Halogen contents of eclogite facies fluid inclusions and minerals: Caledonides, western Norway

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ABSTRACT   Primary multiphase brine fluid inclusions in omphacite and garnet from low- to medium-temperature eclogites have been analysed for Cl, Br, I, F, Li and SO_4. Halogen contents and ratios provide information about trapped lower crustal fluids, even though the major element (Na, K, Ca) contents of inclusion fluids have been modified by fluid–mineral interactions and (step-) daughter-crystal formation after trapping. Halogens in the inclusion fluids were analysed with crush–leach techniques. Cl/Br and Cl/I mass ratios of eclogite fluids are in the range 31–395 and 5000–33 000, respectively. Most fluids have a Cl/Br ratio lower than modern seawater and a Cl/I ratio one order of magnitude lower than modern seawater. Fluids with the lowest Cl/Br and highest Cl/I ratios come from an eclogite that formed by hydration of granulite facies rocks, and may indicate that Br and I are fractionated into hydrous minerals. Reconstructions indicate that the inclusion fluids originally contained 500–4000 ppm Br, 1–14 ppm I and 33–438 ppm Li. Electron microprobe analyses of eclogite facies amphibole, biotite, phengite and apatite indicate that F and Cl fractionate most strongly between phengite (F/Cl mass ratio of 1469 ± 1048) and fluid (F/Cl mass ratio of 0.008), and the least between amphibole and fluid. The chemical evolution of Cl and Br in pore fluids during hydration reactions is in many ways analogous to Cl and Br in seawater during evaporation: the Cl/Br ratio remains constant until the a_{H_2O} value is sufficiently lowered for Cl to be removed from solution by incorporation into hydrous minerals.

Key words: dehydration; eclogite; fluid inclusions; halogen fractionation; halogens; hydration.

INTRODUCTION

Primary brine fluid inclusions in omphacite from eclogites are direct evidence for the presence of a free H_2O-rich fluid phase during deep crustal metamorphism (Philippot & Selverstone, 1991; Selverstone *et al.*, 1992; Andersen *et al.*, 1993; Nadeau *et al.*, 1993; Giaramita & Sorensen, 1994; Vallis & Scambelluri, 1996; Philippot *et al.*, 1998; Scambelluri *et al.*, 1998; Svensen *et al.*, 1999). Omphacite-hosted fluids may provide valuable information about the compositions of fluids in high *P–T* metamorphic systems. However, most of these inclusion fluids have been studied by microthermometry, which only yields estimates of Na–Ca–K–Mg–Cl contents. Because many of these omphacite-hosted fluid inclusions also contain multiple solid phases, the composition of the residual fluids must differ from that of the high-*P–T* fluid. Finally, the fluid chemistry and the composition of the solid phases may have been significantly affected by interactions with the host omphacite during retrograde conditions. Crush–leach analysis of the halogens (Cl, Br, I) has provided information about fluid sources and fluid–rock interactions of eclogites from the Norwegian Caledonides (Cl–Br data; Svensen *et al.*, 1999) and the Alps (Cl–Br–I data; Yardley, 1997). However, the use of halogen data for tracing fluid salinity in high-grade systems is complicated by the incorporation of halogens into minerals. Mineral chemistry studies that focus on halogens exist for amphibolite and granulite facies (e.g. Nijland *et al.*, 1993; Kullerud, 1995, 1996; Léger *et al.*, 1996; Markl *et al.*, 1998) and coesite-bearing rocks (Philippot *et al.*, 1995), but there have been no empirical studies on how halogens are partitioned between fluid and halogen-bearing, high-grade minerals.

Eclogite formation as a result of infiltration of fluids into dry rocks (hydration) is well documented from the Norwegian Caledonides (e.g. Austrheim & Griffin, 1985; Austrheim, 1986; Jamtveit *et al.*, 1990, 2000; Andersen *et al.*, 1993; Austrheim & Engvik, 1997; Austrheim *et al.*, 1997), and emphasizes the importance of fluids in catalysing metamorphic transformations. In addition, field relations indicate a mobility of fluids on scales larger than the size of the eclogite precursors (i.e. kilometre scale). Hydration reactions may lower the a_{H2O} value of the fluid phase significantly, resulting in the incorporation of Cl into minerals (e.g. Sanford, 1981), a lowering of the fluid Cl/Br ratio and enrichment of incompatible elements in the fluid (e.g. Svensen *et al.*, 1999). In contrast, eclogites also form during prograde metamorphism and devolatilization

with a small-scale mobility of fluids (e.g. Philippot & Selverstone, 1991; Selverstone *et al.*, 1992), where the fluid composition is controlled by *P–T* conditions and mineral compositions.

The aim of this study is to analyse omphacite-hosted, brine fluid inclusions from eclogites in western Norway, especially with regard to their halogen systematics. Both microthermometry and crush–leach analyses (liquid ion chromatography and flame emission spectrometry) constrain fluid inclusion composition. In addition, the Cl and F contents of amphibole, biotite, phengite and apatite have been measured for samples from the Bårdsholmen locality. The halogen contents of eclogite facies fluid inclusions and minerals at Bårdsholmen illustrate the effects of high *P–T* fluid–mineral interactions on the compositions of high-pressure fluids.

GEOLOGICAL SETTING

In the Western Gneiss Region (WGR) of south Norway (Fig. 1), continental rocks underwent high-pressure metamorphism during the Caledonian orogeny and at least locally reached the stability field of coesite (e.g. Smith, 1984; Wain, 1997). Eclogite facies rocks occur throughout the WGR, and within allochthonous units of the Bergen arc system (BAS).
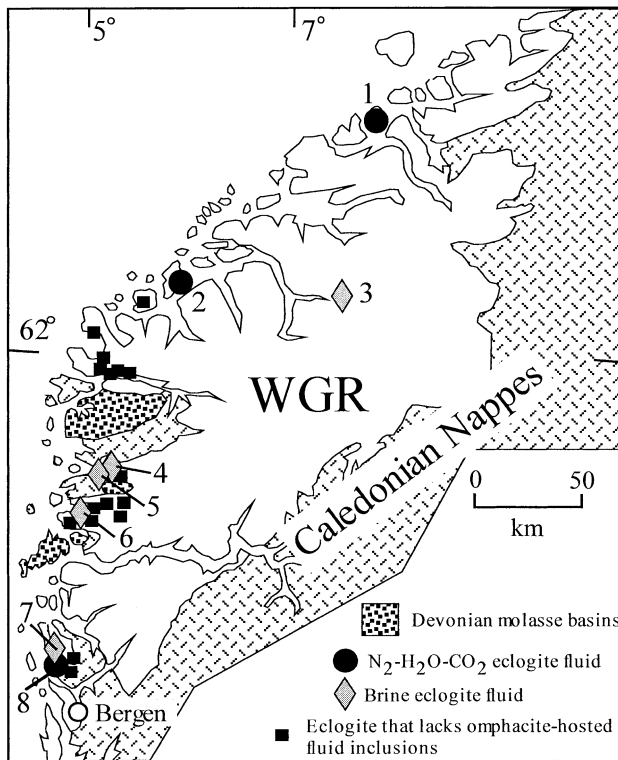

Fig. 1. Simplified geological map of western Norway. Fluid inclusion studies of eclogites from localities 1, 2, 3 and 8 were reported by Andersen *et al.* (1993), and localities 4–7 are described in this paper. WGR, Western Gneiss Region.

They formed by prograde metamorphism of amphibolite facies protoliths (e.g. Krogh, 1982; Jamtveit, 1987) or by hydration of Precambrian granulites or igneous rocks under eclogite facies *P–T* conditions (e.g. Austrheim & Griffin, 1985; Jamtveit *et al.*, 1990; Austrheim & Engvik, 1997; Engvik *et al.*, 2000). We studied eclogites from three localities in the WGR (Bårdsholmen, Engebøfjellet and Leknes) and one locality in the BAS (Sjurdholmen, Holsnøy). All underwent regional, low- to medium-temperature (500–650 °C) eclogite facies metamorphism. The localities are situated within a 140 km long section of southwest Norway (see Fig. 1). Fluid inclusion data from several eclogites in the WGR and BAS suggest that the eclogite facies fluids were both N_2–CO_2–H_2O fluids (Andersen *et al.*, 1989, 1990, 1993) and brines (Andersen *et al.*, 1993; Svensen *et al.*, 1999). This bimodality in the composition of these fluid inclusions has been recognized in other eclogite terranes as well (see Philippot *et al.*, 1995, and references therein), but there is no systematic regional pattern in the distribution of the two types of inclusion fluid in the WGR/BAS. Omphacite-hosted fluid inclusions have not been found in the ultra-high-pressure (UHP) part of the WGR.

The studied eclogites are mafic, i.e. they are dominated by garnet and omphacite and contain relatively small amounts of mica, kyanite and quartz, and are found as lenses or pods (up to several hundred metres in size) in a matrix of quartzofeldspathic gneiss. Field and petrographic evidence, along with bulk rock X-ray fluorescence (XRF) analyses, suggest that some of the eclogites had Fe–Ti-rich gabbroic or granulitic precursors (Table 1).

The eclogites at Bårdsholmen (locality 6, Figs 1 & 2a) probably formed by hydration and deformation of Precambrian granulite (Austrheim & Engvik, 1997; Engvik *et al.*, 2000). Granulite, eclogite and partly eclogitized granulite are all present at the outcrop scale. Eclogitized rocks contain omphacite, garnet, phengite, biotite, amphibole, rutile, quartz and apatite (Fig. 2b). Both omphacite and garnet contain fluid inclusions (Fig. 2c). Some eclogite lenses consist of alternating bands (on a millimetre and centimetre scale) that are dominated by garnet + omphacite or by phengite + biotite. Biotite and phengite comprise up to 18 vol.% of such eclogites (Engvik *et al.*, 2000). Partly eclogitized mafic granulites are preserved near fractures, with the degree of eclogitization decreasing away from the fracture. The eclogitization is characterized by corona formation around granulitic pyroxene, breakdown of plagioclase to clinozoisite and amphibole, and the formation of garnet, omphacite, phengite and biotite at the expense of pyroxene and feldspar (Engvik *et al.*, 2000). Biotite, amphibole and apatite are present in both the granulite and eclogite mineral assemblages. From Bårdsholmen (locality 6, Fig. 1), samples of eclogite (BÅR97-1, BÅR97-3 & B3) were selected for microprobe analyses of hydrous silicates and apatite.

Table 1. Bulk rock major and trace element contents.

Sample	BÅR97-1	EN97-5	LE97-1	HA12/93
SiO_2	44.02	42.73	41.28	40.43
TiO_2	2.33	3.01	5.08	3.99
Al_2O_3	15.27	14.26	11.00	13.92
FeO	15.60	19.50	22.32	18.33
MnO	0.20	0.20	0.22	1.27
MgO	7.14	6.06	5.68	3.21
CaO	9.36	9.94	10.33	12.09
Na_2O	2.83	2.54	2.61	2.72
K_2O	1.53	0.21	0.03	0.05
P_2O_5	0.35	0.01	0.12	1.00
Total	98.63	98.46	98.67	97.01
Cl	n.a.	73	193	45
Zn	160	120	149	125
Rb	58	9	5	< 5
Pb[a]	32	14	8	17
Sr	215	81	62	698
Y	51	22	28	502
Zr	172	47	93	4122
V	265	1196	977	77
Cr	418	379	297	220
Co	57	83	120	25
Ni	132	19	120	25
Cu	35	60	88	16
Nb	7	3	4	108
Th	< 2	< 2	< 2	67
U	< 2	< 2	4	2
La	12.3	1.1	2.6	1459
Ce	34.2	3.39	7.15	3078
Pr	5.11	0.55	0.99	358
Nd	24.5	3	6	1317
Sm	6.4	0.65	1.77	207
Eu	2.27	0.39	0.84	45.4
Gd	7.85	1.21	2.81	144
Dy	7.37	2.08	3.41	95.3
Ho	1.54	0.47	0.71	20.3
Er	4.33	1.19	2.02	56.4
Yb	3.99	1.25	1.92	69.4
Lu	0.62	0.21	0.29	11.9

Abbreviations: n.a., not analysed. [a]Detection limit, 8 ppm.

Amphibole occurs in both eclogite matrix and as inclusions in garnet. Detailed petrography of the granulite and eclogite facies rocks at Bårdsholmen is presented by Engvik *et al.* (2000).

The Engebøfjellet eclogite (locality 4, Fig. 1) is among the largest eclogite bodies in the WGR, covering an area of about 2500×500 m. The protolith is not preserved. From field relations, textures and chemical composition, it is interpreted to have been a layered gabbro. The eclogite lens is surrounded by various amphibolite facies gneisses. It consists of omphacite, garnet, rutile, amphibole, phengite, quartz and dolomite. Veins are common, and consist of omphacite, quartz, rutile, phengite and dolomite (Fig. 3a). The vein omphacite contains fluid inclusions (Fig. 3b). The vein samples (EN97-5 & EN97-8) come from two localities separated by *c*. 500 m. Conventional thermobarometry indicates that the veins formed at temperatures around 600 °C at pressures of 15–17 kbar (Erambert *et al.*, unpublished data).

The Leknes eclogite is situated about 3–4 km from Engebøfjellet (locality 5, Fig. 1). It is geochemically similar to the body at Engebøfjellet, about 100×50 m, and surrounded by gneiss. This eclogite has not previously been studied in detail, but it probably formed at *P–T* conditions similar to the Engebøfjellet eclogite. Veins that contain omphacite, quartz, phengite and amphibole are common. Samples LE-1 and LE-5 come from the same hand specimen of the Leknes eclogite.

The geology of Sjurdholmen (locality 7, Fig. 1; Holsnøy in the BAS) is complex and has not been studied in detail. The *P–T* conditions of eclogite formation were probably similar to those estimated for other nearby eclogite bodies of the BAS, i.e. *c*. 700–750 °C and 16–19 kbar (Jamtveit *et al.*, 1990). The mineral assemblage is dominated by omphacite, garnet and epidote–allanite. Rare earth element (REE)-rich epidote is abundant: the bulk rock contains 3000 ppm Ce, and more than 1000 ppm of La and Nd (Table 1). The degree of retrogression is locally high, and is dominated by the breakdown of omphacite to amphibole.

PETROGRAPHY OF OMPHACITE- AND GARNET-HOSTED FLUID INCLUSIONS

Omphacite- and garnet-hosted multiphase brine-filled fluid inclusions are abundant in only some of the mafic eclogites from west Norway. More than 30 eclogite localities were examined for fluid inclusions (Fig. 1). This study is based on the four localities described above, which were the only ones that were found to contain fluid inclusions.

Omphacite-hosted fluid inclusions

As in all fluid inclusion studies, the timing of entrapment must be established. The following observations (summarized in Fig. 4) form the basis for determining the relative time of inclusion entrapment: (i) fluid inclusions occur as three-dimensional clusters within cores of omphacite grains (Fig. 2c); (ii) rims of omphacite grains are typically free of fluid inclusions (Fig. 2c); (iii) if fluid inclusions occur along planar structures within the omphacite, these terminate at growth zones; (iv) some fluid inclusions have apparently nucleated on rutile in the omphacite (Fig. 5a); and (v) fluid inclusions are elongated and aligned parallel to the *c*-axis of omphacite. Based on these observations, the omphacite-hosted fluid inclusions are interpreted to be of primary or pseudosecondary origin, and thus to represent fluids that were trapped at eclogite facies conditions. On average, the omphacite-hosted fluid inclusions are 15 ± 10 μm long and 4 ± 2 μm wide (based on 121 measurements from samples BÅR97-1 & LE-1). Optical and scanning electron microscopy were used to identify daughter and 'step-daughter' crystals within omphacite-hosted fluid inclusions. A 'step-daughter' crystal is a solid that formed within a fluid inclusion by reaction of the fluid with the host mineral (Svensen *et al.*, 1999). Gypsum, calcite and sulphides are common 'step-daughters' (Fig. 5b). The number of solid phases within the
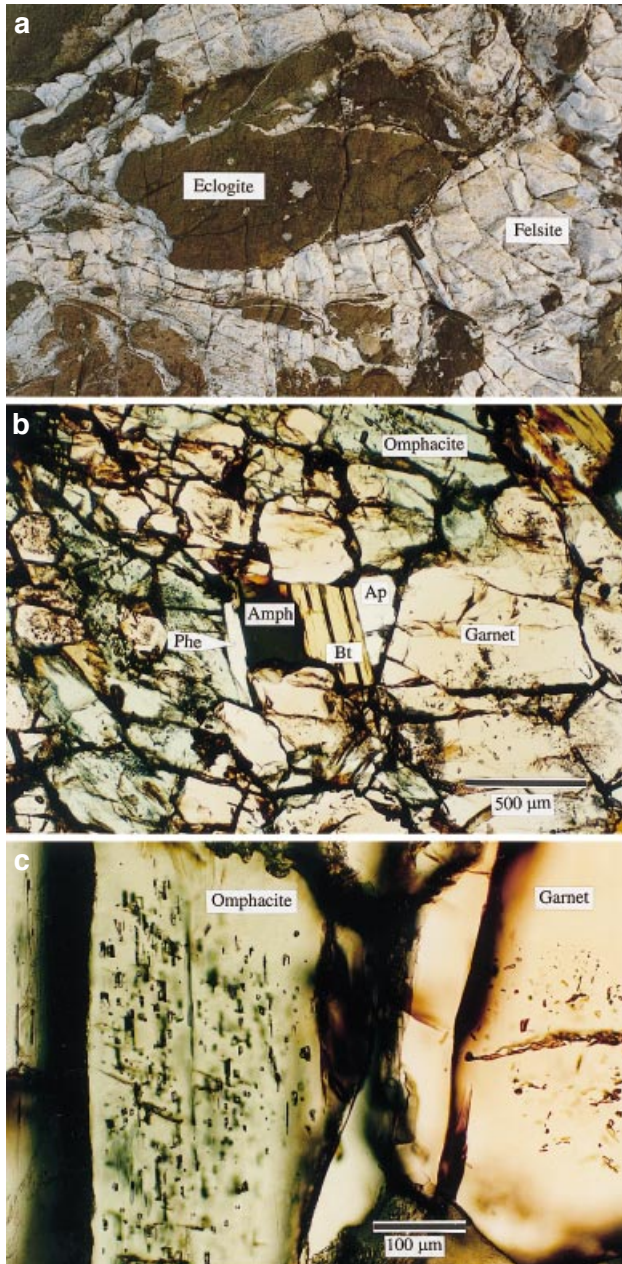
Fig. 2. (a) Eclogite lens in felsic gneiss at Bårdsholmen. The pen is 14 cm long. (b) Photomicrograph of a polished thin section of sample BÅR97-1, showing apparent equilibrium relations among omphacite, garnet, biotite, amphibole (Amph), phengite and apatite. (c) Photomicrograph of omphacite- and garnet-hosted fluid inclusions, BÅR97-1.
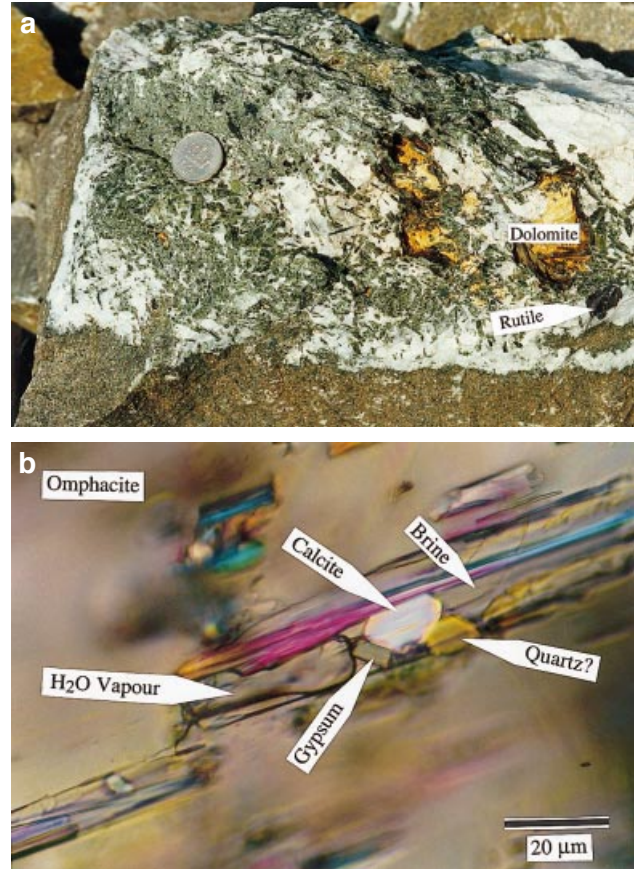

Fig. 3. (a) Omphacite (green)–quartz–dolomite (yellow)–rutile (black) vein in eclogite from Engebøfjellet. Sample EN97-5 comes from this particular vein. The coin is 2.5 cm in diameter. (b) Photomicrograph of omphacite-hosted fluid inclusions, EN97-8.
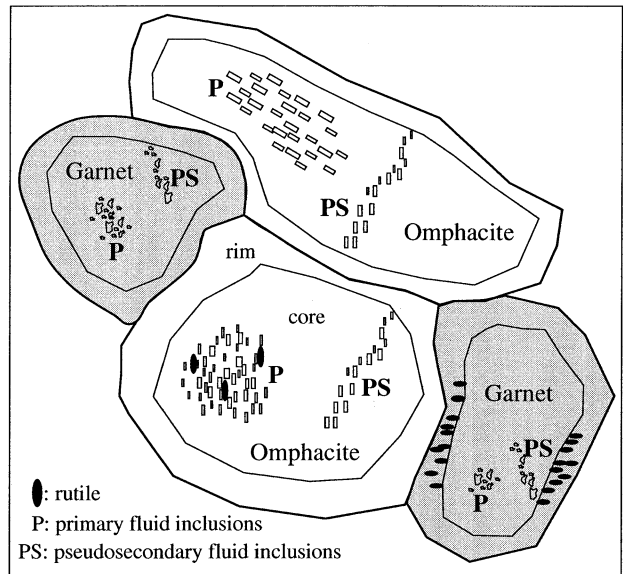
Fig. 4. Schematic textural relations of fluid inclusions in garnet and omphacite.

inclusions in a given sample is variable, usually between two and four, but is as high as 12 in sample BÅR97-1, which contains daughter and 'step-daughter' crystals of calcite, halite, quartz, K-feldspar, rutile, galena and chalcopyrite, and unidentified Zn–S, Fe–Ni–S, Pb–Cl and Pb–Cl–Br phases (Svensen *et al*., 1999; Fig. 5c). A Raman spectrum for the Pb–Cl–Br phase yields two peaks at 282 and 711 cm^{-1}, that can be distinguished

(a)

(b)

(c)

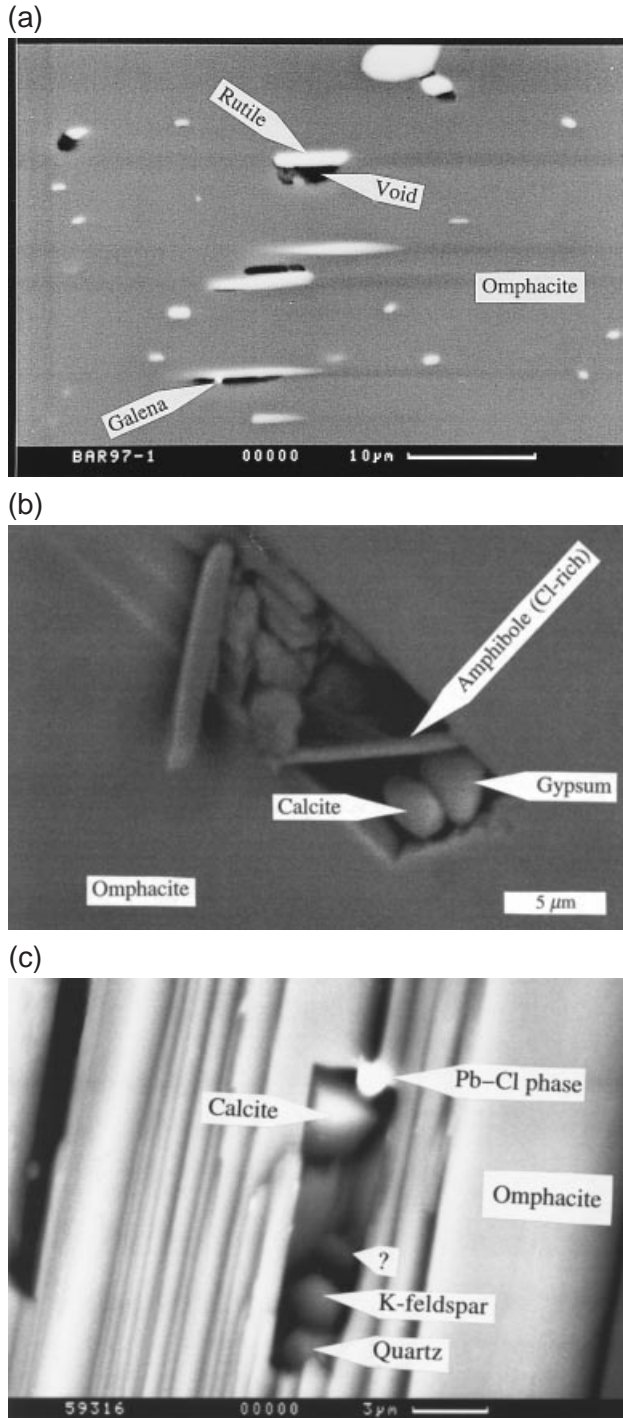**Fig. 5.** (a) Backscattered electron image of a polished section of sample BÅR97-1. The voids represent opened fluid inclusions that apparently nucleated on omphacite-hosted rutile grains. Backscattered images of opened fluid inclusions from sample LE-1 (b) and sample BÅR97-1 (c).

from the background omphacite. Some amphibole inclusions in garnet grains in sample BÅR97-1 occur in aggregates with rutile, ilmenite, Fe-oxide, chalcopyrite, and sometimes calcite, galena, epidote–allanite, voids
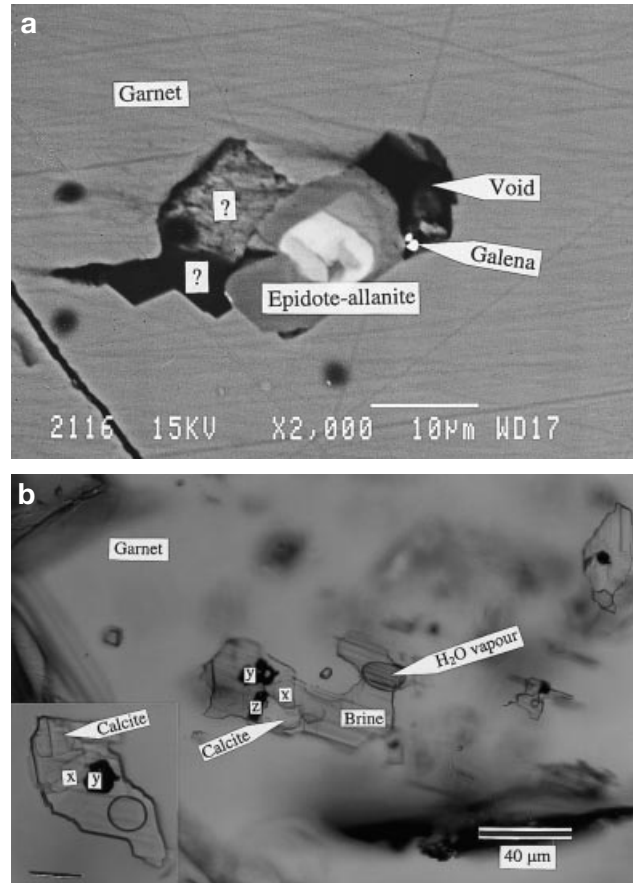

Fig. 6. (a) Backscattered electron image of a polished section of sample BÅR97-1, showing void and solids in garnet. The void contains a Pb–S phase (galena), and probably represents an opened fluid inclusion. (b) Garnet-hosted fluid inclusions (from sample LE-1) which contain three unidentified minerals (x, y, z), in addition to calcite.

and an unidentified Si–Al–Mg phase (Fig. 6a). These aggregates are arranged around voids that probably represent opened fluid inclusions.

Garnet-hosted fluid inclusions

Garnet-hosted, multiphase fluid inclusions are found in all samples that contain omphacite-hosted fluid inclusions. Representative textural relations of fluid inclusions and the garnet host are illustrated in Fig. 4. Fluid inclusions in garnet are irregularly shaped, typically < 20 μm in diameter, and contain a vapour bubble plus several solid phases. The degree of fill of solid phases is, as in the omphacite-hosted fluid inclusions, $< 40\%$. The garnet-hosted fluid inclusions commonly have striated walls (Fig. 6b), which probably represent an approach to negative garnet crystal shapes. Most fluid inclusions are either: (i) isolated or grouped in a garnet core; or (ii) trails that terminate within garnet. Both can probably form during crystal growth. The presence of galena in void spaces in garnet from sample BÅR97-1 (Fig. 6a) is

reminiscent of the galena found in omphacite-hosted fluid inclusions, and differences among suites of step-daughter crystals in omphacite and garnet probably reflect differences in host mineral composition. In contrast to omphacite, halite is not observed in garnet-hosted fluid inclusions from sample BÅR97-1. Raman spectroscopy identified calcite in all these fluid inclusions. An unidentified solid in sample LE-5 has major Raman peaks at 227, 242, 282, 318, 343, 365, 991, 1078, 1153 and 1165 cm^{-1}. The absence of a peak at 3200 cm^{-1} implies a lack of OH-bonds in that mineral.

ANALYTICAL METHODS

Microthermometry

Fluid inclusions were measured on Linkam THM600 heating–freezing stages at the University of Leeds and at the University of Oslo. Apparent salinities in the H_2O–NaCl system were calculated from the freezing point depression relationships given by Bodnar (1993). The stages were calibrated between -56.6 and 450 °C, and the analytical precision is ± 0.1 °C between -30 and $+10$ °C.

Electron microprobe analyses

Minerals were analysed with the CAMECA CAME-BAX electron microprobe at the Mineralogical-Geological Museum, University of Oslo, using natural and synthetic standards. Biotite, phengite, amphibole and apatite were analysed with a beam current of 10 nA and an accelerating voltage of 15 kV. All analyses were carried out with a focused beam of *c.* 1 μm in diameter. Peak and background values were counted for 10 s for major elements and 20 s for Cl and F. Uncertainties (1σ) are higher for F than for Cl, and reach 30% and 8%, respectively, for values < 0.4 wt%. For F concentrations < 0.2 wt%, 1σ uncertainties are $> 70\%$, whereas the 1σ value is within 50% down to 0.03 wt% Cl.

Crush–leach analysis

To separate omphacite and garnet, samples of eclogite or eclogitic vein were crushed to a grain size of 500–1000 μm. Omphacite and garnet were hand picked under a binocular microscope. The separates and crushed bulk eclogite samples were boiled in double distilled water to remove impurities and adsorbed ions. Crush–leach analyses were carried out using the methods described by Bottrell *et al.* (1988) and Banks & Yardley (1992). Some 1–2 g of cleaned sample was ground to a fine powder with an agate pestle and mortar, transferred to a 7 ml sample container, and the dried salts leached with 5–6 ml of double distilled water for anion and alkali analysis. The samples were then filtered through 0.2 μm Whatman nylon membrane filter prior to analysis. Anions (Cl$^-$, Br$^-$, SO$_4^{2-}$, F$^-$) were determined with a Dionex 4500 ion chromatograph fitted with a Ag-14 column and conductivity detector. Iodine was determined separately, using an online preconcentrator and a guard column for separation, due to long retention times with the normal column. For cation analyses, a 2000 ppm La solution (0.2 ml) was added to 2 ml of the sample leachate to inhibit the ionization of K and Li. Sodium, K and Li were determined by flame emission spectrometry. The detection limit is *c.* 0.1 ppb for I and 0.5 ppb for Br.

GEOCHEMISTRY OF MINERALS AND FLUIDS

Halogen contents of eclogite minerals

Mineral chemical analyses were primarily carried out on samples from Bårdsholmen, due to the relatively high halogen contents of minerals from these eclogites. (Halogen contents of hydrous silicates from the other eclogites are below the electron microprobe detection limit.) The mineral chemistry of representative biotite, amphibole and phengite is presented in Table 2, and apatite in Table 3. A graphical presentation of the average mineral F/Cl mass ratios is shown in Fig. 7.

Biotite

Biotite is abundant in the eclogites at Bårdsholmen, but relatively uncommon in the eclogites of western Norway. Fluorine is dominant relative to Cl by a factor of two (by weight), with average values of 0.30 and 0.15 wt%, respectively. Biotite grains in samples BÅR97-1 and BÅR97-3 have F/Cl mass ratios of 2.1 ± 0.6. It is difficult to determine whether Cl and F are correlated with other elements, but there may be a correlation between Cl and Al (negative, with $R^2 = 0.60$), F and Al (negative, with $R^2 = 0.58$) and F and Fe (negative, with $R^2 = 0.59$).

Amphibole

Amphibole inclusions in garnet have higher Cl contents (average of 1.3 ± 0.2 wt%) than matrix amphibole (0.2 ± 0.1 wt%). The opposite is the case for F: 0.1 and 0.2 wt%, respectively. Amphibole inclusions in garnet display an average F/Cl mass ratio of 0.16 ± 0.14, whereas matrix amphibole has a ratio of 1.1 ± 0.6. Potassium is negatively correlated with Cl ($R^2 = 0.72$) and positively correlated with Fe ($R^2 = 0.58$). Amphibole in garnet has the highest Cl and the lowest K content. However, amphibole from eclogite facies felsic rocks at Bårdsholmen show a positive correlation between K and Cl (Austrheim & Engvik, 1997), as is seen in high-grade amphibole (i.e. Kullerud, 1995; Léger *et al.*, 1996; Markl *et al.*, 1998).

Phengite

The F content of phengite is < 0.5 wt%, and the Cl content is very low (< 0.05 wt%). The average F/Cl

Table 2. Biotite, amphibole and phengite compositions.[a]

Mineral sample	Bt BÅR97-1	Bt BÅR97-1	Bt BÅR97-1	Amp BÅR97-1	Amp BÅR97-1	Amp BÅR97-1	Amp BÅR97-1	Amp B3	Amp B3	Phe BÅR97-1	Phe BÅR97-1	Phe B3
SiO_2	39.26	39.62	37.95	42.16	38.94	42.12	37.83	45.26	39.29	53.34	54.66	52.60
TiO_2	2.19	2.06	0.76	0.45	0.06	0.47	0.82	0.52	0.71	0.48	0.74	0.57
Al_2O_3	16.36	15.12	17.84	16.56	18.70	15.71	17.67	14.79	19.46	26.62	27.52	27.18
FeO	13.82	13.08	12.98	13.23	18.94	13.96	18.57	12.33	17.04	2.46	2.37	2.65
MnO	0.13	0.03	0.14	0.03	0.04	0.13	0.19	0.05	0.06	0.04	0.03	0.00
MgO	13.78	16.15	16.06	10.11	5.88	10.07	6.85	11.27	6.82	4.31	4.21	3.97
CaO	0.01	0.11	0.19	10.09	9.28	10.00	11.03	9.51	9.51	0.00	0.02	0.03
Na_2O	0.21	0.29	0.14	3.55	4.18	3.20	2.52	3.46	4.04	0.34	0.41	0.41
K_2O	9.62	9.68	9.76	1.26	0.46	1.25	2.04	1.14	1.08	10.21	9.65	10.98
BaO	0.22	0.19	0.23	0.05	0.00	0.00	0.00	0.04	0.04	0.13	0.22	0.16
Cl	0.16	0.21	0.12	0.34	1.41	0.23	0.15	0.25	1.28	0.02	0.03	0.03
F	0.72	0.73	0.53	0.53	0.27	0.41	0.03	0.41	0.15	0.48	0.00	0.23
Total	96.48	97.27	96.70	98.36	98.16	97.55	97.70	99.03	99.48	98.43	99.86	98.81
O: -Cl-F	0.34	0.36	0.25	0.30	0.43	0.22	0.05	0.23	0.35	0.21	0.01	0.11
Total	96.14	96.91	96.45	98.06	97.73	97.33	97.65	98.80	99.13	98.22	99.85	98.70
Si	2.87	2.87	2.76	6.19	5.91	6.25	5.81	6.52	5.85	3.44	3.45	3.40
Ti	0.12	0.11	0.04	0.05	0.01	0.05	0.09	0.06	0.08	0.02	0.03	0.03
Al	1.41	1.29	1.53	2.87	3.34	2.75	3.20	2.51	3.41	2.02	2.05	2.07
Fe^{2+}	0.84	0.79	0.79	1.62	2.40	1.73	2.38	1.48	2.12	0.13	0.13	0.14
Mn	0.01	0.00	0.01	0.00	0.01	0.02	0.03	0.01	0.01	0.00	0.00	0.00
Mg	1.50	1.74	1.74	2.21	1.33	2.23	1.57	2.42	1.51	0.41	0.40	0.38
Ca	0.00	0.01	0.02	1.59	1.51	1.59	1.81	1.47	1.52	0.00	0.00	0.00
Na	0.03	0.04	0.02	1.01	1.23	0.92	0.75	0.97	1.17	0.04	0.05	0.05
K	0.90	0.89	0.91	0.24	0.09	0.24	0.40	0.21	0.21	0.84	0.78	0.91
Ba	0.01	0.01	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00
Cl	0.02	0.03	0.02	0.09	0.36	0.06	0.04	0.06	0.32	0.00	0.00	0.00
F	0.17	0.17	0.12	0.46	0.13	0.19	0.02	0.19	0.07	0.10	0.00	0.05

Abbreviations: Amp, amphibole; Phe, phengite. [a]Biotite and phengite formulae based on 11 and amphibole on 23 oxygens.

mass ratio (11.4 ± 8.3) is thus significantly affected by analytical uncertainties in addition to possible chemical heterogeneities.

Apatite

Apatite from eclogite sample BÅR97-1 contains up to 3.5 wt% F (0.95 atomic formula units, a.f.u.) and 0.6 wt% Cl (Table 3). The average F/Cl mass ratio in the apatite is 5.3 ± 0.8. Chlorine and F are correlated ($R^2 = 0.81$). The biotite–apatite thermometer of Zhu & Sverjensky (1992), which is based on the F–Cl exchange between biotite and apatite and the Fe, Mg and Al^{VI} contents of biotite, gives 560–650 °C for sample BÅR97-1 and 385 °C for sample BÅR97-3 (at 15 kbar). Independent thermometers (garnet–clinopyroxene) gave temperatures for the eclogite around 500 °C (Engvik *et al.*, 2000). The effect of pressure on estimated biotite–apatite temperatures is small.

Compositions of fluid inclusions

Because Ca-, Na- and K-bearing daughter or step-daughter crystals are present in the fluid inclusions, microthermometry is not a particularly useful tool for reconstructing the eclogite facies fluid chemistry. Microthermometry was only used to obtain salinities
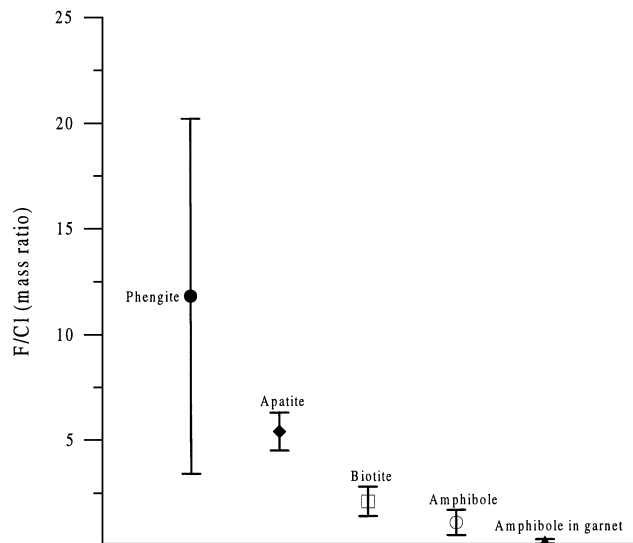
Table 3. Apatite compositions.[a]

Sample	BÅR97-1	BÅR97-1	B3
FeO	0.29	0.26	0.11
CaO	54.89	55.82	55.85
P_2O_5	43.03	43.66	43.57
Cl	0.38	0.25	0.28
F	2.66	2.33	2.06
Total	101.25	102.32	101.87
O: -Cl-F	1.21	1.04	0.93
Total	100.04	101.28	100.94
Fe	0.02	0.02	0.01
Ca	4.94	5.02	5.00
P	3.06	3.18	3.08
Cl	0.05	0.04	0.04
F	0.71	0.62	0.54

[a]Formulae based on 13 (O, OH, F, Cl).

Fig. 7. F/Cl mass ratios of the eclogite minerals. The error bars represent the standard deviation of the spread in the analyses.

in the $NaCl$–H_2O model system, expressed as weight per cent $NaCl$ equivalent in Fig. 8.

Omphacite-hosted inclusion fluids

Apparent salinities in the H_2O–$NaCl$ system were calculated using the freezing point depression relationships of Bodnar (1993), along with microthermometry. Final melting temperatures of ice range from − 20 to − 14 °C in all samples. The resulting salinities of 95 omphacite-hosted fluid inclusions (Fig. 8) are between 17.5 and 26.5 wt% $NaCl$ equivalent. All samples show some spread in the salinity. First-melting temperatures were not systematically measured, but are up to 8 °C lower than the eutectic in the H_2O–$NaCl$ system, which indicates that the brines lack significant Ca or Mg. Heating homogenized the vapour phase to liquid between 300 and 420 °C; decrepitation occurred at temperatures > 450 °C. Halite was identified from sample BÅR97-1 in opened fluid inclusions (using scanning electron microscopy, SEM), and was also observed using the optical microscope. However, within a population of fluid inclusions in a single omphacite crystal from sample BÅR97-1, only some of the fluid inclusions were found to contain halite daughter crystals, i.e. the rest had salinities below 26.5 wt% $NaCl$ equivalent. This can be explained by differential post-entrapment salinity modifications, or salinity variations in the eclogite facies pore fluid composition during trapping of the fluid inclusions. The latter seems more likely considering the possible effects on pore fluid chemistry by local eclogite-forming hydration reactions (Svensen *et al.*, 1999). Dissolution of halite in sample BÅR97-1 occurred between 470 and 555 °C (10 measurements, $\bar{x} = 511$ °C). The minimum apparent salinity for the halite-bearing fluid inclusions (halite dissolution temperatures from Chou, 1987) was
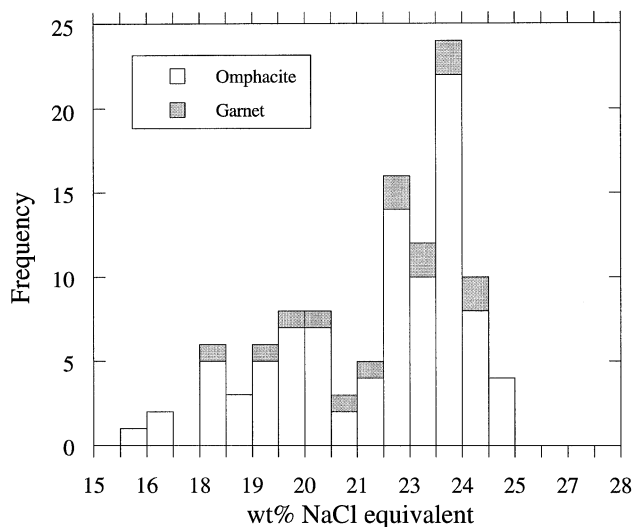
estimated from the maximum vapour–liquid homogenization temperature (i.e. 420 °C), because vapour–liquid homogenization occurred before halite dissolution, giving about 50 wt% $NaCl$ equivalent. The small size of the fluid inclusions and the high degree of fill (of solid phases) precluded measurements of the melting point of ice (and thus the salinity) in halite-free fluid inclusions. When reconstructing the fluid chemistry preserved in sample BÅR97-1, a salinity between 30 and 40 wt% $NaCl$ equivalent seems plausible. The only elemental compositions recalculated for this sample are the halogens.

The crush–leach analyses (Tables 4 & 5) are mass ratio combinations of Cl^-, Br^-, I^-, F^-, SO_4^{2-} and Li^+. Cl/Br and Cl/I mass ratios of the omphacite leachates are shown in Fig. 9. The Cl/Br mass ratios are lower than that of modern seawater (with one exception); Cl/Br mass ratios from sample BÅR97-1 are as low as 33. The low Cl/Br mass ratio of the fluid inclusions from this sample was interpreted by Svensen *et al.* (1999) to reflect fractionation between hydrous minerals and pore fluid during eclogite-forming hydration. Cl/Br ratios in bulk vein samples are higher than in the pure omphacite leaches, and probably reflect contamination. The highest Cl/Br mass ratios (244 and 280) come from sample HA12/93. The Cl/I mass ratios (10 000–35 000) are considerably lower (two orders of magnitude) than that of modern seawater ($c.\ 4 \times 10^5$), whereas the Li/Cl ratios are in general two orders of magnitude higher
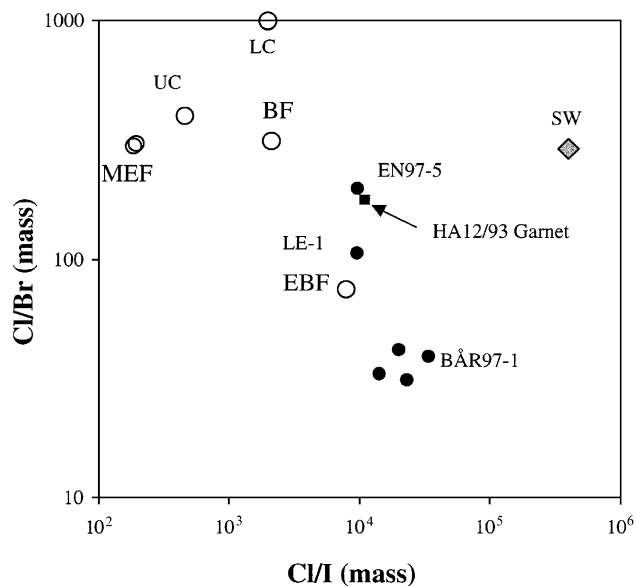
Fig. 9. Halogen systematics of the omphacite and garnet inclusion fluids, with some reference values of rocks and fluids. The Monviso eclogite fluid inclusions (MEF) are described by Philippot & Selverstone (1991); crush–leach analyses from Yardley (1997) are Cl/I = 184 and 193, Cl/Br = 300 and 307. Upper (UC) and lower (LC) crustal halogen ratios of rocks from Wedepohl (1995). The sedimentary basin average fluid analyses (BF) are from Worden (1996), and fluids with the lowest Cl/Br ratios are for evaporite-bearing sequences (EBF). SW is modern seawater.

Fig. 8. Salinity (expressed as wt% $NaCl$ equivalent) of omphacite- and garnet-hosted fluid inclusions.

Table 4. Crush–leach analyses of omphacite and bulk samples (mass ratios).

Omphacite	Br[a]	Cl/Br	F/Cl	SO$_4$/Cl	Cl/I	Li/Cl
LE-1 a	9	190		0.32		0.0012
LE-1 b	27	59	0.004	0.52		
LE-1 c	13	233		0.72		0.0007
LE-1 d	25	59	0.0089	3.97		0.0023
LE-1 e	38	90	0.0006	0.93		0.0006
EN97-5 a	20	198		1.00	9602	0.0003
EN97-5 b	85	111	0.0038	0.60		
EN97-5 c	120	106	0.0013	1.11		0.0003
EN97-8 a	2	183				0.0024
EN97-8 b	3	201				0.0033
EN97-8 c	4	162		3.43		0.0020
EN97-8 d	22	170	0.0046	1.54		
HA12/93 a	21	244	0.0306	0.31		0.0006
HA12/93 b	12	280	0.0079	0.06		
BÅR97-1 a	136	39	0.0094	0.03	33785	0.0006
BÅR97-1 b	136	42	0.0528	0.04	20007	
BÅR97-1 c	110	33	0.0099	0.09	14112	0.0003
BÅR97-1 d	245	38	0.0039	0.01		
Bulk vein						
LE-1 a	50	124	0.0028	0.24		0.0005
LE-1 b	38	106	0.0027	0.24	9542	0.0005
LE-1 c	26	126	0.0022	0.23		
EN97-5 a	35	103		0.30		
EN97-5 b	48	127		0.30		
EN97-5 c	29	108		0.29		
EN97-5 d	27	59	0.0038	0.19		0.0014
EN97-5 e	79	123	0.0020	0.14		0.0001
EN97-8 a	6	395		1.60		0.0009
EN97-8 b	8	353		1.25		0.0008
Bulk eclogite						
HA12/93 a	6	320	0.2788	0.13		0.0006
HA12/93 b	16	225	0.0344	0.04		
HA12/93 c	9	273	0.0376	0.05		
HA12/93 d	8	300	0.0392	0.05		
BÅR97-1 a	156	31	0.1141	0.04	23062	0.0007
BÅR97-1 b	191	34	0.0284	0.04		
BÅR97-1 c	170	34	0.0280	0.04		
BÅR97-1 d	128	36	0.0308	0.02		
LE-1 a	50	47	0.1619	0.50		0.0005
LE-1 b	35	103	0.0070	0.46		
LE-1 c	59	50	0.0068	0.51		
Seawater		291			396000	9×10^{-6}

[a]Concentration in leachate, in ppb.

than that of modern seawater. Furthermore, the leachates display Br/I mass ratios of 48–862 (1360 in modern seawater) and Li/Br ratios of 3–20 (0.003 in modern seawater). The SO$_4$/Cl ratios and reconstructed SO$_4$ concentrations should be treated with care as the eclogites are commonly sulphide-rich (chalcopyrite and pyrite), and some of the analyses have SO$_4$/Cl mass ratios as high as four, obviously due to contamination from a sulphur phase. The solubility of gypsum is low;

Table 5. Repetitive leaches of sample BÅR97-1.

Sample	Cl (ppm)	F (ppm)	SO$_4$ (ppm)	Br (ppb)	Cl/Br (mass)	Cl/F (mass)	Cl/SO$_4$ (mass)
1. leach							
BÅR97-1 a	5.3	0.05	0.18	136	39	106	29
BÅR97-1 b	5.7	0.3	0.22	136	42	19	26
2. leach							
BÅR97-1 a	0.5	0.02	0.08	15.3	33	25	6
BÅR97-1 b	0.5	0.01	0.04	13.9	36	50	13
3. leach							
BÅR97-1 a	0.4	n.d.	0.35	6.5	62	—	1
BÅR97-1 b	0.4	n.d.	0.23	7.4	54	—	2

Abbreviations: n.d., not detected.

thus the leachate should have SO$_4$ contents that correspond to gypsum–sulphide–brine room-temperature equilibrium. Sulphur must have been an important element in the eclogite brines, as gypsum and metal sulphides are found in the fluid inclusions. The SO$_4$/Cl ratio is lowest in the BÅR97-1 leachates (0.01–0.09): the fluid inclusions in this sample do not contain gypsum. Fluid inclusions from both matrix and vein omphacite have been analysed from sample LE-1, and do not indicate compositional differences.

The solubilities of daughter and step-daughter crystals may control the element ratios from the crush–leach analyses, as is the case for Cl, Br and SO$_4$ in sample BÅR97-1, Cl and SO$_4$ in sample LE-1, and SO$_4$ in the other samples. Leachates from three successive crush–leach series of two sets of sample BÅR97-1 show Cl/Br mass ratios of 39/42, 32/34 and 56/47 (Table 5). The differences between the first and the second values are within analytical uncertainty, but the increase in the Cl/Br ratio from the second to the third values is significant, and may reflect the dissolution of Cl from minerals in the eclogite. The Br contents in the third leachate are 5% of those in the first. F/Cl ratios increase by 30% from the first to the second leach. Fluorine was not detected in the third leachate. Bulk eclogite leaches were carried out in order to determine to what extent halogen-bearing minerals release Cl, Br, F or I to the leachate. Halogens in minerals are only detectable by the electron microprobe in samples from Bårdsholmen; thus the dissolution of these minerals during leaching could yield higher Cl/Br ratios in the leachate. This does not seem to have had any effect on the halogen ratios of the bulk leaches, which are within 10% of the values for omphacite.

Fluid inclusion chemistry was reconstructed on the basis of Cl contents derived from microthermometry, and is presented in Tables 6 (omphacite-hosted fluids) and 7 (garnet-hosted fluids). The uncertainties in microthermometry and chromatography are *c*. 10%. All analyses of omphacite leaches showed a large positive deviation from charge balance (positive/

Table 6. Reconstructed composition of omphacite-hosted fluid inclusions (in ppm).

Sample	NaCl (wt%)	Cl	Br	I	F	SO$_4$	Li	K
LE-1 a	24.0	150438	765			45982	179	1698
LE-1 b	24.0	150438	2456		550	75678		
LE-1 c	24.0	150438	624	5		105484	105	210
LE-1 d	24.0	150438	2463	10	1295	577457	329	7558
LE-1 e	24.0	150438	1625	4	85	135825	93	2987
EN97-5 a	20.9	129621	639	14		126906	35	13061
EN97-5 b	20.9	129621	1141		484	76680		2305
EN97-5 c	20.9	129621	1199	1	161	140573	33	8867
EN97-8 a	22.0	136022	728				324	
EN97-8 b	22.0	136022	665				438	
EN97-8 c	22.0	136022	826			457346	261	
EN97-8 d	22.0	136022	784		611	205875		
HA12/93 a	22.9	138911	568	3	4257	43047	88	7980
HA12/93 b	22.9	138911	497		1104	8383		14214
BÅR97-1 min	30	181980	4400	5	700			
BÅR97-1 max	40	242640	7319	17	2400			
Seawater	3.2	19000	65	0.06	1.3	1766	0.17	380

Table 7. Crush–leach analyses of garnet.

Sample	HA12/93	BÅR97-1	LE-1
Cl/Br	178	61	—
F/Cl	0.36	0.07	0.05
SO$_4$/Cl	0.74	0.10	0.42
Cl/I	10940	—	—
Li/Cl	0.001	—	—
K/Cl	0.09	3.64	—
Na/Cl	0.83	0.39	0.74
Na/K	9.78	0.11	—
Cl[a]	131632	154076	138305
Br	740	2534	—
I	12	—	—
F	47888	11005	6649
SO$_4$	96859	15408	57627
Li	132	—	—
K	11190	560508	—
Na	109493	60530	101955
Na[a]	85368	99924	89695

[a]Determined by microthermometry.

negative ratio up to 20), caused by Na contamination from omphacite. Thus the reconstructed element concentrations are likely to be slightly underestimated, as chlorides other than NaCl have not been taken into account. However, microthermometry and the presence of solid phases within the fluid inclusions indicate that the concentrations of $CaCl_2$, $MgCl_2$, KCl and $FeCl_2$ in the fluid are low.

Bromine contents range from 500 ppm (sample HA12/93) to 4400–7300 ppm (sample BÅR97-1). Iodine contents (1–17 ppm) are higher than for most crustal rocks (0.01–10 ppm) and modern seawater (65 ppb), but comparable to high-temperature hydrothermal fluids (<50 ppm; Böhlke & Irwin, 1992). Fluorine contents range from 100 to 2400 ppm, Li ranges from 30 to 400 ppm, and K varies considerably from 200 ppm (LE-1) to 47 000 ppm (BÅR97-1). Because large variations of element contents between replicate leaches are probably due to contamination, average values are more reliable.

Garnet-hosted inclusion fluids

Results from microthermometry of garnet-hosted fluid inclusions are presented (Fig. 8) as wt% NaCl equivalent. Seventeen inclusions were measured, and the final melting temperature of ice was in the same range as that for fluid inclusions in omphacite, yielding 18–26 wt% NaCl equivalent. The two crush–leach analyses from samples HA12/93 and BÅR97-1 show that Cl/Br ratios are lower and higher, respectively, than those for omphacite and bulk eclogite (Table 5). The differences between the Cl/Br ratios from garnet and omphacite leaches of sample BÅR97-1 could imply that the garnet crush–leaches are contaminated by Cl from high Cl-amphibole inclusions in the garnet, or that different retrograde reactions have modified the Cl/Br ratios of the fluids. The possibility that garnet and omphacite originally trapped different fluids seems unlikely considering the fluid inclusion petrography. The F/Cl mass ratio is more than an order of magnitude higher

(0.13) in garnet-hosted fluid inclusions than for the average omphacite-hosted fluid inclusions (0.008). Bromine contents of HA12/93 garnet-hosted fluid inclusions (740 ppm) are about 100–200 ppm higher than the omphacite fluid inclusions. The CO_2 was probably consumed by the calcite-forming reaction, because CO_2 (or N_2) was not detected by Raman spectroscopy. Sodium (*c*. 10 wt% in samples HA12/93 & LE-1, and 6 wt% in sample BÅR97-1) and K (*c*. 11 000 ppm in sample HA12/93) were analysed in garnet leachates because the content of these elements in garnet is negligible. Charge-balance calculations show that the chemistry of fluid inclusions in garnet from samples HA12/93 and LE-1 is representative (showing 18% and 4% deviation from neutrality, respectively), whereas fluids extracted from sample BÅR97-1 are contaminated by K (showing 54% deviation from neutrality).

DISCUSSION

Formation and preservation of fluid inclusions in high *P–T* systems

Fluid inclusions are present only in omphacite from a few eclogite bodies (four localities) from more than 30 localities in Norway. Furthermore, fluid inclusions are only locally present within the individual bodies (except the Bårdsholmen samples). The implications of grain size and deformation for fluid inclusion formation and preservation are seen in the Engebøfjellet samples. Here coarse-grained omphacite grains in veins contain primary/pseudosecondary fluid inclusions, whereas finer grained omphacite grains in the wall-rocks have no fluid inclusions. At Bårdsholmen, fine-grained eclogites are fluid inclusion free, but inclusions are abundant in coarse-grained eclogite. Recrystallization at eclogite facies conditions may explain the absence of omphacite-hosted fluid inclusions from most eclogites from the WGR/BAS. However, the formation of fluid inclusions requires a free fluid phase, which was not necessarily present in all eclogites in the WGR/BAS, even those containing hydrous minerals. Omphacite from the mafic eclogites in the WGR/BAS is more Fe-rich than that from other eclogites in the WGR/BAS, which seems to be connected with the presence of fluid inclusions. In contrast, fluid inclusions in zoned omphacite from Monviso eclogite were confined to zones with high amounts of diopside (Philippot & Selverstone, 1991).

Fluid inclusions in omphacite have played an important role during retrogression. Three sites of omphacite breakdown can be seen in thin section: (i) along grain boundaries; (ii) along fractures; and (iii) at fluid inclusions. The latter is illustrated by the breakdown of omphacite to amphibole at the site of primary or pseudosecondary fluid inclusions. In contrast, garnet breaks down primarily along grain boundaries and fractures. The presence of a variety of solid phases

inside fluid inclusions in high-grade metamorphic minerals might give information about the preservation of the inclusion during retrograde conditions. Perhaps retrograde reactions between fluids and omphacite inhibit the total consumption of fluids, via precipitation on the fluid inclusion walls, and so isolate the fluid from the host mineral.

Omphacite-hosted fluid inclusions commonly contain calcite (in all samples) and gypsum (all samples except BÅR97-1). The presence of these minerals in the fluid inclusions, combined with the fact that CO_2 and SO_4 are not components of omphacite, indicate that the original inclusion fluid must have contained significant amounts of C and S. Svensen *et al.* (1999) estimated the CO_2 contents of fluid inclusions in sample BÅR97-1 to be between 2 and 12 mol.%, based on the sizes of calcite daughter crystals. Calcite and gypsum could have formed by (i) direct precipitation from the inclusion fluid; or (ii) retrograde reactions between the inclusion fluid and its omphacite host, which contributed some of the Ca in the calcite/gypsum step-daughter crystals. Retrograde reactions between omphacite and inclusion fluids could be of the form:

$$Omph + H_2O + CO_2 = Amp + Cal + Qtz \qquad (1)$$

A similar reaction was suggested by Andersen *et al.* (1984) for retrograde formation of amphibole in pyroxene-hosted fluid inclusions within mantle xenoliths. However, amphibole (Cl-bearing) and calcite only appear to coexist inside the fluid inclusions from sample LE-1 (Fig. 5b). No chemical gradients of Ca (or any other major element) are observed in the omphacite surrounding the amphibole + calcite-bearing fluid inclusions. However, on the scale of the omphacite grains, there is typically a difference in the composition of fluid inclusion-rich cores and fluid inclusion-free rims. Whether this reflects post-entrapment diffusion of elements between fluid inclusions and host minerals or geochemical differences during growth is unclear. Despite the lack of petrographic evidence for the formation of calcite and gypsum by retrograde reactions, it is still a plausible mechanism. Some of the Fe released by reaction (1) could form Fe-sulphides, which are also commonly observed in the fluid inclusions. Quartz (see Fig. 5c) may be a product of retrograde reactions, but could also be a daughter crystal, because the solubility of silica (in equilibrium with quartz) at eclogite facies conditions is 20 000–30 000 ppm (Manning, 1994). For a fluid inclusion of $15 \times 4 \times 4$ μm, precipitation of 25 000 ppm silica from solution would form a quartz daughter crystal of about 2.3 μm^3 (i.e. *c.* 10% of the fluid inclusion volume).

Fractionation of Cl and F between minerals and fluid

Apparent mineral–fluid F/Cl partitioning coefficients

(D) have been calculated for sample BÅR97-1 according to:

$$D = (F/Cl)^{mineral}/(F/Cl)^{brine}$$

in which (F/Cl)^{brine} is the average F/Cl mass ratio of the inclusion fluids in sample BÅR97-1 (0.008). Apatite and phengite have the largest mineral–fluid F/Cl partitioning coefficients ($D = 675 \pm 114$ and 1469 ± 1048, respectively); amphibole inclusions in garnet have the smallest ($D = 20 \pm 18$). Amphibole in the matrix has $D = 131 \pm 72$, and biotite $D = 259 \pm 87$. Thus F will be enriched in mineral structures compared to Cl, with the fractionation being greatest for phengite and apatite. Consequently, apatite and phengite growth have the most impact on the F/Cl ratio of the fluid. The uncertainties of the D values are considerable, and they should be regarded as order-of-magnitude estimates. Brenan (1993) determined D values from 900 to 1100 on F–Cl fractionation between apatite and fluid (at $P = 10$–20 kbar, $T = 950$–1050 °C). The bulk eclogite leachates yield larger F/Cl mass ratios than do the omphacite leachates, probably due to contamination in the former.

Halogen systematics in lower crustal fluids

The Cl/I ratios of the fluid inclusions analysed are considerably higher than those of average crustal rocks (Fig. 9). The bulk rock Cl/I ratio may increase with metamorphic grade, as I is released from minerals and organic matter (Fuge & Johnson, 1986; Boneß *et al.*, 1991; Wedepohl, 1995; Muramatsu & Wedepohl, 1998). The I contents of BÅR97-1 fluid inclusions are relatively small (1–17 ppm) compared to those of other high-temperature fluids (see Böhlke & Irwin, 1992). This is in contrast to the fluid inclusions from eclogites
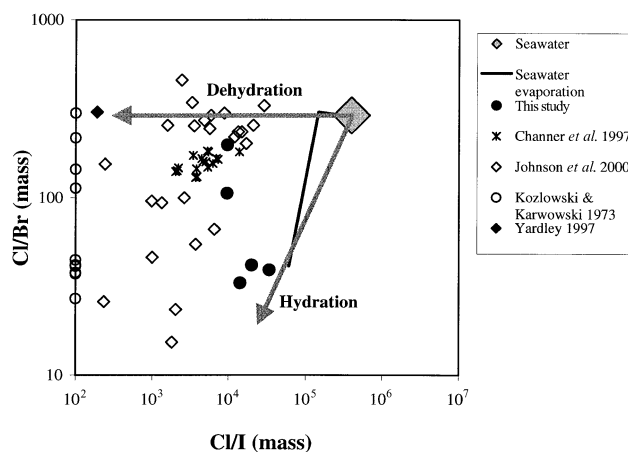

Fig. 10. Evolution of Cl/Br and Cl/I ratios according to metamorphic process. Data from Johnson *et al.* (2000) represent mantle fluids, Channer *et al.* (1997) vein fluids in greenstone, Kozlowski & Karwowski (1973) vein fluids in gneiss, and Yardley (1997) Monviso eclogite fluid. The hydration trend results from preferential removal of Cl relative to Br and I.

of Monviso (Philippot & Selverstone, 1991; Yardley, 1997), in which I contents exceed those of Br. The large Cl/I ratios in Bårdsholmen fluids might reflect the compositional characteristics of the infiltrating fluid, thus being externally controlled.

Whether a metamorphic rock is formed by hydration or by dehydration affects the Cl/Br systematics of the rock and the pore fluid. Figure 10 shows the compositional Cl–Br–I evolution of fluids produced during devolatilization reactions and fluids affected by hydration reactions. Fluids in rocks formed by hydration are subjected to a wide range of water activities, which can fractionate incompatible elements into minerals (e.g. Sanford, 1981). When H_2O is consumed by hydration reactions, the Cl/Br ratio will not change until the a_{H_2O} value is sufficiently lowered to form Cl-bearing minerals. Because the ionic radius of Br is larger than that of Cl, most minerals will not significantly incorporate Br into their structures, even at low a_{H_2O}. In contrast to hydration, dehydration reactions will release I from minerals and organic matter into the fluid phase, decreasing the fluid Cl/I ratio without significantly affecting the Cl/Br ratio. Determining sources for the eclogite-forming fluids is difficult, due to a lack of data for the fractionation of halogens between minerals and fluid, and limited knowledge about the convergence of halogen signatures. However, the most likely fluid source is amphibolite facies rocks undergoing prograde metamorphism, as suggested by Andersen *et al.* (1993).

Eclogite fluids

The setting at Bårdsholmen suggests fluid mobility at larger scales than for Alpine eclogites, where geochemical data indicate fluid mobility on the scale of centimetres, and where eclogites acted as closed systems with respect to fluids (e.g. Philippot & van Roermund, 1992; Selverstone *et al.*, 1992; Nadeau *et al.*, 1993). Philippot (1993) suggested a continuous recycling of fluids originally trapped by omphacite during eclogite-forming dehydration. Furthermore, deformation results in the local release of fluids, followed by trapping in veins. This mechanism cannot be evaluated by our data.

Fluids released during subduction are important catalysts for metamorphism, as recognized in the BAS (e.g. Austrheim, 1986; Jamtveit *et al.*, 1990). The chemistry of such fluids could be continuously modified by dehydration and hydration processes, as fluids are released and consumed along shear zones in subducted rocks. Fluid inclusion data from other Norwegian eclogites (Andersen *et al.*, 1993) suggest that fluids causing eclogitization of dry rocks originally comprised H_2O–CO_2–N_2, and that fluid immiscibility, hydration and/or partial melting resulted in an N_2–CO_2 residual fluid. Our data cannot exclude fluid immiscibility at deeper levels prior to eclogitization, but they do suggest that infiltrating brines caused eclogitization, and that
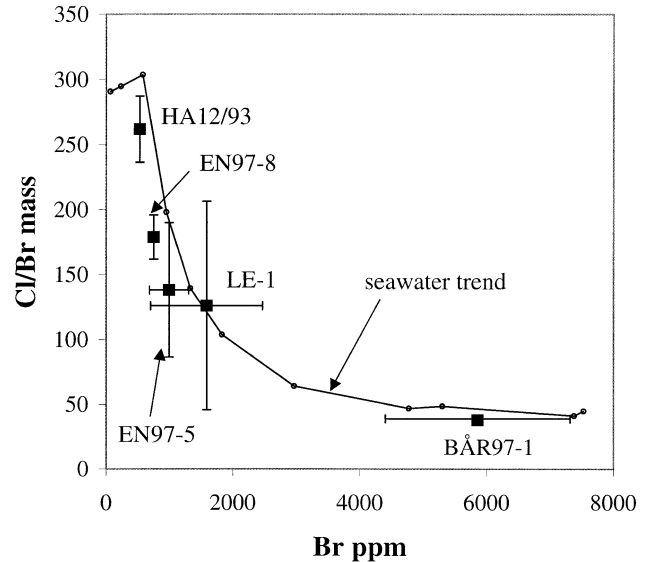

Fig. 11. Cl/Br–Br trend of evaporating seawater (data from Fontes & Matray, 1993) compared to average crush–leach and microthermometry data for omphacite-hosted fluids. It must be emphasized that the positions of the samples in this figure differ from those in a similar figure in Svensen *et al.* (1999). The reason for this is that the dataset is expanded in this paper, and there is some spread in the data.

their composition was modified during hydration. The abundance of elements such as Cu, Zn and Pb in the Bårdsholmen fluid inclusions (especially well recorded by step-daughter crystals) suggests that fluid–rock interactions create brines that resemble ore-forming fluids in their contents of metals and halogens.

The effect of hydration reactions on fluid composition

Figure 11 compares the Cl/Br vs. Br evolution of evaporating seawater with average compositions of fluid from samples BÅR97-1, EN97-5, EN97-8 and LE-1. The evolution of Cl and Br concentrations in fluids caused by hydration reactions appears to be similar to that in evaporating seawater, where H_2O is removed without changing the Cl/Br ratio until halite precipitates. This links the Cl/Br ratio to the water activity of the fluids, and suggests strong fractionations of Cl and Br between minerals and fluid. At room temperature, Cl and Br fractionate through halite precipitation [(Br/Cl)^{halite}/(Br/Cl)^{solution} = 0.032; Fontes & Matray, 1993]. Applying this ratio to fluids from sample BÅR97-1 (which display an average Br/Cl mass ratio of 0.027), the Br/Cl mass ratio in the 'average mineral' responsible for Br removal from the solution is 8.6×10^{-4} (or Cl/Br = 1157). If the Cl content of a mineral is between 1000 and 10 000 ppm (cf. Tables 2 & 3), the corresponding Br value would be 0.9–8.6 ppm. This is considerably higher than that of the average lower continental crust (Br = 0.28 ppm; Wedepohl, 1995), but in agreement the low Cl/Br ratios of the fluid inclusions.

CONCLUSIONS

1 The halogen composition of fluid inclusions hosted by omphacite and garnet in eclogites of the WGR/BAS likely represents the pore fluid composition at eclogite facies conditions. Microthermometry is of limited value in studying the compositions of alkali- and halogen-rich fluid inclusions in these eclogites.

2 Cl/Br ratios range widely among leaches from the studied eclogites, and the fluids with the lowest Cl/Br ratios show the highest Cl/I ratios. This halogen signature shows that I is depleted relative to Cl in the eclogite-hosted fluids, compared to standard reference crustal rocks, and might indicate a high mineral–fluid partitioning coefficient for I.

3 The halogen systematics of eclogite-forming pore fluids are affected by mineral–fluid fractionation. The lower crustal fluid source(s) cannot be determined unless we learn how Br and I behave under high pressure and temperature conditions. More information is needed on the halogen contents of lower crustal fluids, in order to determine end-member fluid compositions. However, metamorphic processes such as dehydration and hydration will result in differing Cl/Br vs. Cl/I evolution of fluids.

4 In the Bårdsholmen eclogites, phengite and apatite show the greatest F/Cl mineral–fluid partitioning and Cl-rich amphibole the least F/Cl mineral–fluid partitioning. The mass ratio of F to Cl in fluid inclusions is about two orders of magnitude less than that of amphibole.

5 The Cl/Br vs. Br characteristics of the fluid inclusions resemble the chemical evolution of evaporating seawater, which might provide a basis for studying halogen mineral–fluid fractionation even at high *P–T* conditions.

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