
| U | 3.46 | 0 . 8 | 3.99 | 3.13 | 1.86 | | | | | | | | | | |
| Th | 12.7 | 5.63 | 13.4 | 9.43 | 25.8 | | | | | | | | | | |
| Pb | 4.17 | 1.86 | 3.53 | 3 . 2 | 2.81 | | | | | | | | | | |
| Mo | 5.02 | 1 . 1 | 8 . 9 | 13.8 | 0.62 | | | | | | | | | | |
| Cs | 0.16 | 0.016 | 0.11 | 0.02 | 0.091 | | | | | | | | | | |

## 6. Discussion

### *6.1. Petrographical Classification of the Breivkbotn Carbonatite and Alkaline Rocks*

The recommendations for the classification of carbonatites by the IUGS Subcommission on the Systematics of Igneous Rocks [31-33] defined carbonatites as “igneous rocks, intrusive as well as extrusive, which contain more than 50% by volume of carbonate minerals”. The samples of the Breivikbotn carbonate rocks studied here contain 20-50 vol. % of carbonate mineral. Strictly speaking the rocks should be referred to as calcitic ijolites, i.e. igneous rocks containing between 10 and 50% of igneous calcite, with additional other primary minerals, such as pyroxene and nepheline. Nevertheless, the rock is extremely taxitic with proportions of carbonate in parts close to the classification boundary at 50 vol. % (e.g. sample 13-3-1, see Fig. 1). In terms of the IUGS chemical classification, the rock can be referred to as silicocarbonatite if the $SiO_2$ content of the rock exceeds

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20%. Of this reason, we prefer to name the Breivikbotn carbonatite-like rocks as nepheline-diopside-calcite-carbonatite, or for simplicity, silicocarbonatite.

Nepheline is an unusual and rare mineral in classic carbonatite complexes world-wide. Nevertheless nepheline-bearing carbonatites have been reported from a number of carbonatite localities; e.g. Laacher See, Germany and Alnø, Sweden (both mentioned in [34]), Fen [35] and Lillebukt [36] in Norway, Chilwa Island and Kangankunde, Malawi [37], Budeda Hill and Homa Bay, Uganda [38, 39], Walloway, Australia [40], Dicker Willem, Namibia [41], and in Ilmeny–Vishnevogorsky, Urals, Russia [42] and from other Russian

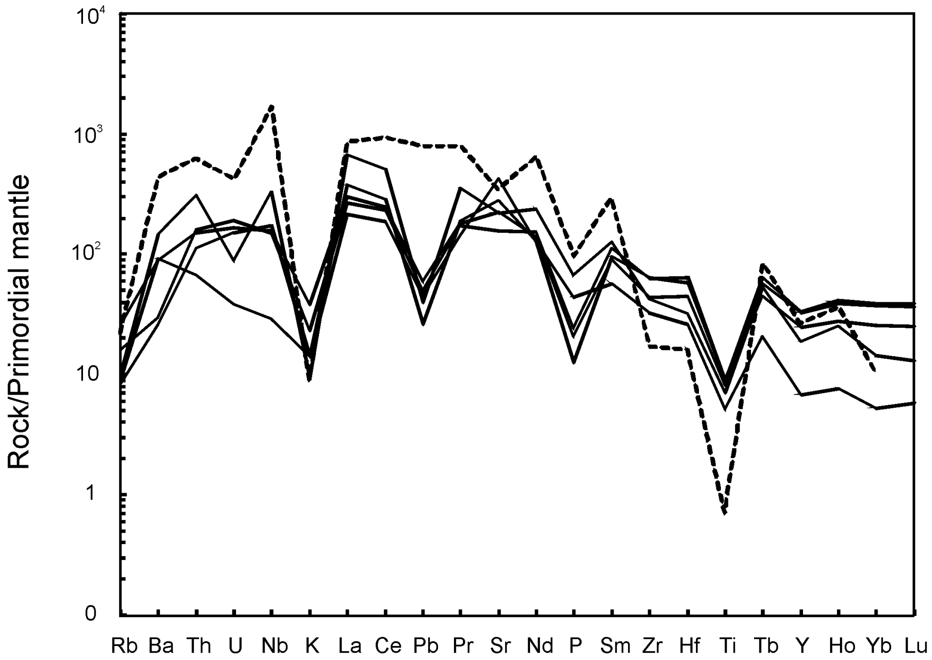


**Figure 5.** Mantle normalized incompatible elements patterns of silicocarbonatite from Breivikbotn occurrence. Curve for average carbonatite [32] is dotted and is shown for reference. Primordial mantle compositions used are taken from [30].

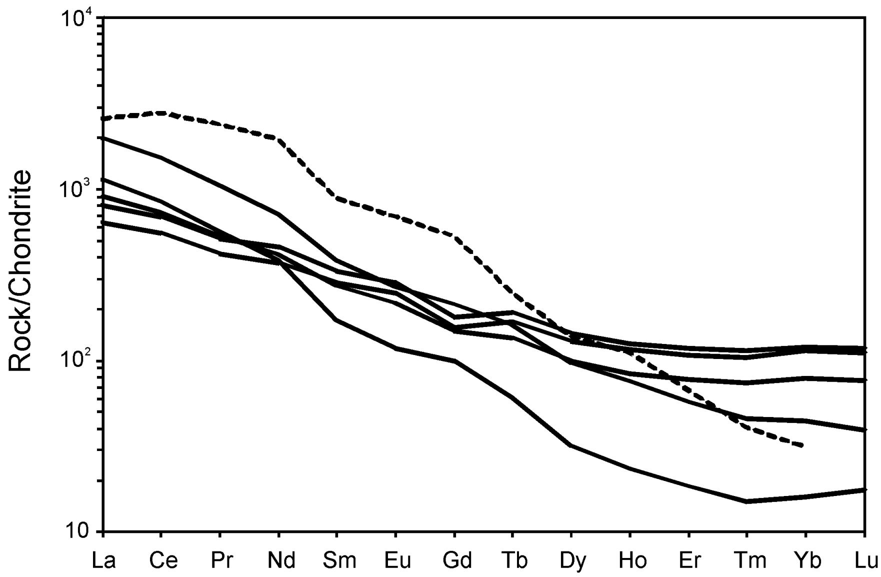


**Figure 6.** Chondrite normalized REE patterns of the silicocarbonatite from Breivikbotn. Curve for average carbonatite [32] is dotted and is shown for reference. Chondrite compositions used are taken from [30].

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occurrences (summarized by [43]. In addition, other carbonatites contain natrolite, analcime or cancrinite, formed by breakdown of nepheline; e.g. Oka, Canada [44]; Legetet Hills, Kenya, Nachendezwaya, Tanzania, Tororo and Bukusu, Uganda (all summarized by [34]). On the basis of this distribution, and the petrographic and mineralogical evidence for the above described localities, it is possible to infer that nepheline-bearing carbonatites are often associated with the "nephelenitic-clan" rather than with the "melilititic-clan" carbonatites (using the terminology of [45]).

The "malignite" from the Breivikbotn occurrence has an unusual mineralogical composition for magmatic rocks. Therefore, the classification based on the chemical composition of the rock was applied. Based on the $SiO_2$ and $Na_2O+K_2O$ contents, the rock can be considered to belong to the clans of melilitolite or ultramafic foidolite. The last clan include melteigite, jakupirangite, ijolite, urtite and missurite, and the Na-rich (Na>K) and melanocratic varieties of the clan are comparable to the rock discussed here, namely melteigite and jakupirangite. Furthermore, the low MgO and relatively high $Al_2O_3$ of the Breivikbotn rock suggest that it could be classified as melteigite, but the elevated $CO_2$ content (0.8 – 3.4 wt %) justify that the rock is referred to as a carbonated melteigite. Broadly speaking, it can be stated that Breivikbotn "malignite" crystallized from a carbonated foidite melt.

Thus the Breivikbotn occurrence provides good field material for studying of the genetic link between carbonatite and foidite magma (i.e. the derivation of carbonatite magma, either by liquid immiscibility or crystal fractionation from carbonated foidite).

### *6.2. Geochemical Constraints on the Petrogenesis and Geodynamic Setting of Breivikbotn Complex*

The proposed origins of carbonatites in association with silicate alkaline rocks include three main processes: (1) direct melting from carbonate-rich mantle peridotite or wehrlite [46-49]; (2) low-pressure liquid immiscibility from parental nephelinite magmas (e.g. [50]); (3) extensive crystal fractionation from carbonated alkaline silicate magmas, i.e. carbonated nephelinite or melilitite [48, 51-53].

Robins [36] suggested that the primary Breivikbotn carbonatite melt was derived from carbonated nephelinite magma through liquid immiscibility. According to the model of Robins [36], nephelinite magma fractionated through mela-phonolite magma to «malignite» and phonolite (yielding nepheline syenite), and through trachyte magma to alkali syenite. Parts of the model are questionable because of its complexity, and the absence at the outcrops of a number of hypothetical magmatic phases. Moreover, if the model is correct, it would be expected that the Breivikbotn carbonatitic and alkaline rocks contain significantly more highly incompatible HFS elements than nephelinite [50]. It is therefore clear that neither liquid immiscibility nor fractional crystallization can explain the formation of the studied rocks from Breivikbotn. We consider it more likely that the primary melt of the studied rocks corresponds to "malignite" or carbonated melteigite.

For the Breivikbotn case, direct melting of silicocarbonatite from carbonate-rich mantle rock seems unlikely because of: (1) low #Mg (17-56, average 37), (2) the association of carbonatite and alkaline silicate rocks at the outcrops, (3) through this process, Mg-carbonate would be expected to dominate.

The derivation of a melt from carbonated foidite by liquid immiscibility or crystal fractionation can be identified by geological, petrographic and mineralogical criteria, but also on the basis of trace-element geochemistry, particularly in relation to the associated silicate lithologies. The HFSE are highly informative in this respect. Data available for both the silicocarbonatitic and the alkaline silicate rocks from Breivikbotn include Zr, Nb and Y. Nb is a highly incompatible element, whereas Zr and Y are more compatible. Therefore it can be expected that Nb partitioned into the carbonatitic melt relative to Zr and Y both during liquid immiscibility and crystal fractionation. The prevalence of Nb in the Breivikbotn silicocarbonatite compared to the alkaline silicate rock supports the proposed petrogenetic processes. Noteworthy, the absence of any gap in the Nb-Zr/Nb diagram (Fig. 7) indicate crystal fractionation from a carbonated foidite melt (of "malignite" or melteigite composition, as proposed above). Zr is strongly partitioned into garnet, and would be incorporated

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in the early magmatic melanite in the Breivikbotn melteigite, which is reflected in Y-Zr/Y diagram (Fig. 7). The role of crystal fractionation during formation of the silicocarbonatite is also supported by its relatively high Th/U ratios (3-14; average 6.2) and low Zr/Hf (35-48; 41) and Nb/Ta (13-34; 27) ratios [54-56]. Fractionation of zircon, titanite and clinopyroxene can explain such element distributions.

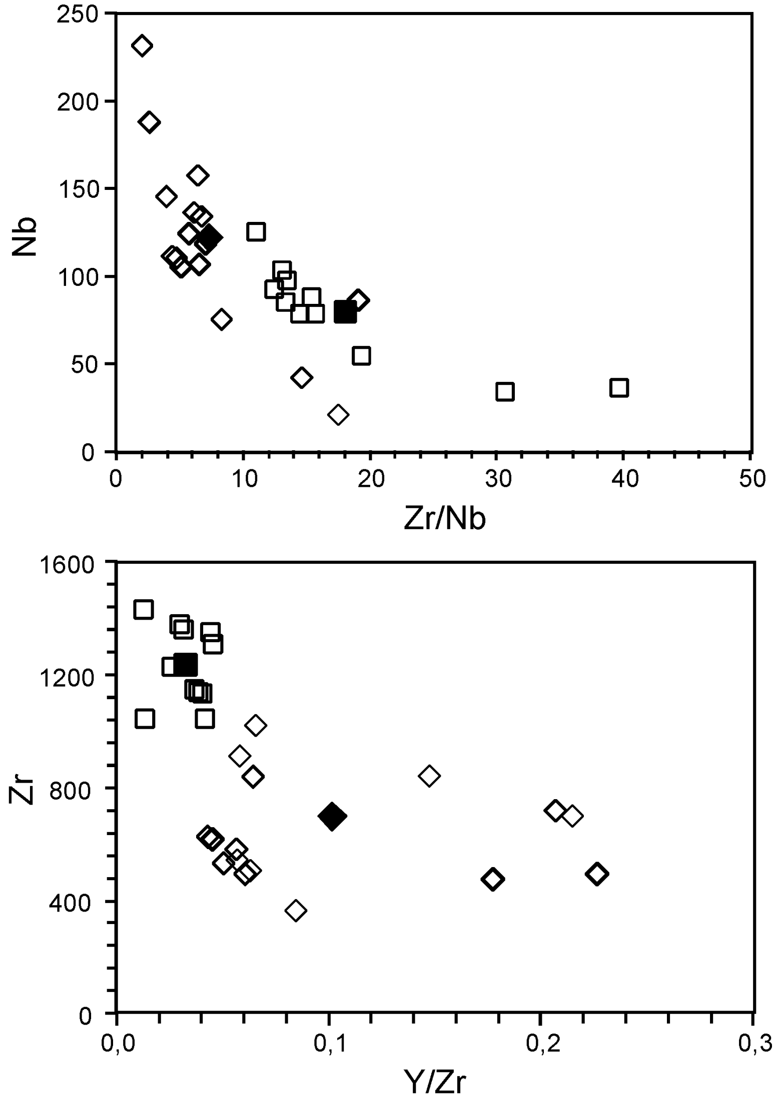


**Figure 7.** Selected paired trace-element ratios for the Breivikbotn silicocarbonatite and associated melteigite (empty diamonds and squares, respectively; solid symbols are average).

Field observations, such as the gradual boundaries between silicocarbonatite and melteigite, the dykes of pure carbonatite cutting the melteigite, and xenoliths of melteigite in silicocarbonatite is in agreement with a process of crystal fractionation of the carbonatitic rocks from a foidite magma.

The concept of synorogenic *vs* extensional origin of the Seiland igneous province (see above) can be addressed using geochemical data. Trace element concentrations can provide important information about the tectonic setting of carbonatites. Most carbonatites in within-plate settings, e.g. continental rifts, are characterized by high concentrations of LILE (especially Sr and Ba) and HFS elements; the average values for Zr, Nb, Hf and Ta are 256.4, 308.9, 4.3 and 8.9 ppm, respectively [54]. Subduction- and collision-related carbonatites in off-craton settings have high LILE contents, but much lower Zr, Nb, Hf and Ta. For example, Chakhmouradian et al. [55] reported for the Eden Lake carbonatite the following values (ppm): 47-98 for Zr, 4.0 for Nb, 1.5-2.4 for Hf, and 0.2 for Ta.

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Similar values are reported from the subduction-related carbonatites in Italy, Antarctica, north-eastern China, Northern Norway [3, 6, 57, 58], and even much lower from collision related carbonatites in southwestern and northern China [2, 4]. The Breivikbotn carbonatite with average values for Sr = 5100 ppm, Ba = 530 ppm, Zr = 700 ppm, Nb = 122 ppm, Hf = 14 ppm, and Ta = 6 ppm has the signature of within-plate carbonatites.

### *6.3. Late- to Post-Magmatic Alteration and Possible Metamorphic Overprints*

Because of the alkalic and volatile-rich character, the silicocarbonatite resulted in the significant fenitization that can be observed at the Breivikbotn outcrops (see above). Moreover, the presence of late-magmatic amphibole and biotite suggests that the silicocarbonatite magma contained sufficient water to produce voluminous hydrothermal fluids during late stages of crystallization. Such deuteric fluids would be able to alter the primary mineral assemblage producing the abundant water-bearing phases that can be observed. The metasomatic alteration can be considered to be the result of autometasomatism, or subsolidus process, in which early magmatic phases reacted with their own residual fluids to form a suite of low-temperature minerals such as natrolite, gonnardite, and thomsonite. Experimental studies, quantitative estimates and field observations indicate that natrolite-type and thomsonite-type zeolites formed in the temperature range of 100-200 °C [59-62].

Earlier petrographic studies have shown that ZGM could be the products of nepheline alteration. Several reactions for nepheline alteration at successive enrichment of $H_2O$, CaO $Al_2O_3$ and $SiO_2$ in hydrothermal fluid might be possible:

$$2NaAlSiO_4(Ne) + SiO_2(aq) + 2H_2O(aq) = Na_2(Si_3Al_2)O_{10}\cdot 2H_2O\ (Ntr)$$

$$NaAlSiO_4(Ne) + SiO_2(aq) + CaO(aq) + Al_2O_3(aq) + 3H_2O(aq) = (Na,Ca)_2(Si,Al)_5O_{10}\cdot 3H_2O\ (Gonn)$$

$$NaAlSiO_4(Ne) + 4SiO_2(aq) + 2CaO(aq) + 2Al_2O_3(aq) + 6H_2O(aq) = NaCa_2(Al_5Si_5)O_{20}\cdot 6H_2O\ (Toms)$$

Textural evidence for replacements of natrolite by gonnardite is supported by the successive enrichment of the fluid phase by $H_2O$, CaO, $Al_2O_3$ and $SiO_2$ according to following reaction:

$$Na_2(Si_3Al_2)O_{10}\cdot 2H_2O\ (Ntr) + 2SiO_2(aq) + 2CaO(aq) + 2Al_2O_3(aq) + 4H_2O(aq) = 2(Na,Ca)_2(Si,Al)_5O_{10}\cdot 3H_2O\ (Gonn)$$

Ti-rich garnets can form over a wide range of temperatures and pressures. Particularly in alkaline rocks, they appear to reflect complex late stage metasomatic reactions between earlier-formed mafic minerals and late-stage fluids [63, 64]. This idea is consistent with the petrographic observations from the garnet-bearing rocks from the Breivikbotn outcrops, which indicate that Ti-rich garnets formed from clinopyroxene and amphibole, possibly driven by metasomatic fluids. We suggest that these reactions either occurred as a result of alkali metasomatism influencing the evolution of the Breivikbotn complex in a fluid-enriched environment, or in respons to a late metamorphic event during the Caledonian orogeny. Metamorphic overprint of the Breivikbotn rocks can also explain the elevated contents of Al and Fe in titanite from the silicocarbonatite (see above).

## 7. Conclusions

The Breivikbotn carbonatite-alkaline complex consists of silicocarbonatite and alkaline ultramafic rock of an unusual mineralogic composition. The silicocarbonatite is composed mainly of primary magmatic calcite, pyroxene, altered nepheline, late-magmatic amphibole, post-magmatic garnet (andradite) and zeolite group minerals. The alkaline ultramafic rock, which has a composition similar to melteigite, is composed of garnet (melanite), pyroxene, alkali feldspar and altered nepheline. Geological observations and geochemical data suggest that the melteigite is an early magmatic phase, being parental for the silicocarbonatite, which formed by fractional crystallization from carbonated foidite melt.

Zeolites are the products of late/post-magmatic alteration of magmatic nepheline at low temperature (<200°C), Ca-, Al- and Si-bearing hydrous fluids. The alteration of nepheline to zeolite group minerals progressed through several steps: (1) "altered nepheline", (2) natrolite, (3) gonnardite , and (4) Sr-rich thomsonite-(Ca). The main prerequisites for zeolite formation in the carbonatite complex were (1) silicocarbonatite composition of the parent magma and crystallization of nepheline, (2) fractional crystallization of primary magmas leading to fluid enrichment of residual melts/hydrothermal solutions, (3) extensive fluid alteration of the rock at late- and post-magmatic stages.

Textural and compositional data for andradite distinctly point to its formation either due to alkali metasomatism at late-, post-magmatic stage of the Breivikbotn complex, or during the Caledonian metamorphic event.

**Supplementary Materials:** The following is available online at www.mdpi.com/xxx/s1, Table S1: Chemical composition and CIPW norms of the Breivikbotn carbonatites and alkaline rocks.

**Author Contributions:** Conceptualization, D.Z. and K.K.; Methodology, E.S., Y.S. and M.T..; Investigation, D.Z., K.K. and E.R.; Writing-Original Draft Preparation, D.Z.; Writing-Review & Editing, K.K. and E.R.; Visualization, D.Z. and Y.S.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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