Orogenic degassing, scapolitization and K-metasomatism during Caledonian exhumation, Helgeland, Norway

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Metasomatism associated with barren quartz veins cross-cutting hornblende schist in the upper part of the metasedimentary sequence of the W–Mo-mineralized Bjellatinden area was studied using P–T modeling of the mafic schist host and alteration assemblages, fluid inclusions, and isocon diagrams. Two main alteration assemblages are associated with the quartz veins: A biotite-scapolite-dominated zone (BSZ), and a zone dominated by chlorite, muscovite and calcite (CMZ), replacing the former BSZ and extending further into the wall rock. Three types of fluid inclusions are observed: CO₂-rich (type I), low-salinity H₂O-rich (type II), and high-salinity H₂O-rich (type III). Type III fluids are strongly associated with the CMZ, and are not observed in the BSZ, whereas type I and II fluids are observed in quartz and scapolite in the BSZ. Type I and II fluid inclusions found in quartz in the CMZ are interpreted to be relics from the earlier BSZ alteration. Plagioclase-amphibole thermobarometry of grains in the hornblende schist indicates a near-isothermal pressure drop from ca. 660 to ca. 560 MPa at ca. 690°C. Type I and II fluids are interpreted to have infiltrated extension fractures at ca. 240 MPa and 600°C giving rise to the BSZ, and only minor mass exchange with the hornblende schist. Type III fluids are responsible for K-metasomatism of the existing alteration assemblage, and infiltrated the pre-existing fracture system over a large temperature range, from ca. 600°C to ca. 350°C during near-isobaric cooling at ca. 200 MPa. The CMZ was stabilized towards the end of this fluid infiltration. The high-salinity fluids are interpreted to have exsolved from an underlying, unexposed, granitic intrusion. This intrusion may also be the source of the W–Mo mineralizations in the area.

Keywords: Metasomatism, W–Mo mineralization, scapolite, fluid inclusions, Caledonides

Electronic Supplement 1: Mineral chemical analyses.

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Introduction

The release of volatiles during orogenic exhumation has been well studied (e.g., Larsen et al., 1998; Smith & Yardley, 1999; Vry et al., 2010). While large volumes of metamorphic fluids may be produced, their potential to metasomatically alter rocks is generally much lower than fluids generated from intrusions or sedimentary formation waters (Yardley, 2013). In this study we focus on hydrothermal alteration by fluids produced during the late stages of the Caledonian (Scandian) orogeny in the Bjellatinden area, located in the Meløy and Gildeskål

municipalities, Nordland County, northern Norway. A number of W and Mo skarn and vein mineralizations are recorded in the area, two of which were mined periodically for molybdenite between 1917 and 1944. Scheelite was first discovered in 1953 in samples from the abandoned Oterstrand mine. Later, several Mo, W and U anomalies were found in the contact zone between the Caledonian nappe complexes and Precambrian basement rocks, and in stream sediments in the larger Bodø–Glomfjord region (Stendahl et al., 1993). Stratabound W mineralizations in Nordland have been explained as volcanic exhalative (Skaarup, 1974), magmatic (Bowitz-Ihlen, 1973), and most recently, metamorphic (Larsen,

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1991). The apparent lack of intrusive bodies related to the Bjellatinden mineralizations, its semi-conformable character, and abundance of scapolite in the richest mineralized skarn body, make it comparable to examples from Massif Central, France (Gibert et al., 1992), Brazil (Barbosa et al., 1986), and the Archean Malene supracrustals (Appel, 1986). This area has had a complex fluid history, with at least two generations and sources of fluids infiltrating the metasedimentary stratigraphy. Most prominent, and not previously observed, are the brine fluids responsible for the K-metasomatism of the hornblende-mica schist during extensional collapse of the Caledonian orogeny in the Early Devonian.

A major enigma in explaining the W mineralization has been the dominance of nearly pure CO₂ fluid inclusions related to the scheelite-bearing lithologies (Larsen, 1991). Given that W ions preferably are transported as various tungstates (Wood & Samson, 2000), an aqueous component is required. In this study we report results from a poorly studied part of the area where several generations of volatiles were discovered. The fluids occur in structures and textural settings that relate to various episodes of exhumation of the orogenic roots, from possibly peak T metamorphism to the more advanced stages of orogenic collapse. Accordingly, when appended to the previous results, the Bjellatinden area exposes a well-preserved record of fluid evolution during the late-Scandian extensional collapse of the Caledonian orogeny.

Geological setting

Regional geology

The Scandinavian Caledonides are commonly divided into four tectonostratigraphic units: Lower, Middle, Upper, and Uppermost Allochthons (Roberts & Gee, 1985), emplaced onto autochthonous Precambrian crystalline basement (Fig. 1A). Slices of crystalline basement and overlying shelf and continental-rise successions of Baltican origin, constitute the Lower and Middle Allochthons (Roberts & Gee, 1985; Andréasson, 1994). More recently, the Seve Nappe, interpreted to represent the Baltica–Iapetus continent-ocean transition (Stephens & Gee, 1985), has also been included in the Middle Allochthon (Gee et al., 2008). The Upper Allochthon comprises sedimentary and igneous rocks derived from the Iapetus Ocean, including ophiolites and island-arc complexes (e.g., Stephens, 1988; Grenne et al., 1999; Roberts, 2003) with faunas of Laurentian affinity (e.g., Bergström, 1979; Spjeldnæs, 1985; Bruton & Harper, 1988). The Uppermost Allochthon was formed in a continental margin setting of Laurentian origin (e.g., Melezhik et al., 2002; Roberts et al., 2002, 2007), consisting of various metasedimentary, carbonate and magmatic units, and is divided geographically by the Paleoproterozoic Tysfjord basement window (Skår,

2002). North of the Tysfjord window, a number of unnamed and named thrust sheets, divided into Niingen, Tromsø, Gratangen, Nakkedal, Evenes, Lyngsfjell and Bjerkvik nappe complexes, are included in the Uppermost Allochthon (Barker, 1986; Zwaan et al., 1998; Melezhik et al., 2003). The Gratangen Nappe (including the Bogen Group) has been correlated with the Tromsø Nappe farther north, and the ophiolitic Bjerkvik Nappe has been correlated with the Lyngen Magmatic Complex of the Lyngsfjell Nappe (Andresen & Steltenpohl, 1994), making the tectonostratigraphy (top to bottom) Niingen, Tromsø, Nakkedal, Evenes, and Lyngsfjell. South of the Tysfjord window, in tectonostratigraphic order top to bottom, Helgeland Nappe Complex (HNC), Beiarn Nappe Complex (BNC), and Rödingsfjället Nappe Complex (RNC) constitute the Uppermost Allochthon, with the HNC and RNC being the dominating units in terms of areal extent. The Fauske Nappe is in some maps shown as a separate unit (Roberts et al., 2007), but is generally interpreted to form the base of the RNC (Gustavson, 1996; Roberts et al., 2002). Correlations of the BNC to either the HNC (Stephens et al., 1985) or the RNC (Gustavson & Gjelle, 1991; Gustavson, 1996) have been proposed, but it may very well be seen as a separate complex (Augland et al., 2012). Zircon ages in the HNC resemble those in sedimentary zircons from the Dalradian Supergroup of Scotland (Barnes et al., 2007), while $\epsilon_{\text{Hf}(t)}$ -values in zircons from magmatic intrusions in the Beiarn Nappe correspond to those in the Hurry Inlet plutonic terrane in Liverpool Land, NE Greenland (Augland et al., 2012). Furthermore, Taconian structures in the HNC (Yoshinobu et al., 2002; Barnes et al., 2007) and the RNC (Roberts et al., 2002) underline the exotic origin of the Uppermost Allochthon.

Dating of migmatites and plutonic intrusions have indicated that the continental collision during peak Scandian orogenesis lasted ca. 25 million years from 425 Ma to 401 Ma, including a period of uplift of basement windows and related decompression melting responsible for the emplacement of pegmatite bodies from 409 ± 5 Ma to 401 ± 2 Ma (Larsen et al., 2002; Nordgulen et al., 2002), followed by extensional collapse and unroofing between 401 Ma and 398 Ma (Braathen et al., 2002; Eide et al., 2002; Osmundsen et al., 2003).

Geology of the Bjellatinden area

The quartz monzonitic 'Bjellatind granite' comprises, together with the larger Glomfjord, Svartisen, Høgtuva, and Sjona windows, a group of Proterozoic, autochthonous basement windows yielding zircon U–Pb ages of 1800 ± 3 to 1795 ± 5 Ma (Skår, 2002). Coinciding with late-Svecofennian magmatism and the earliest intrusions of the Transscandinavian Igneous Belt (Skår & Pedersen, 2003), they are interpreted to be a part of the western margin of the Fennoscandian Shield.

Figure 1. (A) Overview of the principal allochthonous units in northern Norway (Roberts et al., 2007). The studied area is indicated by the small rectangle. (B) Local map of the Bjellatinden area based on Wells & Bradshaw (1970), Larsen (1988) and this study. (C) Lithological column through the metasedimentary units. The column is representative of the area near the quartz veins 'X'. Based on Larsen (1988) and this study.

Overlying the Bjellatind granite is a metasedimentary suite of gneisses, schists, amphibolites, and marbles (Fig. 1B, C) known as the Meløy Group, or Meløy Nappe (Hollingworth et al., 1960). The lowermost lithologies of the Meløy Group ("*Lower Meløy Group*" of Hollingworth et al. (1960), and "*basal succession*" of Holmes, 1966) consisting of garnet augen gneiss and thin layers of psammitic and mica schist towards the top, are thought to be (par)autochthonous. The presumed original unconformity between the basement and the directly overlying strata was destroyed by intense F_1 folding (Rutland & Nicholson, 1965). The Gildeskål marbles have been correlated with the Fauske marbles around Saltfjord, a part of the Fauske Nappe (Rutland & Nicholson, 1965), and the Upper Meløy Group of Hollingworth et al. (1960) (corresponding to the Galtskartind succession of Holmes, 1966) is thus a part of the RNC of the Uppermost Allochthon.

The boundary between the hornblende gneiss and the overlying kyanite-bearing garnet- two- mica schist is in places marked by an intensely deformed marble layer of varying thickness (Larsen, 1988). Cribb (1981) also noted a difference in age between these two lithologies (1199 $\pm$ 161 Ma for the kyanite schist, and a maximum age of 758 Ma for the precursor of the hornblende gneiss) based on whole-rock Rb–Sr geochronology. This dating technique is today regarded as imprecise, and the 758 Ma age may

well represent a resetting of the whole-rock Rb–Sr system. However, intensive shearing of the marble unit associated with thinning and even disappearance implies a tectonic boundary (Larsen, 1991). The metasedimentary rocks occurring some tens of meters below the hornblende gneiss and the marble, where present, are characterized by intense deformation and W–Mo mineralized fractures, and are dubbed the W–Mo zone. All W–Mo mineralizations are situated in this 15–40 m-thick composite layer, comprising calcareous biotite schist, marbles, and calc-silicate rocks. Within this layer, scheelite and molybdenite occur in a scapolite replacement skarn, in addition to quartz, quartz-plagioclase, and quartz-calcite veins (Larsen, 1991). Scheelite also occurs in a 0.3–8 m-thick diopside replacement skarn, but here Mo is absent (Drivenes, 2010). A 20 cm-thick layer of diopside skarn is situated above a garnet-bearing schist in the upper lithologies, but no scheelite was observed in this layer. Typical grades vary from a few hundred $\mu\text{g/g}$ to 1.5 mass% W, and up to 0.2 mass% Mo. CO_2 dominated fluids imply formation at 450 to 525°C and 200 MPa (Larsen, 1991).

A ca. eight meter-thick layer of hornblende schist, situated above the garnet- two-mica schist, is in the lower three meters intersected by a stockwork of 5–15 cm-wide quartz veins with alteration halos up to 5 cm in width. The veins also cross-cut earlier quartz veins parallel to the foliation (Fig. 2). These veins and their alterations

Figure 2. Field appearance of cross-cutting quartz veins in location 'X' in Fig. 1B.

are the focus of this study. The veins gradually taper upwards into unfilled joint-like fractures continuing 1–2 meters on the very top. The vein swarm can be followed for 250 meters along the dip direction of the hornblende schist. Similar veins with the same orientation are located 200 meters along strike, but lacking the alteration halos. In these veins, dense clusters of coarse biotite grains (~5 cm) occur in the center of the veins. The boundary between the garnet- two-mica schist and the hornblende schist is marked by a one meter-thick quartzite layer. A one meter-thick marble layer, situated above the hornblende schist, is overlain by at least 100 meters of various amphibole-, garnet-, and carbonate-bearing schists and psammites.

A tourmaline-rich, S-type granitic pegmatite body, similar to other pegmatites that formed from decompression melting during gneiss dome uplift (Barnes et al., 2007), intruded at the stratigraphic level of the hornblende gneiss and the W–Mo zone. It consists of subconformable sheets 5–20 m thick, with subvertical dike offshoots intersecting the overlying lithologies.

Structural setting

Three major folding episodes dominate the area. The early F_1 episode is represented by isoclinal folds in the basement gneisses with an axial plane schistosity parallel to the cover rock contacts (Rutland & Nicholson, 1965), which formed during nappe emplacement (Wells & Bradshaw, 1970). These features were in turn refolded during a later F_2 episode producing tight, overturned folds, such as those observed in the marble layers between the hornblende gneiss and the garnet- two-mica schist, as well as larger fold structures such as the Spilderdalen antiform (Holmes, 1966). The most obvious structural element of the Bjellatinden area is the asymmetrical dome shape caused by F_3 folding during exhumations in the late stages of the orogeny. Large, open, antiformal domes were created by interfering folds with N–S and E–W fold axes (Fig. 1B). Layers on the Bjellatinden plateau are sub-horizontal, dipping ca. 10° W–SW, with increasing dips toward the flanks of the dome. On the northern slope of the dome, the layering is near-vertical to slightly overturned. Boudinage structures on the flanks of the dome, as exemplified by chocolate tablet boudins of hornblende gneiss in marble in one of the W-mineralized zones, and other boudinage phenomena on a smaller scale, were caused by extension and thinning during the F_3 doming event. Brittle fracturing of competent lithologies, such as the hornblende gneiss, is also ascribed to extensional thinning (Osmundsen et al., 2003).

Analytical methods

Alteration zones were identified based on mineralogy from macroscopic observations. The different zones were then cut out and analyzed microscopically. Bulk chemical analyses of major elements (Table 1) were done by wavelength dispersive XRF on a Bruker S8 Tiger, and trace elements (Table 2) by a combination of XRF and aqua regia ICP–MS, using an Elan DRC–II. Nickel, Cu, Zn, Rb, Sr and Zr were analyzed by XRF using the standardized Geoquant software package. The remaining trace elements in Table 2 were analyzed by ICP–MS. Electron microprobe spot analyses were performed using a JEOL JXA–8500F hyper probe with beam current and acceleration voltage set to 30 nA and 15 kV, respectively. During construction of element maps of decrepitated fluid inclusions, voltage was set to 10 kV. For the plagioclase-hornblende geothermometry, only spots with totals above 98.0% for plagioclase, and 96.0 mass% for amphibole were selected. All microprobe runs were calibrated using mineral standards from Astimex Scientific Ltd.

Microthermometry was performed using a Linkam THMSG 600 heating/freezing stage controlled by Linkam 32 software, and calibrated using synthetic fluid inclusions from Bubbles Inc. Isochores were calculated using the FLUIDS 1 package (Bakker, 2003) and equations of Bodnar & Vityk (1994).

Results

Mineralogy

Two distinct types of alteration can be observed in the hornblende schist, adjacent to the quartz veins: One dominated by chlorite, muscovite, and calcite (the chlorite-muscovite zone, CMZ), most prominent in the lower parts, and a visually more subtle zone, dominated by biotite, scapolite, and calcite (biotite-scapolite zone, BSZ).

Table 1. Major element compositions (mass%).

Sample	Unaltered	Bt-scp zone	Chl zone	Ms-cc zone
Fe ₂ O ₃	6.21	5.53	8.94	4.47
TiO ₂	0.60	0.65	1.04	0.37
CaO	6.18	2.60	3.65	5.31
K ₂ O	1.77	2.44	1.58	6.38
P ₂ O ₅	0.09	0.06	0.19	0.11
SiO ₂	60.37	72.85	60.92	56.76
Al ₂ O ₃	16.54	8.94	15.92	22.58
MgO	3.24	3.24	3.32	1.78
Na ₂ O	2.89	0.65	2.38	0.00
MnO	0.09	0.05	0.04	0.07
Sum	97.98	97.00	98.98	97.82
LOI	0.74	1.36	3.76	7.52

Table 2. Trace element compositions ($\mu\text{g/g}$).

Sample	Unaltered	Bt-scp zone	Chl zone	Ms-cc zone
Li	19.2	45.5	42.7	37.2
B	4.2	1.6	2.0	13.7
Sc	41.0	49.8	42.9	35.5
V	122.7	128.4	163.6	96.3
Cr	19.0	18.6	18.6	9.9
Ni	93.1	76.1	119.6	61.5
Co	15.7	8.4	12.9	4.2
Cu	4.4	8.4	2.6	1.8
Zn	64.7	78.0	75.2	53.3
Ga	18.0	16.1	16.4	31.7
Ge	<3	<3	<3	<3
As	2.6	<0.5	1.2	0.9
Se	<30	<30	<30	<30
Rb	16.1	31.0	20.2	46.4
Sr	131.6	53.4	112.5	17.2
Y	3.9	2.5	4.6	0.0
Zr	20.3	17.6	26.5	13.3
Nb	9.1	9.5	13.7	5.9
Mo	<0.4	<0.4	<0.4	<0.4
Ru	<3	<3	<3	<3
Rh	<0.01	<0.01	<0.01	<0.01
Ag	1.6	<0.8	<0.8	2.1
Cd	<2	<2	<2	<2
Sn	2.2	0.9	1.0	4.2
Sb	<2	<2	<2	<2
Te	<600	<600	<600	<600
Cs	0.4	3.3	3.3	3.0
Ba	184.9	219.4	118.4	379.0
La	4.8	4.6	7.9	0.0
Ce	33.9	22.7	35.9	9.1
Pr	1.5	1.4	2.3	0.0
Nd	<100	<100	<100	<100
Sm	<3	<3	<3	<3
Eu	<0.3	<0.3	<0.3	<0.3
Gd	<4	<4	<4	<4
Tb	<1	<1	<1	<1
Dy	<1	<1	<1	<1
Ho	<0.5	<0.5	<0.5	<0.5
Er	<2	<2	<2	<2
Tm	<1	<1	<1	<1
Yb	<3	<3	<3	<3
Lu	<1	<1	<1	<1
Hf	1.0	0.8	1.1	<0.7
Ta	0.8	0.7	1.2	0.4
W	<2	<2	<2	<2
Os	<2	<2	<2	<2
Ir	<0.1	<0.1	<0.1	<0.1
Pt	<8	<8	<8	<8
Au	<40	<40	<40	<40
Tl	0.2	0.8	0.8	0.8
Pb	168.4	121.9	0.0	0.0
Bi	<0.7	<0.7	<0.7	<0.7
Th	3.1	3.5	8.6	<0.6
U	2.2	<2	<2	<2

Figure 3. Example of the CMZ and the transition to unaltered hornblende schist between two quartz veins. The chlorite and muscovite-calcite subzones are indicated.

The extent of the alteration zones varies from a few mm to several cm (Fig. 3). Major and trace element compositions of the alteration zones are listed in Tables 1 and 2, respectively. All electron microprobe analyses are listed in Electronic Supplement 1.

Hydrothermally unaltered hornblende schist

The hydrothermally unaltered schist is composed mainly of plagioclase, green hornblende, brown biotite, and quartz, with minor K-feldspar and accessory titanite, clinozoisite, zircon, apatite, and calcite. Foliation is marked by 200 μm to 2 mm elongated, subhedral nematoblastic hornblende, mostly of edenitic to pargasitic composition (Fig. 4A), and lepidoblastic biotite grains. Reverse-zoned plagioclase (Fig. 5) is less clearly aligned. Antiperthite, possibly of replacement origin, may constitute up to 40% of the plagioclase grains as laths or well-defined patches, but is generally absent. Quartz occurs as 100–300 μm inclusions in plagioclase and less commonly in hornblende, but also as 200–600 μm grains in the matrix. Larger grains in the matrix show undulose extinction, whereas rounded inclusions appear undeformed. Myrmekite is common along grain boundaries with K-feldspar and larger antiperthite patches, with up to 20 μm -wide quartz vermicules. Inclusions of zircon, clinozoisite, and titanite are common in hornblende, and less common in plagioclase. Clinozoisite is zoned, with cores showing yellow birefringence and blue birefringent rims, and is commonly surrounded (more so than zircon) by pleochroic halos. Clinozoisite and titanite also occur as matrix minerals, with titanite oriented parallel to the foliation. A few biotite and plagioclase grains are situated inside the older quartz veins running parallel to the main foliation. This plagioclase is antiperthitic and weakly altered to sericite and coarser muscovite.

The first sign of retrograde alteration is the alteration of hornblende to biotite-quartz symplectites (Fig. 6A). K-feldspar disappears, and plagioclase is weakly altered to calcite. Calcite commonly has a dusty appearance

Figure 4. Classification of central minerals from EPMA analyses. (A) Amphibole compositions from the hornblende schist. White and grey circles are rim and core compositions used for P–T modeling. (B) Scapolite classification based on Cl content and anorthite equivalents. (C) Biotite compositions based on total Al and Fe/(Fe + Mg) ratio.

Figure 5. (A) Color-coded SEM picture of reverse-zoned plagioclase. (B) Same as (A) in cross-polarized transmitted light. The insert shows the An content of the plagioclase grain along the black line.

due to microscopic Fe-oxide inclusions. This alteration occurs furthest from the vertical quartz veins in both the BSZ and the CMZ. Some samples showed a depletion zone between the unaltered schist and the alteration zones (Fig. 6G, H). This zone lacks amphibole, but without extra biotite, and consists mostly of quartz and feldspar, and similar amounts of biotite as the protolith.

Biotite–scapolite zone

Near the quartz vein, in the schist, mica grains change orientation by 90° from schistosity-parallel to quartz vein-parallel. This area is dominated by biotite with a zone of scapolite bordering the vein. Biotite and scapo-

lite grains accumulate in separate distinctive zones but are intergrown at the contact between the two zones. Here, plagioclase is mostly unaltered, only weakly altered to calcite. However, there are rare examples of partial replacement of plagioclase by scapolite (Me_{53-59} ; Figs. 4B, 6B; Table 3).

Both the grain size and the mineral chemistry of the biotite in the alteration zone are similar to biotite in the unaltered schist, which plots in the phlogopite and annite areas in the biotite quadrilateral (Fig. 4C). Both biotite generations have low Cl contents (<0.06 apfu). In addition to biotite-quartz symplectites, amphibole is altered

Figure 6. (A) Biotite-quartz symplectites after breakdown of amphibole. Cross-polarized transmitted light. (B) Partial replacement of plagioclase to scapolite in the biotite-scapolite zone. Cross-polarized transmitted light. (C) Direct replacement of biotite by chlorite and ilmenite in the chlorite subzone. Plane-polarized transmitted light. (D) Partial replacement of biotite by chlorite, and titanite by ilmenite. Located in the transition zone between the unaltered schist and the chlorite subzone. Plane-polarized transmitted light. (E) Ilmenite overgrowths on rutile in the CMZ. Plane-polarized transmitted light. (F) Partial replacement of scapolite to muscovite, calcite, and quartz in the scapolite subzone of the BSZ. Cross-polarized transmitted light. (G) Depletion zone between the unaltered schist (left) and minor biotite-scapolite alteration (right; the quartz vein is far right). Plane-polarized transmitted light. (H) Same as (G) but with cross-polarized transmitted light.

Table 3. Average mineral compositions.

<i>n</i>	<i>Biotite</i> ^a 44	<i>Chlorite</i> ^b 20	<i>Muscovite</i> ^c 18	<i>Scapolite</i> ^d 16
SiO ₂	37.12 ± 1.32	23.99 ± 0.32	49.17 ± 0.32	51.22 ± 0.49
Al ₂ O ₃	16.90 ± 0.58	21.74 ± 0.35	33.13 ± 0.57	24.95 ± 0.28
Na ₂ O	0.13 ± 0.05	0.03 ± 0.04	0.49 ± 0.07	5.51 ± 0.34
MgO	10.90 ± 0.57	13.17 ± 0.29	1.11 ± 0.12	<0.02
K ₂ O	8.44 ± 0.37	<0.02	8.14 ± 0.27	0.33 ± 0.11
MnO	0.10 ± 0.07	0.03 ± 0.03	<0.02	<0.02
TiO ₂	2.43 ± 0.28	0.05 ± 0.04	0.22 ± 0.12	<0.02
CaO	0.04 ± 0.05	<0.02	0.02 ± 0.02	13.35 ± 0.46
FeO (tot)	18.80 ± 1.53	28.78 ± 0.61	1.87 ± 0.25	0.05 ± 0.03
BaO	0.11 ± 0.06	-	0.14 ± 0.06	-
Cl	0.27 ± 0.11	-	0.02 ± 0.03	1.18 ± 0.17
Total	95.35 ± 0.68	88.02 ± 0.90	94.35 ± 0.35	96.42 ± 0.34
Si	2.81 ± 0.07	2.58 ± 0.03	3.25 ± 0.02	7.72 ± 0.05
Al	1.51 ± 0.05	2.75 ± 0.05	2.58 ± 0.04	4.43 ± 0.06
Na	-	-	0.06 ± 0.01	1.61 ± 0.09
Mg	1.23 ± 0.06	2.11 ± 0.05	0.11 ± 0.01	-
K	0.81 ± 0.03	-	0.69 ± 0.02	0.06 ± 0.02
Mn	-	-	-	-
Ti	0.14 ± 0.02	-	-	-
Ca	-	-	-	2.16 ± 0.08
Fe (tot)	1.19 ± 0.11	2.58 ± 0.04	0.10 ± 0.01	-
Ba	0.00 ± 0.00	-	-	-
Cl	0.03 ± 0.01	-	-	0.30 ± 0.04
Fe/Fe+Mg	0.49 ± 0.03	0.55 ± 0.01		
Me				0.56 ± 0.02
An eq				0.48 ± 0.02

^a Calculated based on 11 O^b Calculated based on 14 O^c Calculated based on 11 O^d Calculated based on 16 cations

to a yellowish mineral. The alteration product was analyzed using three, large, defocused electron microprobe spots. They showed a mineral low in alkalis, and rich in Fe, Mg, Al, and Si. Totals were below 85%, but this may be an indication of a more complex reaction. Ilmenite is the dominating Ti-phase, and is most easily observed in areas between hornblende breakdown and change in biotite orientation.

Table 4. Summary of microthermometric results.

Associated alteration assemblage	Fluid type	Fluid components	Reaction	Average temperature (°C)
Chl + ms + cc	III	H ₂ O + NaCl ± CO ₂ ± CH ₄	Th (LV-L)	469
Chl + ms + cc	III	H ₂ O + NaCl ± CO ₂ ± CH ₄	Th (LV-L)	257
Chl + ms + cc	III	H ₂ O + NaCl ± CO ₂ ± CH ₄	Tm hal	284
Bt + scp	II	H ₂ O	Tm ice	-0.7
Bt + scp	II	H ₂ O	Th (LV-L)	325
Bt + scp	I	CO ₂	Th (LV-L)	21

Chlorite–muscovite–calcite zone (CMZ)

The BSZ is overprinted by a mineral assemblage consisting of chlorite, muscovite, and calcite. This zone can be divided into two subzones, with chlorite replacing the former biotite zone, and muscovite and calcite replacing the scapolite-dominated zone. The CMZ penetrates further into the hornblende schist than the BSZ, and additional alteration of the wall rock has occurred during this stage. Partial replacement of biotite by chlorite (Fig. 6C) can be observed together with the formation of ilmenite at the expense of biotite and titanite while coexisting with clinozoisite (Fig. 6D). Ilmenite-rutile intergrowths (Fig. 6C) are more common closer to the vein, where chlorite replacement is more advanced. Ti ± Fe-phases are more common in this zone than in the BSZ. Fe/(Fe + Mg) ratios in biotite (average 0.49) are slightly lower than that in replacement chlorite (average 0.55). Plagioclase is sericitized to a much greater degree compared to the BSZ. Partial replacement of scapolite can also be observed (Fig. 6F), but in most thin-sections scapolite is absent in the CMZ.

Fluid inclusions

Three types of primary fluid inclusions are observed: Liquid (L) CO₂ inclusions (type I); liquid-vapor (LV) H₂O inclusions (type II); and liquid-vapor-solid (LVS) NaCl–H₂O–CO₂ inclusions (type III). Effort was made to distinguish between apparently primary fluid inclusions in separate clusters from secondary fluid inclusions clearly associated with fractures, and microthermometric measurements were performed only on primary fluid inclusions. Microthermometric results are summarized in Table 4.

Types I and II

Scapolite hosts mainly type I inclusions, but a few type II inclusions were found to coexist with type I fluid inclusions (Fig. 7A). Both type I and type II occur commonly in quartz, and are generally small (<20 µm). Most type I inclusions are single-phase at room temperature, and homogenize into liquid at average 21°C. Type II fluid inclusions are two-phase inclusions at room temperature with 5–10 vol.% vapor (Fig. 7B). Average total homogenization (LV→L) for type II inclusions was 325°C. Freezing point depressions for type II inclusions were 0.5 to 1.0°C,

Figure 7. (A) Coexisting type I and II fluid inclusions in scapolite. (B) Coexisting type I and II fluid inclusions in quartz. (C) Type III inclusions in quartz.

yielding a salinity of 1–2 mass% NaCl equiv. using the equations from Bodnar (1993). Type II inclusions were measured in a quartz grain inside the chlorite zone free of type III inclusions.

Type III

Type III inclusions (Fig. 7C) are large (10–150 μm in the long direction), three phase (LVS) inclusions, only observed in quartz associated with chlorite-muscovite alteration. Multiple solid inclusions in addition to the halite crystal were observed, one of which was analyzed by Raman and verified as calcite. These solids vary in size and frequency, and are thus interpreted to be accidentally trapped minerals. Multiple melting and homogenization events occurred during heating from -195 to ca. 600° C, indicating a complex fluid composition. A solid, likely ice, appeared in and around the bubble between -91 and -88°C, gradually deforming the bubble to a maximum at -70°C. The solid phase gradually melted and disappeared entirely between -45 and -37°C. In a few samples a birefringent, second, solid phase nucleated around accidentally trapped minerals, and melted between -25 and -28°C. The halite crystal dissolved between 250 and 331°C, mostly between 270 and 305°C (Fig. 8B). Homogenization temperatures (LV→L) showed a bimodal distribution with one group homogenizing mainly from 465 to 495°C, and another group homogenizing before or at halite dissolution, mainly from 235 to 285°C. Several inclusions decrepitated prior to homogenization, generally from 350°C and up, and during analyses of specific fluid inclusions, groups of nearby inclusions were observed to homogenize from 420°C and up. These are not listed in the histogram, since precise homogenization temperatures are not known, and the high-temperature group is thus under-represented in the histogram (Fig. 8A). The birefringent, low-temperature solids were only observed in inclusions with high homogenization temperatures. Differences in halite dissolution could not be distinguished between the two groups, and $T_{m(\text{hal})}$ for both groups contributes to the peak in the histogram. Using the equation of Sterner et al. (1988), the dissolution of the halite crystal gives a salinity of 37.4 mass% NaCl equiv., calculated for the median $T_{m(\text{hal})}$ of 289°C in the H_2O -NaCl system. The slope of the isochore for the low-temperature type III inclusions

should be steeper in the liquid–halite (L–H) field than in the one-phase field (Bodnar, 1994), but as the salinity is fairly close to the pressure independent halite liquidus at 50 mass% NaCl (Bodnar, 1994), and $T_{h(\text{LV-L})}$ is within a few tens of degrees of $T_{m(\text{hal})}$, errors in isochore calculations for the group that homogenize by halite dissolution are small, and thus ignored. Combined measurements of both $T_{m(\text{ice})}$ and $T_{m(\text{hal})}$ are limited, but calculating the fluid composition for the range of $T_{m(\text{ice})}$ and median $T_{m(\text{hal})}$ in the H_2O -NaCl- CaCl_2 system (Steele-MacInnis et al., 2011) yields a total salinity of 41.9–43.1 mass% (NaCl + CaCl_2), and a molar Na/Ca ratio between 1.8 and 2.1.

In a few inclusions, CO_2 melting could be observed from -60.6 to -58.3°C. Liquid-vapor homogenization of the carbonic phase was not observed, and density cannot be calculated, but the observed melting temperature close to the triple point of CO_2 indicates a low X_{CH_4} , likely less than 0.2 (Thiery et al., 1994). Raman spectroscopic analyses of this generation of inclusions also indicated a minor CH_4 component, but greater than in mixed H_2O - CO_2 inclusions from the scheelite-bearing tremolite-diopside skarn in the W–Mo zone (Drivenes, 2010). Melting of CO_2 was observed only in the low- $T_{h(\text{LV-L})}$ inclusions.

Variation in the homogenization temperatures may result from stretching of the fluid inclusion during post-entrapment decompression (e.g., Bodnar et al., 1989). However, the strongly bimodal $T_{h(\text{LV-L})}$ distribution (Fig. 8A), uniform vapor volume, different CO_2 content, and proposed near isobaric cooling history (see section ‘Pressure and temperature constraints’) indicate that the high T_h fluids represent a fluid pulse infiltrating prior to the low T_h fluids.

All isochores were calculated for the system H_2O -NaCl, due to poorly constrained equations of state for gas-salt mixtures for salinities above the eutectic composition, and few observations of ice melting. Electron microprobe element maps of a selection of the decrepitated fluid inclusions showed elevated levels of Na, Ca, and Fe, with less K and Mn. (Fig. 9).

Figure 8. Histograms showing microthermometric analyses of type III fluid inclusions. (A) Liquid-vapor homogenization temperatures. (B) Halite dissolution temperatures.

Figure 9. Element maps of surface condensates of a decrepitated type III fluid inclusion determined by EPMA. The numbers on the right are counts detected by the microprobe in the element maps, and intensity levels in the secondary electron and backscatter images.

Mass exchange

Isocon plots (Fig. 10; Grant, 1986, 2005) were drawn for the BSZ and CMZ alteration assemblages vs. the original hornblende schist. The two alteration assemblages were also plotted against each other. Overlap between the zones during sample preparation was unavoidable. Quartz and Si are thus not regarded in zones bordering the quartz vein due to cutting separation issues. In the isocon diagrams, the traditionally hydrothermally

immobile element Zr (Finlow-Bates & Stumpfl, 1981) is used as an isocon. Petrographic evidence also suggests that zircon is preserved during alteration (Fig. 6C). When plotting the unaltered schist vs. the BSZ, the isocon intersected the Al point, indicating that Al was immobile during alteration. Since the muscovite-calcite and chlorite zones were analyzed separately, the combined CMZ was calculated under the presumption that Al also was immobile. That resulted in a mix consisting of ca. 2/3 of the chlorite sample, and ca. 1/3 of the

Figure 10. Isocon diagrams of the alteration zones plotted against the unaltered schist and each other, using Zr as isocon. (A–C) Major elements. (D–F) Trace elements.

muscovite-calcite sample. Only one sample of each zone was analyzed. Also, the extent of subzones within each alteration zone varies relative to each other (Fig. 3). This introduces further uncertainties in that the difference in chemical composition between the minerals within each zone will have an effect on the isocon plots. The results are thus used as indications for mass transfer, rather than statistical, quantitative implications for the complete system. The effects of variable mineral ratios are discussed in the subchapters dealing with the respective alteration assemblages. Both alteration assemblages are enriched in K, and depleted in Na and Ca compared to the unaltered schist. The BSZ is also enriched in Mg, Fe, and Ba, whereas the CMZ is depleted in Mn.

Plagioclase-hornblende geothermometry

Six pairs of adjacent hornblende and plagioclase grains were analyzed and used for plagioclase-hornblende geothermobarometry (Tables 5, 6). Traverses of point analyses with step size 10 μm throughout the zoned plagioclase showed a distinct chemical zoning, correlated to the variations in extinction angle. Typical An contents in the rims vary from 0.45 to 0.47, with core values from 0.37 to 0.43 (Fig. 5B; Table 5; Electronic Supplement 1). Orthoclase contents are less than 0.015 in the cores, and

slightly lower in the rims. Hornblende is less drastically zoned, with ΣAl values from 2.01 to 2.14 apfu in the rims, and 2.29 to 2.36 in the cores (Table 6). Temperatures were estimated using the amphibole-plagioclase geothermometer (Blundy & Holland, 1990; Holland & Blundy, 1994), and pressure was calculated using Al-in-hornblende geobarometry (Johnson & Rutherford, 1989). Combined, the thermobarometric calculations yielded P–T intervals from 678 to 708°C and 530 to 580 MPa in the rims, and 680 to 704°C and 640 to 670 MPa in the cores (Fig. 11).

Discussion

Metamorphic and metasomatic reactions

Biotite-scapolite zone

Based on petrographic observations, the replacement of calcic amphibole is controlled by the formation of new biotite, quartz, and minor calcite. In order to preserve stoichiometric balance, and given the limited amount of K-feldspar available to react, K⁺ has to be added. This can, to a certain degree, be canceled out by taking the alkali-depleted alteration product into account, assigning some of the Fe and Mg to that, and thus minimize the required K⁺. There are no Na-bearing alteration products

Figure 11. Summary of P - T estimates. The titanite-out line is from Spear (1981) at the QFM buffer. Upper stability of muscovite + calcite + quartz is extrapolated from data of Hewitt (1973) for $X_{\text{CO}_2} = 0.1$. Isochores for type I and II fluid inclusions are calculated using the BULK and ISOC programs of Bakker (2003), and type III are calculated from equations of Bodnar & Vityk (1994) for a H_2O -NaCl fluid with 40 mass% NaCl. Minimum and maximum temperature of chlorite is calculated using the thermometer of Cathelineau (1988).

directly replacing amphibole. Scapolite is the only major host for Na, but the extent of scapolite is not enough to accommodate the amount of Na^+ released at amphibole decomposition. Also, alteration of amphibole is observed several cm into the host schist, while scapolite is only recorded bordering the quartz vein, and a direct replacement of amphibole by scapolite is not observed. Newly formed albite was not observed, and the loss in Na_2O in the BSZ (Fig. 10A) indicates that Na was extracted into the fluid phase. Given the phases observed in the unaltered protolith, the reaction may have been:

Scapolite has replaced plagioclase close to the quartz vein. Meionite 0.53–0.59 corresponds to anorthite equivalents (An_{eq}) of 0.45–0.50, coinciding roughly with the rim values of the original plagioclase, and serves as additional evidence for a direct replacement. Biotite is the main Cl-bearing phase in the original rock, with much higher levels than amphibole, and since the original biotite coexists with scapolite, Cl in scapolite (~ 0.3 apfu) has to originate from the fluid. Only minor calcite is observed, and it occurs only in areas where amphibole is altered. No carbonates are

thus a major part of the protolith, and the CO_3^{2-} -component in scapolite is likely derived from the fluid. A possible reaction for the replacement could be as follows:

Scapolite composition is largely related to the infiltrating fluid (e.g., Orville, 1975; Ellis, 1978; Aitken, 1983), and although not completely carbonated, these scapolites must have been formed in contact with a low-salinity, CO_2 -dominated fluid.

Ilmenite coexists with Ti-bearing biotite, and examples of partial replacement of titanite by ilmenite can be observed (Fig. 6D). Rutile is scarce in samples which have not equilibrated with the high-salinity fluid, and commonly occurs in the zone between the chlorite zone and the unaltered zone. Titanite is the only Ti-phase that reacts out, and assuming Fe is available from amphibole breakdown, the main Ti reaction is:

Frost et al. (2001) suggested the breakdown of titanite, combined with amphibole dehydration, formed ilmen-

Table 5. Plagioclase compositions used for thermobarometric calculations.

Grain	Rim						Core					
	1	2	3	4	5	6	1	2	3	4	5	6
SiO ₂	57.25	58.17	56.41	57.93	56.86	56.83	57.25	58.17	56.41	57.93	56.86	56.83
Al ₂ O ₃	27.02	27.12	26.89	27.19	26.67	26.70	27.02	27.12	26.89	27.19	26.67	26.70
FeO	0.05	0.09	0.14	0.08	0.08	0.06	0.05	0.09	0.14	0.08	0.08	0.06
CaO	9.33	9.45	9.48	9.54	9.14	9.25	9.33	9.45	9.48	9.54	9.14	9.25
Na ₂ O	5.77	5.93	5.88	5.89	5.92	5.85	5.77	5.93	5.88	5.89	5.92	5.85
K ₂ O	0.13	0.19	0.16	0.16	0.16	0.14	0.13	0.19	0.16	0.16	0.16	0.14
Sum	99.56	100.96	98.96	100.78	98.85	98.83	99.56	100.96	98.96	100.78	98.85	98.83
Si	2.57	2.58	2.56	2.57	2.58	2.57	2.57	2.58	2.56	2.57	2.58	2.57
Al	1.43	1.42	1.44	1.42	1.42	1.43	1.43	1.42	1.44	1.42	1.42	1.43
Fe (tot)	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00
Ca	0.45	0.45	0.46	0.45	0.44	0.45	0.45	0.45	0.46	0.45	0.44	0.45
Na	0.50	0.51	0.52	0.51	0.52	0.51	0.50	0.51	0.52	0.51	0.52	0.51
K	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01
An	0.468	0.463	0.467	0.468	0.456	0.463	0.468	0.463	0.467	0.468	0.456	0.463
Ab	0.524	0.526	0.524	0.523	0.534	0.529	0.524	0.526	0.524	0.523	0.534	0.529
Or	0.008	0.011	0.009	0.009	0.010	0.009	0.008	0.011	0.009	0.009	0.010	0.009

Structural formula calculated based on 8 O

Table 6. Amphibole compositions used for thermobarometric calculations.

Grain	Rim						Core					
	1	2	3	4	5	6	1	2	3	4	5	6
SiO ₂	43.52	42.91	42.95	43.13	43.35	43.63	42.25	41.95	42.08	41.66	42.19	41.60
TiO ₂	0.58	0.60	0.63	0.66	1.20	0.60	0.74	0.71	0.75	0.78	0.74	0.72
Al ₂ O ₃	11.46	11.84	12.03	12.11	11.84	11.36	12.90	13.13	13.03	13.21	13.10	13.22
FeO	18.03	18.49	18.32	18.24	18.18	18.05	18.44	18.53	18.61	18.70	18.54	18.33
MnO	0.35	0.37	0.37	0.37	0.36	0.37	0.37	0.37	0.35	0.36	0.36	0.36
MgO	9.18	9.22	8.83	8.95	9.30	9.22	8.51	8.60	8.45	8.32	8.65	8.54
CaO	11.88	11.67	12.09	11.82	11.58	11.83	12.04	11.97	12.08	11.92	11.88	11.69
Na ₂ O	0.97	1.01	0.91	1.00	0.84	0.93	1.00	1.19	0.96	1.05	1.17	1.10
K ₂ O	1.48	1.50	1.50	1.56	1.41	1.41	1.76	1.84	1.78	1.90	1.84	1.85
Cl	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.07	0.00
Sum	97.45	97.62	97.64	97.86	98.06	97.41	98.02	98.29	98.10	97.91	98.54	97.41
Si	6.54	6.44	6.46	6.46	6.46	6.55	6.35	6.30	6.33	6.29	6.32	6.29
Al ^{IV}	1.46	1.56	1.54	1.54	1.54	1.45	1.65	1.70	1.67	1.71	1.68	1.71
Al ^{VI}	0.57	0.54	0.60	0.60	0.54	0.56	0.64	0.62	0.64	0.64	0.63	0.65
Ti	0.07	0.07	0.07	0.07	0.13	0.07	0.08	0.08	0.08	0.09	0.08	0.08
Fe ³⁺	0.36	0.48	0.35	0.38	0.42	0.38	0.34	0.36	0.35	0.36	0.38	0.40
Fe ²⁺	1.90	1.85	1.96	1.90	1.84	1.88	1.98	1.97	1.99	2.00	1.95	1.91
Mn	0.05	0.05	0.05	0.05	0.04	0.05	0.05	0.05	0.04	0.05	0.05	0.05
Mg	2.06	2.06	1.98	2.00	2.07	2.06	1.91	1.92	1.89	1.87	1.93	1.92
Ca	1.91	1.88	1.95	1.90	1.85	1.90	1.94	1.93	1.95	1.93	1.91	1.89
Na	0.28	0.29	0.27	0.29	0.24	0.27	0.29	0.35	0.28	0.31	0.34	0.32
K	0.28	0.29	0.29	0.30	0.27	0.27	0.34	0.35	0.34	0.37	0.35	0.36

Structural formula calculated on basis of 23 O

ite and anorthite with increasing temperature in metabasites. In our samples, the titanite-out reaction occurs simultaneously with a hydration reaction of hornblende to biotite (Fig. 6D). It is more likely that ilmenite formed by a drop in pressure or reduction by fluid infiltration (Spear, 1981), or in fact, a combination of the two. However, a potential increase in temperature during ilmenite formation cannot be ruled out.

The isocon analysis for the BSZ shows a depletion in Na and Ca, compared to the original rock (Fig. 10A). Loss of Ca and Na from amphibole breakdown may be the cause of this depletion. It is unlikely that the fluid responsible for the scapolitization was a pure CO_2 - CH_4 fluid (Larsen, 1991), since mobilization of Na and Ca requires an aqueous component (e.g., Bai & Koster van Groos, 1999). The increase in K, Mg, and Fe in this zone in the isocon plot may be a result of the specific sample having less than representative amounts of scapolite. The ratio between the biotite and scapolite contents is highly variable, and since K, Mg and Fe are major components in biotite, and only occur as trace concentrations in scapolite, the increase in these elements may be artificially high in the isocon plot. Likewise, Na and Ca are major components in scapolite and trace components in biotite (Electronic Supplement 1), and would be closer to the isocon if the sample had more scapolite.

Chlorite-muscovite zone

In some samples, a gradual transition from the BSZ to the CMZ was observed. Here, chlorite replaces biotite, while scapolite is still stable (Fig. 12). This indicates that the overprinting has happened in variable degrees as a result of differing fluid/rock ratios. The remaining titanite, although restricted, is also altered to ilmenite during this fluid pulse. Ilmenite intergrowths with rutile are common, and rutile may also be an alteration product of ilmenite, possibly the result of Fe-leaching during the later stages. When the CMZ is plotted against the BSZ in the isocon diagram (Fig. 10C, F), the only significant change is a depletion in Fe and Mg. The uncertainties regarding the representability of the BSZ sample discussed above also apply here, and Fe, Mg, and K may be over-represented in the BSZ, while Na and Ca are under-represented, and the depletion of Mg and Fe in the CMZ relative to the BSZ may be exaggerated. Likewise, K and Ca may be enriched in the CMZ relative to the BSZ. The petrographic observations (Fig. 6) indicate a two-step alteration, where biotite is altered to chlorite, and scapolite is altered to muscovite, calcite, and quartz. The $\text{Fe}/(\text{Fe} + \text{Mg})$ ratio for biotite and chlorite is similar, and the main resultant ion of the reaction is K^+ .

Closest to the vein, the replacement of scapolite by muscovite, calcite, and quartz requires K^+ and produces Na^+ and Cl^- after the reaction:

Seen as a whole, the overprinting of the BSZ can be written as:

Whether K^+ is required in equation 6 depends on the ratio of K-bearing alteration products (i.e., biotite and muscovite). However, plagioclase in the CMZ is more sericitized than in the BSZ, and it is our interpretation that K^+ was added during this fluid pulse. The net element budget for the complete metasomatic event is represented by the protolith vs. CMZ plot (Fig. 10B, E). The only element that is added during fluid infiltration is K, and the protolith has thus experienced K-metasomatism. The CMZ extends further into the hornblende schist than the observed relics of the BSZ, and the bulk equation for the last metasomatic stage may have been:

The element exchange between the different zones is summarized in Fig. 12. As seen from the isocon diagrams, Na, Ca, K, and Mn have been mobile elements in the system, as expected from their elevated levels in the fluid inclusions (Fig. 9). Additionally, there was a significant proportion of Fe in the type III fluids, but Fe did not increase in the altered rock.

Fluid evolution and source

All observed mineral reactions are fluid-driven, either as hydration, carbonation or other volatilization events. As a result, the newly formed mineral assemblages depend on the availability of volatiles. The close relationship between different fluid inclusions and the two different alteration zones implies that two separate fluid pulses interacted with the rock at different times. The lower-temperature mineral assemblage in the CMZ, its greater extent in the lower parts compared with the upper parts of the fracture system, and textural overprinting places this alteration later on the timeline. The close affiliation of high-salinity type III fluid inclusions to the CMZ, and the lack of these fluids in the BSZ, where only types I and II fluid inclusions occur, implies that alteration to the BSZ served as a preparatory stage for the main metasomatic event of the CMZ observed in the field.

Figure 12. Overview of the metasomatic evolution during fluid influx.

For types I and II fluids to exist as separate inclusions, trapping must have occurred in the two-phase field. The intersection between the two isochores is in the one-phase field of the CO₂-H₂O system, but the addition of other volatiles will greatly expand the immiscibility field (Diamond, 2003). However, it cannot be ruled out that the two inclusion types represent different fluid pulses. Type III inclusions, given their uniform volumetric liquid and vapor distribution, were trapped in the one-phase field. In addition, the fluid inclusions that homogenized by halite dissolution, cannot have been trapped in equilibrium with a vapor phase (Bodnar, 1994), excluding the evolution of a moderately saline fluid to a high-salinity

fluid through phase separation (e.g., Burnham, 1979; Hedenquist & Lowenstern, 1994; Ulrich et al., 2001).

Trapping of fluid inclusions that homogenize by halite dissolution and LV-L may be due to a rapid drop in temperature after fluid infiltration during near isobaric conditions. Systems with fluid populations that homogenize in similar fashion as in our study have been described from magmatic-hydrothermal ore deposits, such as the Bismark Pb-Zn skarn deposit, Mexico (Baker & Lang, 2003). A hot fluid pumped into a relatively cool host rock will lead to rapid cooling of the fluid system, given that it is not a long-lived system. This is implied from

the hydrothermal overprinting being restricted to the lower parts of the fracture system, and only penetrating a few cm into the hornblende schist. The trapping conditions of types I and II inclusions are 600°C and 240 MPa, whereas the minimum pressure and temperature for trapping of low-temperature type III inclusions are ca. 50 MPa and 290°C, where the isochore intersects the halite liquidus. This gives a gentle slope between the two events in the P–T diagram, especially if the type III fluid inclusions with low homogenization temperatures were trapped further up along the isochore.

The oxidation state of the low-salinity fluid is probably close to the QFM buffer, since the titanite-out curve in P–T space (Fig. 11) for the QFM buffer goes through the intersection of the isochores of the types I and II fluids, and ilmenite is stable instead of hematite.

The pH of a fluid in the H₂O–CO₂–NaCl system decreases with increasing pressure and salinity, and increases with increasing temperature (Li & Duan, 2007). Also, the high-salinity fluid was apparently in equilibrium with muscovite instead of K-feldspar, indicating a low to moderate a_{K^+}/a_{H^+} ratio (Bowers et al., 1984). The K content of the fluid was significant (Fig. 9), giving a low pH. If a hot, acidic, and reactive fluid, such as the type III fluid, was to be transported over a great distance, it probably would have been buffered by the host rocks, including marble and other carbonate-bearing rocks, thus losing much of its metasomatic potential. Alternatively, it could also have been transported from the source through a refractory channel. It is more likely that the fluid source was relatively local and that the fluids were far from equilibrium with the host. High-salinity fluids have not been observed in any other lithology in the metasedimentary stratigraphy (Larsen, 1988, 1991), and this fluid pulse was not pervasive, unlike CO₂–H₂O fluids, which are found as inclusions in most of the studied rocks from the area (Larsen, 1988, 1991; Drivenes, 2010). The sheared appearance of the lower part of the vein system indicates a tectonic contact, and the high-salinity fluids may have flowed through the shear zone from the source, and reused the already existing vein set. Alternatively, the hornblende schist was thrust onto the underlying garnet-mica schist after fluids had altered the hornblende schist.

Metamorphic fluids with salinities resembling those of the type III fluids may form from evaporites (Yardley, 2013), but they are also commonly described as originating from S-type, granitic magmas (e.g., Lowenstern, 1994; Sirbescu & Nabelek, 2003; Drivenes et al., 2016). Evaporites are not common in the Norwegian Caledonides, although meta-evaporitic magnesite and scapolite deposits have been described from the Skipa formation in the Swedish part of the Seve Nappe (Svenningsen, 1994). Also, if evaporates had been deposited along with the abundant carbonate bodies on the Laurentian continental margin, evidence of high-salinity fluids should be

much more widespread than what has been observed. A possible source of the type III fluids observed could be a granitic body formed by decompression melting during rapid exhumation. Further rise of the granitic body would cause contact-metamorphic dehydration and decarbonation of the surrounding metasedimentary rocks and carbonates, forming the type I and type II fluids. A high-salinity fluid may later have been exsolved from the crystallizing magma to form the type III fluids. The CO₂-dominated fluids responsible for the BSZ are not likely to have exsolved directly from an underlying magmatic source due to the low solubility of CO₂ in felsic melts at pressures below 2 GPa (e.g., Fogel & Rutherford, 1990; Blank et al., 1993; Guillot & Sator, 2011). On the contrary, CO₂-rich fluids are common in high-grade metamorphic rocks (Touret & Nijland, 2013), and the multiple marble and carbonate-bearing bodies in the area could be likely sources of CO₂. Also, fluids similar to those of types I and II, are common in the retrograde alteration of the surrounding metasedimentary rocks, and a magmatic body is not a prerequisite for these fluids. No high-salinity fluid inclusions have been observed in the pegmatite intruding the metasedimentary sequence, or in the quartz veins closely associated with the pegmatite, thus rendering the pegmatite an unlikely source. The low Cl content of the hornblende schist makes it unlikely for an originally low-salinity fluid to have reached the salinities of the type III fluids through metamorphic reactions. The fluid may have increased in salinity due to water consumption into the hydrous minerals, but we argue that the initial type III fluid was Cl-rich prior to infiltration.

Pressure and temperature constraints

The highest recorded pressure and temperature in the rock is calculated from the cores of hornblende and plagioclase. Typical high-temperature minerals, such as pyroxene and wollastonite, are not observed. Hence, the temperatures of ca. 700°C from plagioclase-hornblende thermometry are interpreted to represent the peak temperature of the system during regional metamorphism. The highest pressures are calculated from Al-in-hornblende barometry (Fig. 11), indicating a near-isothermal pressure drop, and thus a rapid uplift in the early retrograde stage.

End-member meionite is stable above 875°C (Goldsmith & Newton, 1977), while Me₈₃ (Aitken, 1983) and Me₇₅ (Oterdoom & Gunter, 1983), both CO₃-scapolites, have been determined to be stable for T > 625 and 550°C, respectively, in contact with CO₂-rich fluids. For Cl-bearing scapolites there is a general decrease in X_{Cl} and increase in An_{eq} with increasing metamorphic grade, with local variations due to fluid composition (Mora & Valley, 1989). The lowest-temperature scapolites form around 400°C, and are the Al–Si ordered with an An_{eq} of 33 (Oterdoom & Wenk, 1983). At the time of scapolitization,

titanite was altered to ilmenite, restricting the event to high temperatures (Fig. 11). Assuming fO_2 was close to the QFM buffer, the intersection between the type I and II isochores and the titanite-out curve, at ca. 600°C and 240 MPa, gives us the temperature and pressure for the first fluid infiltration and scapolitization. Similar temperature and pressure estimates can also be achieved when assuming that the types I and II inclusions did not form during immiscibility. Both types predate the high salinity pulse. The high homogenization temperatures for both type I and type II inclusions require a high trapping temperature (type II), and elevated trapping pressure (type I), given trapping conditions at higher P–T than the formation of the CMZ.

The temperature estimates are well within the stability field of our analyzed scapolite, and in agreement temperature-wise with scapolites of similar composition from the kyanite-staurolite zone and kyanite-sillimanite zone of the Wallace formation, USA (Mora & Valley, 1989), but at lower pressure in our example. Other examples of high-temperature scapolitization include Mary Kathleen, Mt Isa, Australia (Oliver et al., 1994) and the Bamble sector, Norway (Engvik et al., 2011). However, scapolite in both examples is considerably more marialitic than scapolites from Bjellatinden. These temperature and pressure estimates are also in agreement with the conclusions of Larsen (1991) for scapolite formation in a scheelite-bearing scapolite skarn.

Between the P–T points from plagioclase-hornblende thermobarometry and the scapolitization, no additional firm pressure or temperature constraints can be made. There are two possible paths to explain the uplift history: a near-isothermal pressure drop during rapid uplift and crustal thinning, or uplift along a less steep thermal gradient and reheating by a magmatic intrusion. In the first scenario, thermal lag during rapid exhumation would produce a steep P–T path during the early stage followed by a gently sloping cooling path. In the case of the second scenario, a magmatic heat source will create a significant isobaric temperature increase. Without an infiltrating fluid, the amphibolitic assemblage could have persisted well into greenschist facies before being reheated on a local scale by hot fluids causing scapolitization. A granitic body has not been observed, nor even a contact-metamorphic aureole. However, as discussed above, a magmatic intrusion is the most likely source for the high-salinity fluids, and an intrusion may also explain the P–T path during the late stages of the metamorphic history. The pegmatites in the area are rich in tourmaline, and given that the proposed underlying granitic magma had a similar composition, it could also have been B-rich. Boron suppresses the liquidus significantly, and a completely molten magma may not have been hotter than 750–800°C (Pichavant, 1987). Heat production from such a system would have been sufficient to induce both the scapolitization and the high-temperature type III fluids. Typical retrograde, post-intrusion P–T paths

are (near) isobaric cooling (Spear, 1995), similar to our path for Bjellatinden.

The mineral assemblage observed in the CMZ is not stable at the high temperatures of the high-temperature type III fluid group. At 200 MPa, the thermal maximum stability of muscovite + calcite + quartz is 475°C in contact with a CO_2 - H_2O fluid with X_{CO_2} of 0.5 (Hewitt, 1973). X_{CO_2} of the high-salinity fluid is low, and the muscovite alteration occurred at lower temperatures. Extrapolation of the muscovite-calcite-quartz line to $X_{CO_2} = 0.1$ gives temperatures of around 380 and 425°C for 100 and 200 MPa, respectively. Tetrahedral-Al-in-chlorite thermometry (Cathelineau, 1988) gave temperatures from 370 to 410°C, and although criticized (e.g., de Caritat et al., 1993), it may give an indication of the formation temperature for the CMZ. Assuming a maximum pressure of 240 MPa, and using the chlorite temperatures, muscovite-calcite-quartz stability line and the isochore for the low-temperature type III fluids as boundaries, a limited window from 370°C and 75 MPa to 410°C and 240 MPa can be drawn for the formation of the CMZ.

Implications for W–Mo mineralizations

The model with an underlying granitic magma solves two major problems: The source of high-salinity fluids and the source of tungsten. The W content of the metasedimentary rocks is insignificant (Larsen, 1988), and these rocks are unlikely to have been leached of enough W to form the mineralizations in the area. As discussed earlier, the high-salinity type III fluids are also most likely magmatic in origin.

A granitic intrusion may have driven the hydrothermal convection system which eventually formed the mineralizations. Devolatilization of carbonates and pelites during an intrusive stage formed the metamorphic, low-salinity, CO_2 - H_2O fluids. These fluids interacted with the crystallizing granitoid intrusion, scavenging W and Mo, which subsequently allowed for the formation of the W (–Mo) veins and replacement mineralizations. When the magma reached water saturation, the exsolved, high-salinity fluids migrated through existing fractures and shear zones, and eventually overprinted the pre-existing mineral assemblage. The vein system in this study was barren, and occurred stratigraphically above the mineralized layers.

The sheared appearance of the lower parts of the quartz veins indicate significant shearing. The shear zone may have accommodated greater fluid flow compared to lithological and structural boundaries in the underlying units, which may explain why late high-salinity fluids are only observed in this particular zone. The main W mineralizations are strongly associated with marble units in the W–Mo zone. CO_2 inclusions in scapolite, related to the scheelite-mineralized scapolite skarn in

the richest mineralized zone, have similar T_h as the type I inclusions in our study (Larsen, 1991). However, the mineralized zones are associated with the tapering and disappearance of marble bodies. The stability of scheelite was controlled by the availability of Ca^{2+} and pH (Wesolowski, 1984). The lack of Ca-carbonate bodies as a source of Ca^{2+} and pH buffering during breakdown, may have restricted scheelite precipitation in the studied lithology. Fluid inclusions associated with scheelite mineralization in the underlying diopside skarn body are mixed H_2O - CO_2 fluids (Drivenes, 2010). These fluids are not observed in the vein system, and a clear relationship between the ore-forming event and the fluids observed in the quartz veins cannot be established. The source of W for the W-mineralized diopside and garnet-diopside skarn from Målvika, Nordland, has been interpreted to be the W-enriched granites of the Vistindan massif up to 1 km away (Müller & Furuhaug, 2008), implying that long-distance transportation of ore-bearing fluids in similar environments is not unique.

Conclusions

This study documents reactions during fluid-rock interaction in a hornblende schist where two fluid pulses have altered and metasomatized the original rock at different stages during amphibolite- to greenschist-facies metamorphism. The main aspects of the study can be summarized as:

1. Low-salinity, CO_2 - H_2O fluids responsible for the breakdown of hornblende to biotite-quartz symplectites, the alteration of titanite to ilmenite, and of plagioclase to scapolite (Me_{53-59}), formed distinct biotite and scapolite zones. Alteration occurred at ca. 600°C and 240 MPa. Only minor element exchange occurred during this alteration.
2. High-salinity fluids overprinted the former alteration assemblage and extended further into the host rock, resulting in bulk K-metasomatism. Biotite is replaced by chlorite and ilmenite, and scapolite is altered to muscovite, calcite, and quartz. This alteration happened during cooling somewhere between 370°C; 75 MPa, and 410°C; 240 MPa.
3. The low-salinity fluids are of metamorphic origin, and are a part of the regional retrograde stage, whereas the high-salinity fluids were exsolved from an underlying S-type granitic magma and transported through local shear zones. These overprinted the existing alteration assemblage using the same vein system.

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