

# Chapter 11

## Prograde, Peak and Retrograde Metamorphic Fluids and Associated Metasomatism in Upper Amphibolite to Granulite Facies Transition Zones

J.L.R. Touret and T.G. Nijland

**Abstract** Granulites constitute a major part of the (lower) continental crust, occurring on a regional scale in many metamorphic belts. Their origin is generally discussed in terms of vapour-absent melting and fluid-assisted dehydration. This last model is notably supported by the occurrence of two immiscible free fluids at peak- and retrograde conditions, viz. CO<sub>2</sub> and highly saline brines. Evidence includes fluid remnants preserved in mineral inclusions, but also large scale metasomatic effects. The current paper discusses the presence and action of these fluids in granulites, with special attention to amphibolite to granulite facies transition zones (e.g. the Bamble sector, south Norway). Metasomatic effects induced by fluid percolation at different scales and stages include: (1) Control of state variables (H<sub>2</sub>O activity or O<sub>2</sub> fugacity), regional oxidation and so-called ‘granulite facies’ islands. (2) Small scale metasomatism at mineral intergrain boundaries (e.g. K-feldspar microveins and/or myrmekites). (3) Large scale metasomatism at the amphibolite to granulite facies transition zone, evidenced by: (a) Incipient charnockites, (b) Metasomatic redistribution of elements traditionally considered as immobile (e.g. Zr, Th, REE), (c) Peak metamorphic to retrograde bulk chemical processes (scapolitization, albitization), (d) Long distance action of granulite fluids. The importance and widespread occurrence of these effects call for large fluid quantities stored in the lower crust at peak metamorphic conditions, later expelled towards shallower crustal levels during retrogradation. Fluid origin, only briefly discussed in this paper, is complicated, not unique. Some fluids are crustal, either far remnants of sedimentary waters (brines) or linked to metamorphic/melt reactions. But, especially for high-temperature granulites, the greatest amount, notably for CO<sub>2</sub>, is issued from the upper mantle, which contain also the same fluid remnants as those found in the lower crust.

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## 11.1 Introduction

Granulites constitute a major part of the (lower) continental crust and are spread, on a regional scale, in many metamorphic belts, especially in Precambrian shield areas. They are typically characterized by water-free (pyroxene, garnet) or water-poor mineral assemblages. Though water-poor, granulites are by no means fluid-free rocks. Since the early 1970s, it has been claimed, notably by the senior author, that two immiscible free fluids are present at peak conditions, viz. CO<sub>2</sub> and highly saline brines (e.g. Touret 1985). These findings were in line with early suggestions by D.S. Korzhinskii, first proposed in 1940 and elaborated in a series of papers culminating in his 1959 English language treatise *Physicochemical basis of the analysis of the paragenesis of minerals*. Korzhinskii explained charnockite/granulite-forming mineral reactions by variation in chemical potentials, such as a high  $\mu_{\text{K}_2\text{O}}$  for the formation of K-feldspar and a low  $\mu_{\text{H}_2\text{O}}$  for the stabilisation of anhydrous mineral assemblages. He concluded that such variations are due to processes of fluid-rock interaction. This line of research has since been developed by many Russian geologists, on the basis of field observations, theory and experiments (see e.g. reviews by Perchuk and Gerya 1992, 1993). This model can be designed as *fluid-assisted metamorphism*. Fluids play an active role, being able to dissolve, transport and deliver chemical element to form minerals. In contrast, many Western geologists researchers have been inclined to favour a *vapour-absent* or *fluid-absent model* (*fluid-absent metamorphism*; Thompson 1983). In this model, no free fluid is present at peak metamorphic conditions, all fluid components (e.g. H<sub>2</sub>O for water-bearing mineral phases) are either bound in mineral structures or dissolved in melts. Though the presence of fluids during granulite facies metamorphism is still debated (e.g. Rigby and Droop 2011), it is clear, at least for present authors, that the so-called granulite controversy, which has lasted for more than 40 years now, has come to an end. Nobody denies that dehydration melting, the major process involved in vapour-absent models, is of major importance for the formation of many granulite facies terrains. But, as discussed in Sect. 11.3 of this chapter, dehydration melting is quite compatible with the occurrence of different low H<sub>2</sub>O-activity fluids. Two major fluids are found in granulites, viz. CO<sub>2</sub> and brines. An impressive set of arguments demonstrates that these occur in granulites at both peak and retrograde metamorphic conditions. They differ radically in terms of transport properties and the ability to dissolve solid phases (Newton and Manning 2010). Their occurrence, however, implies that large-scale, fluid-assisted mass transport of elements, called *metasomatism* by Naumann (1826), could occur. The relative roles of metamorphic versus metasomatic effects have been discussed since, e.g. Goldschmidt (1922), Ramberg (1951, 1952), Korzhinskii (1959) and Putnis and Austrheim (2010). However, if such metasomatic effects do exist, they provide (additional) proof of the reality of these fluids. Consequently, understanding the role and significance of these fluids is of major importance for the formation of the continental crust. To do so, it is necessary to first consider the position of granulites in a vertical section of the continental crust. Subsequently,

major evidence for the presence and action of fluids during granulite facies metamorphism will be presented and discussed as a basis for an integral understanding of granulite formation, paying special attention to the amphibolite to granulite facies transition zones. As the Bamble sector, southern Norway, provides one of the best records of fluids in granulites, we will describe it in several parts of this chapter, incorporating crucial and salient observations from other granulite facies terranes.

## 11.2 Background: A Granulite Continental Lower Crust

Compared to high pressure rocks within the eclogite facies, high temperature rocks comprise various types of granulites, from the (relatively) low pressure varieties (LP,  $P >$  about 5 kb,  $T >$  about  $750^{\circ}\text{C}$ ) to high temperature (HT) and ultra-high temperature (UHT) types, which can reach temperatures covering part of the magmatic range ( $> 1,000^{\circ}\text{C}$ ). Most common are intermediate granulites, with  $T$  about  $700\text{--}800^{\circ}\text{C}$ ,  $P = 7\text{--}8$  kb, occurring notably in large, regional-scale complexes (e.g. Harley 1993). Granulites, characterized primarily by water-deficient mineral assemblages (orthopyroxenes, clinopyroxenes, garnets; Fig. 11.1) are transitional to amphibolite facies rocks, notably the granitic migmatites, which are themselves the most commonly occurring igneous rocks in the continental crust. A great number of data, including P-T mineral estimates, LILE and trace element geochemistry as well as geophysical density and electrical conductivity measurements, suggest that the amphibolite and granulite facies correspond broadly to a vertical section of the continental crust (Belousov 1966; Rudnick and Fountain 1995).

Granitic migmatites are the most common rock type in the middle crust between approximately 5 km, the typical level of emplacement of most granites, and about 20 km (e.g. Guillot et al. 1995). The lower crust down to the Moho (generally at a depth of about 40 km under the majority of the continental cratons) is the domain of granulites. This implies that the latter constitute about one third of the entire continental land mass. Only small parts of this huge domain have reached the Earth's surface through orogenic uplift, often suffering intense deformation and retrogression. Granulites were for a long time considered as petrographical oddities, restricted to few exotic countries (India, Madagascar; e.g. Lacroix 1910). For instance, in Rinne's 1928 textbook, which served as the standard reference for all French petrologists until the late 1950s, the name charnockite is hardly mentioned (orthopyroxene-bearing granite). Granulites, to which Rinne devoted only two whole pages of description, are presented in a very confusing way. They are considered as purely magmatic rocks and defined as muscovite-bearing granites, following the French tradition of that time. However, at the same time, examples are given of typical granulites in current sense such as the Sri Lankan graphite-bearing granulites, as well as of textural features such as platy quartz that are typical for Saxonian granulites. Recognition of the geological



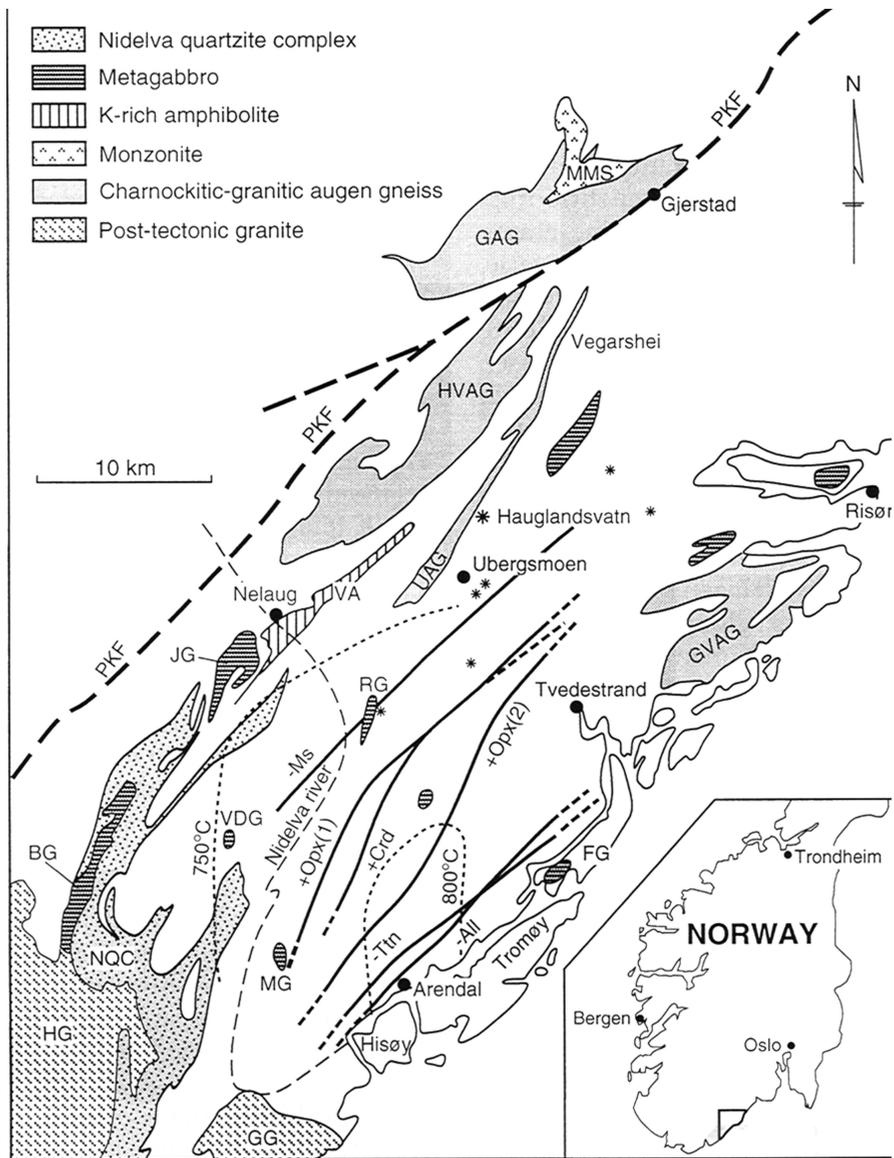
**Fig. 11.1** Example of typical granulite facies orthopyroxene-bearing veins at Hove, Tromøy, Bamble sector, Norway

importance of granulites occurred in the late 1970s, when it was progressively realized that the average composition of the continental crust was less magmatic (granitic) than had been assumed since the nineteenth century, but instead metamorphic, with remnants of supracrustal (i.e. sediments and eruptive rocks) that survived at the highest metamorphic grades. In the same period, deep crustal granulites were recognized amongst xenoliths from active volcanoes, in addition to a greater number of upper mantle rocks (e.g. Lovering and White 1969; Nixon 1987; Amundsen et al. 1988; Kovacs and Szabo 2005; Montanini and Harlov 2006), confirming the observations a century before from the French Massif Central (gneiss à pyroxène; Lacroix 1893). They document a systematic change in the composition of the granulite lower crust with depth (Bohlen and Mezger 1989; Bohlen 1991). The upper part is purely metamorphic, corresponding to regionally exposed granulite complexes, whereas recrystallized magmatic rocks, mostly gabbroic in composition (two pyroxene-granulites), dominate the lower part down to the upper mantle (Moho). The source of these gabbroic rocks is upper mantle plagioclase- or spinel-lherzolites residing at depths not exceeding 60–70 km. The timing of the emplacement of these magmas is often close to peak metamorphism, e.g. the French Massif Central (Vielzeuf and Pin 1989). Such intrusions provide the required heat for HT and UHT-metamorphism. As will be discussed below, we believe that they also play a key role in establishing the fluid regime in this part of the crust.

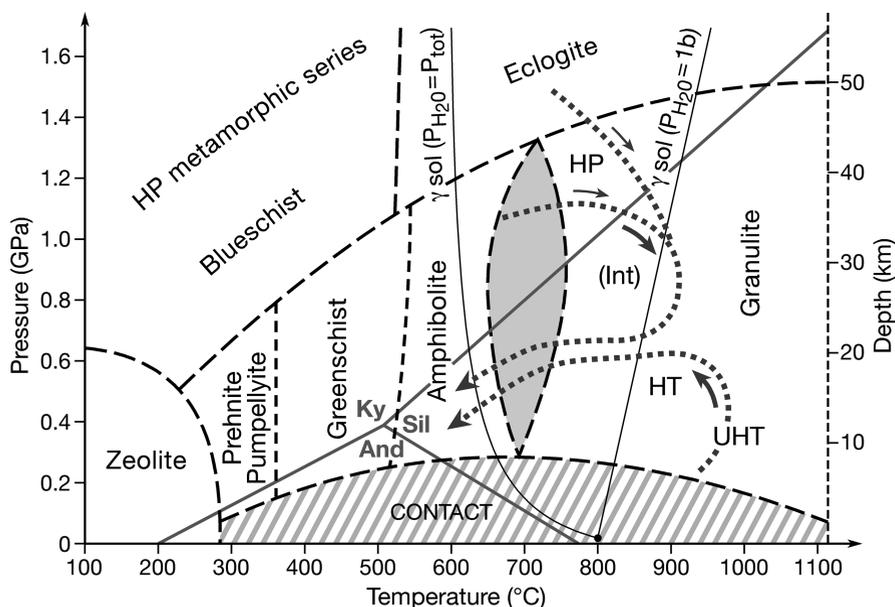
The transition zone between amphibolite and granulite facies rocks is quite complicated. It is marked by a number of prograde mineral isograds, corresponding to the (dis)appearance of diagnostic minerals, e.g. muscovite-out, biotite-out, orthopyroxene-in, cordierite-in, garnet-in, in terranes like the Adirondacks, US

(Engel and Engel 1958, 1960), Broken Hill, Australia (Binns 1964), the Bamble sector, Norway (Touret 1971; Nijland and Maijer 1993) and the classical granulites from Saxony (Rötzler 1992) or even osumilite-in in UHT granulites, like Rogaland, Norway (Tobi et al. 1985). Other index minerals in progressive granulite facies metamorphism include the more localized occurrence of minerals like sapphirine, spinel + quartz, kornepupine, etc. Additional petrological indicators that can be used include the degree of partial melting, the occurrence of quartz-garnet symplectites in some mobilisates, etc. However, the interpretation of these features in terms of variable P and T is even more difficult than that of the mineral reactions, though the latter is far from being straight forward (Chap. 3). Most minerals are complex solid solutions connected to each other by multivariant equilibria. The large number of chemical and physical variables affecting an isograd can lead to complicated map patterns with intersecting isograds (Fig. 11.2). In some cases, it is hard to identify whether these isograds are prograde (transition amphibolite to granulite) or retrograde (retrogressed granulite). Identification is only possible if the relationships between the consumed phases and the product phases are unambiguous. In some areas, the amphibolite to granulite facies transition is considered to be a retrograde feature (e.g. the Limpopo belt; Van Reenen 1986; Ungava orogen, Canada; St Onge and Lucas 1995). The complex nature of the granulite facies is clear on a conventional P-T diagram (Fig. 11.3). Whereas the boundaries separating high pressure facies (blueschist and eclogite) from high temperature facies (greenschist, amphibolite and granulite) are relatively well defined, boundaries delimiting the granulite facies are never represented by a sharp line, but by a rather broad, diffuse band.

On a regional scale, it is clear that temperature plays an important role in the amphibolite to granulite transition. Overall, temperature tends to be higher in granulites than in neighbouring amphibolites. In detail, however, the situation is often much more complicated, as illustrated by the case of the Bamble sector, southern Norway (Fig. 11.2). Here, mineral isograds differ grossly from isotherms, maximum temperatures (above 800°C, typical for most intermediate pressure granulites) being reached in a dome situated in the northern part of the granulite core east of the town of Arendal extending in this direction within the neighbouring amphibolite (Nijland and Maijer 1993). This situation is by no means exceptional. Even if it has been rarely documented in detail, it is our experience, from various field studies in Finland, Sweden and the Variscan belt that it occurs in many granulite facies terranes worldwide. It is for this reason that, on Fig. 11.3, we have represented the granulite and amphibolite fields as partly overlapping (shaded area on Fig. 11.3). In this zone, another variable of state is required to initiate and control the transition from amphibolite to granulite facies mineral assemblages. This variable is  $P_{\text{H}_2\text{O}}$  as most reactions involve dehydration. It is significantly lower than lithostatic pressure in the granulite facies terrane, whilst nearly equal to the lithostatic pressure in the amphibolite domain.



**Fig. 11.2** Generalized metamorphic map of the Bamble sector, south Norway (Nijland et al. 1998), indicating metamorphic isograds (continuous lines, + = appearance, - = disappearance of diagnostic minerals; +Opx(1): in basic rocks, +Opx(2): in metapelites) and isotherms (dashed lines), as well as so-called granulite facies islands (stars; see text). Geological units: BG Blengsvatn gabbro, FG Flosta gabbro, GAG Gjerstad augen gneiss, GG Grimstad granite, GVAG Gjeving augen gneiss, HG Herefoss granite, HVAG Hovdefjell-Vegårshei augen gneiss, JG Jomåsknutene gabbro, MG Messel gabbro, MMS Morkheia monzonite suite, NQC Nidelva quartzite complex, PKF Porsgrunn-Kristiansand fault, RG Ripåsen gabbro, VA Vimme amphibolite, VDG Vestre Dale gabbro



**Fig. 11.3** Major metamorphic facies in P-T space for  $P_{H_2O} = P_{Total}$ , except in granulite; this explains the partial overlap of amphibolite and granulite facies, indicated as a *shaded area* on the figure. In this portion of an amphibolite to granulite facies transition zone is not explained by an increase of temperature but by a decrease of  $P_{H_2O}$  relative to  $P_{Total}$ . Subdivisions of the granulite facies are indicated as *HP* high pressure, *HT* and *UHT* high to ultrahigh temperature, and *Int*-Intermediate. *Dots with arrows* indicate typical P-T paths, either clockwise (in HP-granulites) or anticlockwise (in HT-UHT-granulites)

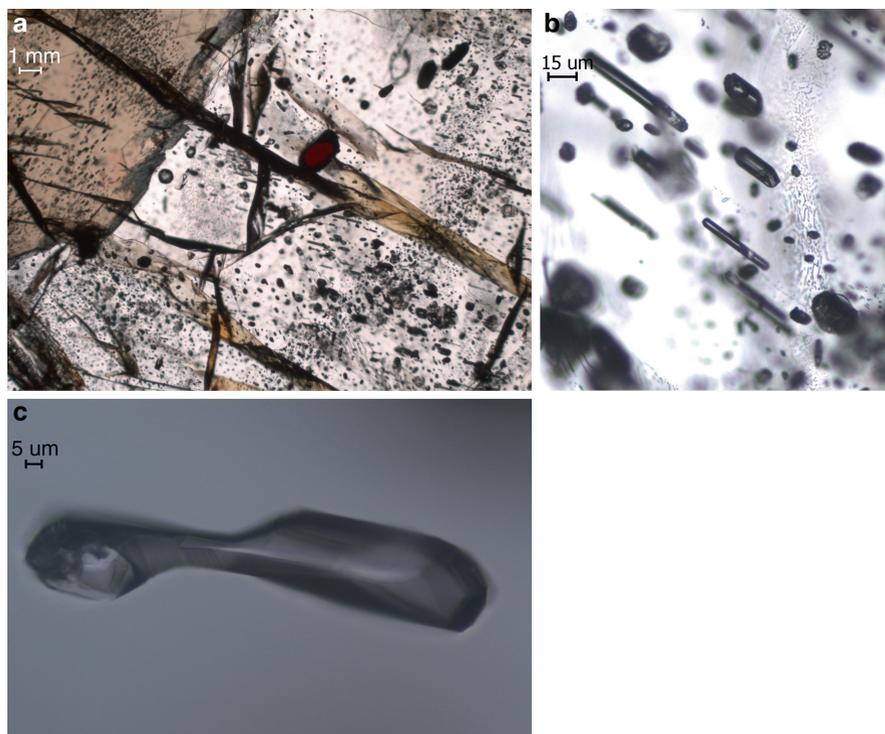
### 11.3 Granulites: Vapour-Absent Melting or Fluid-Assisted Dehydration?

P-T conditions for both upper amphibolite and granulite facies rocks are all above the hydrous granite minimum melting curve. These rocks are the source of crustal granites (typically S-types) which, when formed in the amphibolite facies domain, are  $H_2O$ -saturated and crystallize rapidly above the migmatite source (midcrustal granites). Water undersaturated lower crustal granites, on the other hand, may rise to higher levels, reaching the upper crust or even the Earth's surface in the form of rhyolite eruptions (Clemens 2006). Being able to dissolve and transport fluids, notably  $H_2O$ , these granitic magmas obviously can play an important role for regulating the fluid regime at peak metamorphic conditions. Following a seminal paper by W.S. Fyfe (1973) on the Lewisian complex in Scotland, a first, purely magmatic model was proposed for the formation of granulites, in line with ideas prevailing during the first half of the twentieth century (see above). This model was later extended to many other terranes including Broken Hill, Australia (Phillips 1980),

New England, US (Tracy and Robinson 1983) and Namaqualand, South Africa (Waters 1988).

The model of vapour-absent (dehydration) melting involves breakdown of hydrous phases such as muscovite, biotite and amphibole, releasing water as well as other interstitial fluids (e.g. Le Breton and Thompson 1988). The released water is partitioned into melts and subsequently removed by melt extraction. In this hypothesis, granulites are restites, with no fluids remaining at peak metamorphic conditions (e.g. Vielzeuf et al. 1990). This model is supported by a number of microstructural, experimental, and theoretical investigations (e.g. Thompson 1982; Brown and White 2008). Being purely magmatic, it does not leave much room for any large scale metasomatic processes. Only limited diffusion metasomatism on the millimetre to centimetre scales between coexisting phases of contrasting compositions, permitted by high metamorphic temperatures, is possible. These include corona textures, spectacular in some high temperature (HT) granulites, but even more developed in high pressure metamorphic rocks (HP granulites, eclogites). For this reason, also because they are well discussed in the literature (e.g. Yardley 1989; Harley 1989), as well as in Chaps. 5 and 10 of this volume, these textures will only be marginally considered in this chapter. Moreover, at least in some cases, including the coronitic gabbros of southern Norway (formerly referred to as hyperites), compelling evidence suggest that in some of these gabbros the coronas are caused by interaction with residual melts, rather than being of metasomatic origin (Joesten 1986; De Haas et al. 2002).

The discovery of low water activity fluids ( $\text{CO}_2$ , concentrated brines) included in many granulite minerals (Touret 1971, 1972, 1973; Figs. 11.4, 11.5), combined with theoretical and experimental work done by D.S. Korzhinskii and his followers (e.g. Aranovich et al. 1987) has led the other model for granulite genesis, namely *fluid-assisted dehydration* (e.g. Janardhan et al. 1979; Newton et al. 1980). Under the influence of these low water activity fluids, hydrous mineral such as muscovite, biotite, amphiboles breakdown to granulite mineral assemblages whilst producing water. The water produced becomes diluted. This model does not negate partial melting at all. In fact, partial melting is an efficient way to produce some of these low water activity fluids such as  $\text{CO}_2$ , either due to preferential dissolution of  $\text{H}_2\text{O}$  into granitic melts (Kadik et al. 1972), reaction of graphite (former organic matter) with  $\text{H}_2\text{O}$  (Touret and Dietvorst 1983) or possibly residual brines. There is no doubt that some melts are extracted leaving behind residual granulites, especially in zones of intense shearing and deformation. However, the astonishing preservation of delicate sedimentary structures such as graded or cross bedding, mudcracks and conglomerates in quartzitic sediments in the transition zone (Nijland et al. 1993) and even in migmatitic quartzo-feldspathic gneisses where partial melts remained in situ (e.g. Selås gneisses, Touret 1966) shows that, even in the most structurally complicated terranes, strain-free domains exist, in which post-crystalline deformation is virtually absent. These features illustrate the metasomatic character of the amphibolite-granulite transition, namely mineralogical changes at nearly constant volume without melting. This is illustrated by an impressive series of features including granulite oxidation states, grain boundary microtextures and

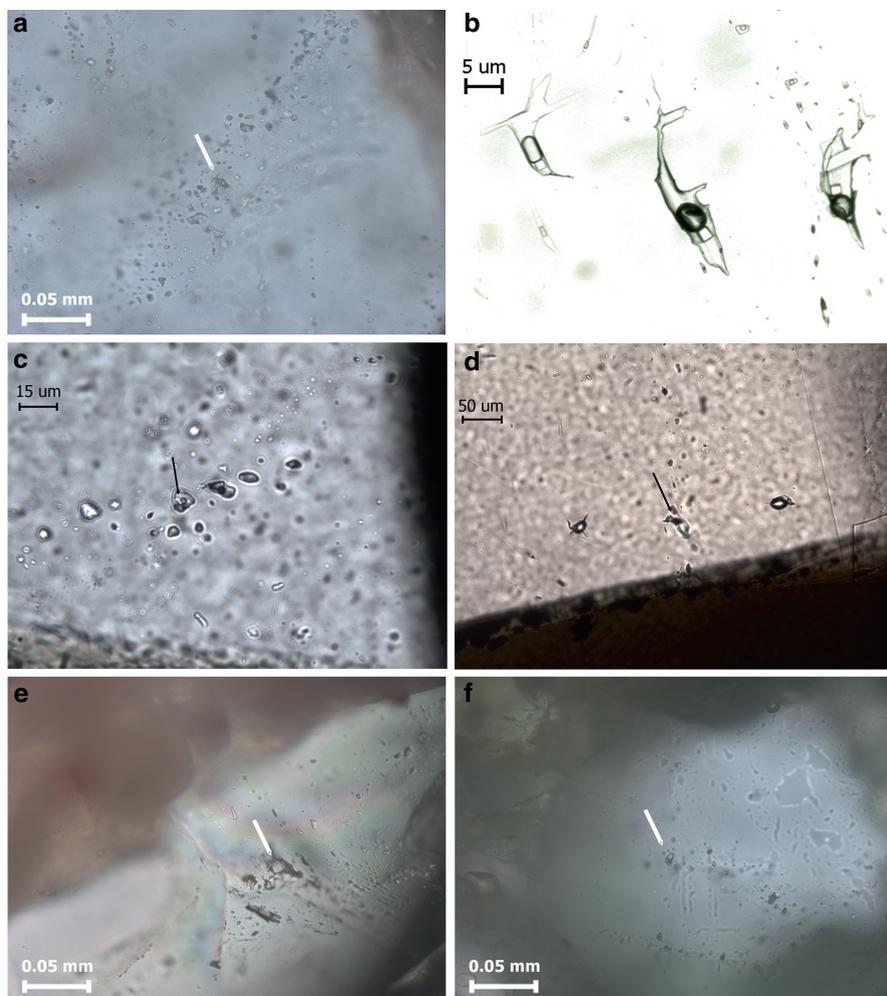


**Fig. 11.4** Examples of CO<sub>2</sub>-rich fluid inclusions in sapphirine-bearing granulites from central Sri Lanka (Bolder-Schrijver et al. 2000). (a): Overall view of major rock-forming minerals in thick section with garnet to the *right* and orthopyroxene to the *left*. (b): Array of tubular primary inclusions in garnet; *dark* inclusions are filled by high-density CO<sub>2</sub>, *white* inclusions are daughter crystals of magnesite; detail of CO<sub>2</sub>-inclusion in garnet

incipient charnockites (see below). The scale and extent of these features requires infiltration (percolation) metasomatism in the sense of Korzhinskii (1959, 1968). It also requires a number of necessary conditions, some of them at odds with commonly accepted hypotheses, notably by advocates of vapour-absent models:

- A significant quantity of fluids is present in the lower granulite crust (and in the underlying mantle) at peak metamorphic conditions.
- These fluids are able to move through the rock masses, at a level where, classically, no fluid movement is supposed to occur, except along rare, discrete shear zones (Etheridge et al. 1983).
- The fluids can have a profound geochemical action, either in the control of some variables of states (first of all P<sub>H<sub>2</sub>O</sub>) or in dissolution and transport of chemical components involved in metasomatic reactions and mineral genesis.

Except for the minute remnants preserved as fluid inclusions in minerals, most deep fluids have left the rock system before eventual uplift and exposure at the



**Fig. 11.5** Examples of brines inclusions in granulites and associated rocks from the Bamble sector, Norway. (a): brine inclusion from a so-called granulite facies island, Hauglandsvatn (Nijland et al. 1998). (b): brine inclusion in quartz-actinolite rock. (c,d): brine inclusions in cordierite-anthophyllite rock hosting giant myrmekites from Åkland with C a squeezed brine inclusion in cordierite accompanied by small solid inclusions of zircon and D in quartz blebs in cordierite. (e): brine in apatite from the former Lykkens mine, Kragerø. (f): brine inclusion in scapolite in rutile-scapolite-pargasite rock from Ødegårdens Verk

Earth's surface. But, as discussed in detail below, the existence and abundance of fluids in granulite facies is documented by the numerous traces that they have left in the rocks. Description of these features is the core of this chapter. However, it is first necessary to justify the presence of these fluids, as they are critical for the existence of metasomatic processes at this deep level of the continental crust.

## 11.4 Free Fluid Phase During Granulite Metamorphism

### 11.4.1 Fluid Inclusion Data

The revolutionary idea that some free fluids can exist at peak metamorphic granulite conditions, at a temperature of 800–1,000°C and a depth of many 10s of kilometres, come primarily from fluid inclusion data, first described in southern Norway (Touret 1971), then in virtually all granulite facies terrains (Newton 1989). Ignoring minor components such as N<sub>2</sub> or CH<sub>4</sub>, found in some high grade terranes, two types of fluids are systematically observed:

- Pure CO<sub>2</sub> of variable density (Fig. 11.4), reaching more than 1.1 g cm<sup>-3</sup> in the case of ultradense inclusions as described by Van den Kerkhof and Olsen (1990) from the Colorado Front Range, USA, from many localities in India including Dodabetta (Touret and Hansteen 1988; Srikantappa et al. 1992), Nuliyam, Kerala (Fonarev et al. 2001), eastern Ghats (Mohan et al. 2003), Salem area south of the Shevaroy Hills (Santosh and Tsunogae 2003), Nagercoil (Santosh et al. 2003) and the Chottanagpur complex (Mishraa et al. 2007), as well as the Napier complex, Antarctica (Tsunogae et al. 2002), In Ouzal, Algeria (Cuney et al. 2007) and the Gandese charnockites, southern Tibet (Zhang et al. 2011).
- Highly saline aqueous inclusions (brines; Fig. 11.5), typically with 20–30 mol% equivalent NaCl, approaching pure salt in a few extreme cases (Touret 1985; Frost and Touret 1989). In the literature, brine inclusions are much less frequently reported than CO<sub>2</sub>-bearing ones. However, brines also occur in classic granulite areas from which up to recent only CO<sub>2</sub>-bearing ones have been described, like southern India (Srikantappa and Arash Zargar 2009). It is noteworthy that rather similar fluids have also been encountered in rocks from deeper levels in the mantle (Izraeli et al. 2001; Klein-BenDavid et al. 2004).

It has been argued that the CO<sub>2</sub> (brines were not considered at this time) represents late fluids, unrelated to granulite metamorphism (Lamb et al. 1987). It is true that both fluids are constantly found during all the steps of retrograde metamorphic evolution. In fact, recent detailed investigations of the Bamble sector rocks (Sørensen 2007) show that they act as a sort of shield, protecting high grade mineral assemblages against surficial retrogression. Traces of this superficial retrogression, due to encounters with surface waters during the final stages of uplift, are quite widespread. This retrogression is, for example, responsible for the fact that French petrologists have ignored until very recently that the southern Vosges and Massif Central are typical granulite provinces. They considered only relatively late, intrusive granites, that make up only a small fraction of the exposed rocks.

But, as discussed in many papers (Coolen 1980, 1982; Touret and Hartel 1990; Santosh 1992; Santosh et al. 1991), the reality of synmetamorphic granulite fluids can hardly be questioned. Their existence is proven by a set of three, concordant arguments: synchronism of inclusion formation and mineral crystallization (primary inclusions), concordance between CO<sub>2</sub> density (isochores) and P-T mineral

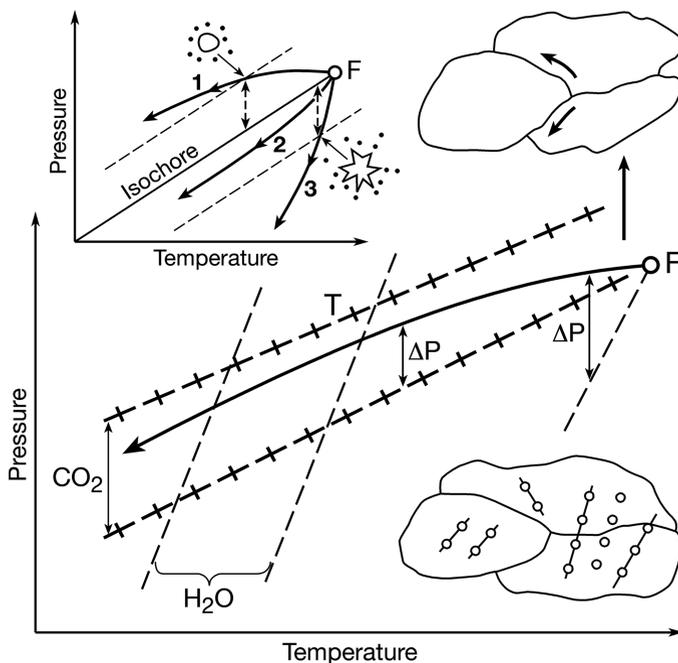
data, and fluid compositions in agreement with those predicted from mineral assemblages (Touret 2009). Contrary to conventional wisdom, quartz is not the best mineral host in granulites. Other minerals (garnet, feldspars) may contain many more, larger and better preserved inclusions than the neighbouring quartz (Srikantappa et al. 1992).

Not only fluid inclusions demonstrate the presence of a free fluid phase during granulite facies metamorphism. Mineral assemblages may also indicate the former presence of free CO<sub>2</sub> and/or saline solutions. This holds notably for CO<sub>2</sub>-scapolite-bearing granulites (Goldsmith 1976; Aitken 1983; Moecher and Essene 1990, 1992; Moecher 1993) which have been described from a number of granulite terranes, including SW Finland (Parras 1958), Furuu, Tanzania (Coolen 1980, 1982; Hoefs et al. 1981), the Aldan shield, Siberia (Perchuk et al. 1985), Fiordland, New Zealand (Bradshaw 1989) and the Kohistan arc, NW Himalayas (Yoshino and Satish-Kumar 2001). Though once considered as common (e.g. Turner and Verhoogen 1960), the number of documented occurrences is relatively small, possibly indicating that most CO<sub>2</sub>-bearing scapolites were destroyed during uplift, leading to liberation of CO<sub>2</sub> during this stage of evolution.

Anyone studying fluid inclusions will immediately find that if remnants of both CO<sub>2</sub> and brines can be found in most granulites, their relative abundance and state of preservation are rather different. CO<sub>2</sub> may occur as large, spectacular inclusions several tens of micrometres in size and often negative-shaped (Fig. 11.4). Most granulite complexes show post-metamorphic P-T paths with generally isobaric cooling, i.e. roughly parallel to high density CO<sub>2</sub> isochores, favouring the preservation of CO<sub>2</sub> inclusions (cf. Touret and Huizenga 2011) (Fig. 11.6). Brines, on the other hand, occur only in minute inclusions of only a few micrometres, with an irregular cavity often squeezed around one of several solids (halides) (so-called collapsed inclusions; Touret 2001) (Fig. 11.5). The steeper brine isochores cause these inclusions to be constantly underpressured and transposed until final decompression, which typically occurs at temperatures of about 300–350°C (Fig. 11.6).

### ***11.4.2 Deep Fluids Can Move Through the Rocks***

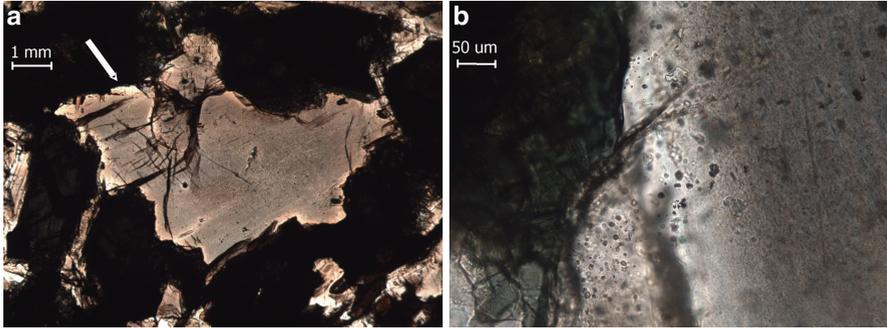
Permeability estimates of the lower crustal conditions are very low (Fyfe et al. 1978), which is apparently incompatible with large fluid movements through these rocks. Nevertheless, focussed fluid flow has been demonstrated to occur at all levels (e.g. Ague 2003 and references therein). The absence of large fluid movements in the lower crust does not comply with some common observations on fluid inclusions. Most granulites contain numerous trails of secondary inclusions, most of them CO<sub>2</sub>, trapped over a wide P-T range. Detailed observations on the CO<sub>2</sub> density evolution show that it follows the inferred P-T path closely, with a density increase for relatively late inclusions in the case of isobaric cooling and a density decrease in the case of isothermal decompression (Touret 2001). This indicates that significant quantities of dense CO<sub>2</sub> were able to move through the rocks through



**Fig. 11.6** Diagram illustrating how deep fluid inclusions can (or cannot) reach the Earth's surface. *Crosses and dashed lines*: relevant  $\text{CO}_2$  and  $\text{H}_2\text{O}$  isochores, respectively; *solid lines (with arrow)*: postmetamorphic P-T paths. At any temperature, the distance between both lines defines a pressure difference ( $\Delta P$ ) which, when exceeding the strength of a mineral host, leads to inclusion rupture, either by implosion (collapsed inclusions; path 1 (insert)) or explosion (decrepitation; path 3 (insert)). If a P-T path remains close to an isochore (path 2 (insert)), inclusions may remain undisturbed until the surface. On the insert, F indicates an inclusion formation at peak P-T conditions, whereas the grain sketches show typical ways of fluid migration through rocks at high P-T:  $\text{H}_2\text{O}$  moves along interstitial grain boundaries;  $\text{CO}_2$  moves through successive generations of microfractures, forming trails of secondary inclusions

a system of microfractures, which were successively opened and then healed after a short period of time (*fluid-induced fracturing*, Putnis and Austrheim 2010, Chap. 5). In many cases, most inclusions belonging to the same fracture represent a group of synchronous inclusions (GSI), i.e. they have formed at almost the same time, as shown by the same homogenisation temperature with a typical variation of only a few degrees Celsius (Touret 2001).

Healing occurred at nearly constant temperature by the solubility change induced by surface tension effects in very narrow cavities (Lemlein 1951). Most fluids initially contained in the open crack are expelled during this healing, causing the formation of another microcrack in the neighbourhood. But, due to the large wetting angle of  $\text{CO}_2$  (Holness 1992; Watson and Brenan 1987; Gibert et al. 1998), some inclusions will remain regularly aligned in the plane of the former fissure, forming the typical trail that we see today. It is much more difficult for  $\text{CO}_2$



**Fig. 11.7** Metagabbro from the Anápolis-Itaçu complex, Brazil: *Left*: Magmatic plagioclase appearing grey with along its margin clear white metamorphic plagioclase (*arrow*). *Right*: detail of the recrystallized margin with CO<sub>2</sub> fluid inclusions appearing as *black dots*

than H<sub>2</sub>O to travel along grain boundaries, as noted by many authors (e.g. Watson and Brenan 1987). It is, however, to a limited extent not impossible as is illustrated by many granulite facies metagabbroic rocks. In many of deep-seated gabbroic and anorthositic intrusions transformed into two-pyroxene granulites during metamorphism, magmatic feldspars are still present, as indicated by numerous inclusions of ilmenites and other Fe-Ti oxides. A good example is the metagabbro shown in Fig. 11.7, from the Anápolis-Itaçu complex in central Brazil, a key locality for the characterization of UHT granulite metamorphism (Harley 2008). In this metagabbro, magmatic plagioclases, coeval with granulite metamorphism are very dark, due to a great number of minute opaque ilmenite inclusions (Della Giustina et al. 2010) (Fig. 11.7a). In contrast, recrystallized metamorphic plagioclase, formed along the borders or major fractures of magmatic crystals, is very clear and free of solid inclusions. It contains a number of minute, primary CO<sub>2</sub> inclusions, which only occasionally grade into the periphery of the magmatic plagioclase in the form of very small, pseudo-secondary trails (Fig 11.7b). It is interesting to note that the Anápolis-Itaçu case is precisely a case of *fluid-resaturation* of an initially dry, CO<sub>2</sub>-undersaturated, mineral assemblage (Table 4 in Rigby and Droop 2011). This shows that CO<sub>2</sub>-rich fluids, which are normally much less mobile than brines at intergrain boundaries (see below), can move to a certain extent along grain boundaries, provided that fluid movement is accompanied by sufficient recrystallization of the mineral host. Also granitic melts may, to some extent, dissolve CO<sub>2</sub> (Eggler and Kadik 1979). This implies that migmatitic leucosomes may provide a transport medium (e.g. Frost and Frost 1987). The occurrence of dense CO<sub>2</sub> inclusions in granites contemporaneous with granulite facies metamorphism (Santosh et al. 2005) shows that this may be of significance in at least some granulite terrains.

Brines move in a very different way than CO<sub>2</sub> (cf. Gibert et al. 1998). During typical post-metamorphic granulite isobaric cooling paths, brine inclusions behave differently than CO<sub>2</sub> inclusions due to the different slopes of the H<sub>2</sub>O and CO<sub>2</sub> isochores (Fig. 11.6). Also, the very small wetting angle of brines prohibits the

formation of secondary trails, in contrast to CO<sub>2</sub>. Compared to CO<sub>2</sub>, brines exhibit high mobility along grain boundaries, as evidenced by the numerous trails that they have left within rocks. With this respect, it is interesting to recall the so-called catastrophic textures described by Trommsdorff and Skippen (1986). These consist of a sudden increase in the mineral grain size, sometimes by one or two orders of magnitude, at places where concentrated brines, originating from fluid immiscibility, occur in high grade rocks. This role of brines, i.e. favouring large crystal sizes, illustrates the large capacity of element transport of this medium, quite different in this respect of the more *inert* CO<sub>2</sub> fluids (Newton and Manning 2010).

## 11.5 Metasomatic Effects Induced by Fluid Percolation

The fact that CO<sub>2</sub> and brines may have been present at granulite conditions and able to move through rock masses, notably during the early stages of post-peak metamorphic uplift, implies that they can induce large-scale metasomatic effects. This requires that mineral solubilities are high enough at the P-T conditions of interest to cause mineral transformations through dissolution-recrystallization/reprecipitation processes (Putnis 2002, 2009, Chap. 5). Solubilities of rock-forming minerals in CO<sub>2</sub> are generally very low. For example, quartz solubilities can only be measured in H<sub>2</sub>O + CO<sub>2</sub> fluids, not in pure CO<sub>2</sub> (Novgorodov 1977; Newton and Manning 2010). The CO<sub>2</sub> fluid can only act as a neutral component in the overall fluid. The primary silicate solvent in the fluid is the H<sub>2</sub>O component. However, the presence of alkali carbonates, such as nahcolite, NaHCO<sub>3</sub>, in carbonic inclusions from several terrains, e.g. In Ouzal, Algeria (Cuney et al. 2007), demonstrates the capability of this fluid to transport alkali, as well as trace elements commonly associated with carbonates, such as REE (e.g. Kontak and Morteani 1983), with a preference of the HREE over the LREE (Hellman et al. 1979; Humphries 1984; Michard and Albarède 1986).

In contrast to CO<sub>2</sub>, mineral solubilities can be quite high in brines, as documented by the ongoing work of the UCLA group (Newton and Manning 2002, 2006, 2007, 2008, 2010). Presence of NaCl in the fluid will enhance solubility of phases like monazite and xenotime (Tropper et al. 2011). If P and T variations are large enough, – a necessary condition for some minerals to be dissolved and others to be precipitated–, metasomatic processes may leave their tracks within rocks. The mechanism behind mineral re-equilibration in these metasomatic reactions is covered extensively by Putnis (2002, 2009), Putnis and Austrheim (2010) as well as in Chap. 5 of this book. There are many lines of evidence for metasomatic alteration in granulite facies rocks, which can occur on all scales, from minute structures at intergranular boundaries to larger scale replacement of entire rock masses. Previously, they have either been ignored or considered to be limited to a few odd areas. Nowadays, such metasomatic effects have been documented from many areas and are especially obvious in the amphibolite to granulite facies transition zone.

Evidence for metasomatism will be described in an order which reflects an increasing scale of alteration:

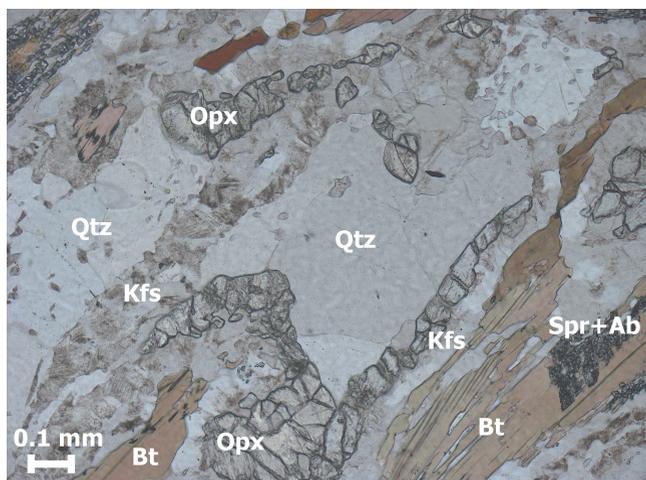
- Control of state variables ( $H_2O$  activity or  $O_2$  fugacity).
- Small scale metasomatism at intergranular boundaries (myrmekites, K-feldspar microveins)
- Large scale prograde and retrograde metasomatism in the amphibolite to granulite facies transition zone:
  - Incipient charnockites
  - Metasomatic redistribution of elements traditionally considered as *immobile* (e.g. Zr, Th, REE)
  - Peak metamorphic to retrograde bulk chemical processes (scapolitization, albitization)
- Long distance action of granulite fluids by fluid focussing in shear zones.

### ***11.5.1 Control of the Variables of States, Regional Oxidation, and Granulite Facies Islands***

One of the most obvious effects of granulite facies fluids is the control of some variables of state, first of all the partial pressure of  $H_2O$ , which for low values (typically up to about 0.3 of  $P_{Total}$ ) is compatible with granulite facies mineral assemblages. This can be realized by the presence of a low  $H_2O$  activity fluid phase, rapidly released during prograde metamorphism (Chap. 14), as argued above either  $CO_2$ -rich fluids or concentrated brines. The widespread occurrence of  $CO_2$  within the rock masses, not exclusively along grain boundaries, suggests that, within granulite domains, a low  $H_2O$  activity is mainly due to dilution by  $CO_2$ . The scarcity of adequate mineral reactions, as well as required  $CO_2$  amounts, requires an external origin for this fluid. Demand for an external source is illustrated by the Bamble sector, south Norway, in which  $CO_2$ -bearing fluids are a prominent feature, but which is almost devoid of carbonate rocks whose decarbonation could have provided free  $CO_2$ . Many arguments, including C- or He-isotopes (e.g. Pineau et al. 1981; Dunai and Touret 1993; Santosh et al. 1993), can be made that the source is mainly the mantle (see discussion in Touret 2009). This  $CO_2$ -rich fluid is then transported into the lower crust by synmetamorphic intrusions during peak metamorphic granulite facies conditions (e.g. Frost et al. 1989; Farquhar and Chacko 1991; Hansen et al. 1995). These intrusions contribute to the high temperatures reached during granulite facies metamorphism, especially for HT or UHT-types (Kelsey 2008; Touret 2009). It had been argued that infiltration by  $CO_2$ -rich fluids should lead to the deposition of large quantities of graphite (Lamb and Valley 1984, see also discussion in Touret 2009). This may account for some of the major graphite deposits found worldwide, e.g. in the Adirondacks, New York State, USA (Ticonderoga; Luke et al. 1998) and Sri Lanka (Katz 1987). On a smaller

scale, graphite-bearing hydrothermal veins in New Hampshire, USA, have been ascribed to the reduction of infiltrating CO<sub>2</sub>-rich fluids that resulted in local granulite facies hot spots (Chamberlain and Rumble 1989; Rumble et al. 1989). The reducing trends are, however, rarely evident. CO<sub>2</sub> itself has some oxidizing capacity (Frost 1991) and its introduction at high temperature will, on the contrary, result in a high degree of oxidation, typical for many granulite terranes (Harlov 2000). This has notably been demonstrated along a traverse of Archean lower crust in southern India including the Shevaroy Hills, southern India (Harlov et al. 1997; Harlov and Hansen 2005). Other examples include the granulite facies rocks of the Bamble sector, south Norway, where magnetite-hematite and ferrosilite-magnetite-quartz oxygen barometers show that peak metamorphic oxidation states were well above fayalite-magnetite-quartz buffer (Harlov 1992, 2000). Considerations on the buffering capacity of the oxygen fugacity of the C-H-O system (Huizenga 2005; Huizenga and Touret, 2012) show that this high degree of oxidation can be understood if CO<sub>2</sub> is introduced into the rock system at high, magmatic or submagmatic temperatures, well above the graphite stability curve. Except in a few cases, i.e. the above mentioned graphite deposits, the fluids will not re-equilibrate during retrogression; firstly, because of the difficulty for graphite to nucleate; secondly, because most of the free fluids probably leave the rock system during the first stages of the retrogression, at temperatures still too high for graphite to crystallize. Recent modelling by J.M. Huizenga on the C-O system shows that if CO<sub>2</sub> is introduced in completely dry rocks (hence the absence of H<sup>+</sup>), the quantity of graphite which can be deposited is extremely small, leaving excess oxygen which can then be responsible large scale oxidation effects (Huizenga and Touret, 2012). It should be also noted that, in addition to CO<sub>2</sub>, fluids may introduce other oxidizing components such as SO<sub>2</sub> (Cameron and Hattori 1994; Harlov et al. 1997; see also discussion in Newton and Manning 2005). The presence of SO<sub>2</sub>-bearing fluids in (some) granulites is also indicated by assemblages of sulfate scapolites, reported from, amongst others, Ghana (Von Knorring and Kennedy 1958), xenoliths in deep seated pipes from eastern Australia (Lovering and White 1964) and the Kohistan arc, NW Himalayas (Yoshino and Satish-Kumar 2001).

As far as the stability of granulite mineral assemblages is concerned, the role of brines is significantly different. An important feature is the development of so-called granulite facies islands, i.e. scattered, isolated occurrences of granulite facies rocks well beyond the orthopyroxene-in isograds for both basic and felsic rocks in the amphibolite to granulite facies transition zone of the Bamble sector, southern Norway (Nijland et al. 1998) (Fig. 11.2). The occurrences are not related to magmatic intrusions that might have provided a local additional heat source. They have up to now been only documented in this terrain, but likely occur in other amphibolite to granulite facies transition zones too. These granulite facies islands are characterized by diagnostic granulite facies minerals like orthopyroxene + K-feldspar (Fig. 11.8) and sapphirine + quartz in ordinary metapelitic rocks. This common bulk chemistry distinguishes these granulite facies islands from granulite facies mineral assemblages that may occur in very Mg-rich lithologies like cordierite-orthoamphibole rocks (e.g. Munz 1990; Visser and Senior 1990) or



**Fig. 11.8** Orthopyroxene-K-feldspar coronas between biotite and quartz, formed in response to saline brines, Hauglandsvatn, Bamble, Norway (See Nijland et al. 1998)

Mg-rich assemblages in scapolitized metagabbros (Engvik and Austrheim 2010; Engvik et al. 2011). In the latter, the Mg-rich bulk chemistry apparently plays a role in stabilizing granulite facies assemblages to lower temperature. The presence of B may also serve to stabilize typical granulite facies minerals, such as kornepine, to considerably lower temperature, as demonstrated by kornepine + andalusite assemblages (Girault 1952; Grew et al. 1990). In the case of common metapelites from the Bamble sector, low  $a_{\text{H}_2\text{O}}$  fluids are probably responsible for the formation of such granulite facies islands, characterized by low-Al orthopyroxene + K-feldspar coronas between biotite and quartz (Fig. 11.8) and spinel  $\pm$  corundum + albite/oligoclase symplectites at the expense of biotite and sillimanite (which have subsequently been partially replaced by sapphirine  $\pm$  albite/oligoclase symplectites) (Nijland et al. 1998). Brines are closely associated with the anhydrous assemblages (Fig. 11.5) and regarded the cause behind the formation of these granulite facies islands. Current fluid inclusions studies indicate salinities of c. 25 wt% NaCl, which are too low to stabilize low-Al orthopyroxene + K-feldspar assemblages. However, daughter minerals like halite are not taken into account in this salinity estimate from heating-freezing data implying that this salinity estimate represents the lower limits of synmetamorphic salinity. The low Al-content of the orthopyroxene and presence of low Ca-plagioclase fits with observations by Perchuk and Gerya (1995) that these Al- and Ca-contents decrease with alkali activity.

This raises further questions concerning the nature of fluids involved in other examples of anomalous formation of granulite facies assemblages. Examples of such assemblages include orthopyroxene + andalusite symplectites at expense of cordierite in granulites from the Arunta block, Australia (Ballèvre et al. 1997) and the retrograde formation of sapphirine at the expense of spinel in quartz-rich

granulites from the Sharyzhalgay complex, Siberia (Podlesskii and Kurdyukov 1992).

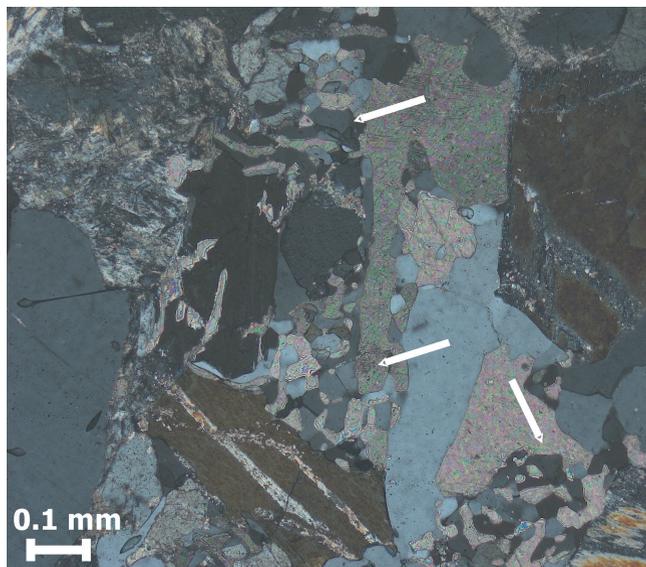
The action of brines in controlling mineral stabilities is not limited to peak metamorphic conditions. Brines may exert their influence during cooling and uplift. The Ubergsmoen augen gneiss is a syntectonic granitic-charnockitic intrusion in the Bamble area, with a fluid inclusion population dominated by H<sub>2</sub>O and CO<sub>2</sub>, respectively (Touret 1985). It generally shows a postmagmatic evolution of orthopyroxene + clinopyroxene → hornblende + garnet → hastingsite + biotite. The later assemblage is in particular abundant in white pinkish granitic zones between fresh, greenish charnockitic varieties, and is accompanied by late trails of aqueous fluid inclusions. The zones might be termed decharnockitization. In one section, however, the late stage evolution is different, with the hastingsite + biotite being overgrown by new, coarser grained coronas of garnet, with contemporaneous replacement of plagioclase by aggregates of scapolite and carbonate.

Associated with this assemblage (and different from other samples), are small fluid inclusions, generally about 5 μm and occurring in trails, often ending near larger, irregular shaped empty inclusions; sporadically, a salt cube, without fluid, is present in the latter inclusions. Inclusions, saturated with NaCl at room temperature have final melting temperatures between -4.7°C and -27.8°C, homogenizing to liquid between 122.3°C and 188.7°C (Nijland and Touret 2000). Fluid inclusion textures, the large spread in freezing point depression, and the relatively low homogenization temperatures indicate that the inclusions likely reflect reset samples of the original fluid that was contained in the larger, now decrepitated inclusions. This fluid was saturated with NaCl and possibly with other salts. The close spatial relationship between brine inclusions and a late anhydrous assemblage demonstrates that, as in prograde granulite facies metamorphism, brines with reduced water activity may provoke *progressive dehydration* during retrograde cooling and uplift in upper greenschist – lower amphibolite facies and give rise to *apparently* high grade mineral assemblages (Nijland and Touret 2000).

The example above illustrates that peak metamorphic granulite facies fluids may stay present during the post-peak metamorphic evolution, instead of being expelled. This holds for both brines and CO<sub>2</sub>. Salient examples include the formation of calcite + quartz symplectites after wollastonite in the granulite facies Rogaland area, south Norway (Fig. 11.9); similar symplectites have been described by Harley and Santosh (1995) from Kerala, south India.

### ***11.5.2 K-Feldspar Microveins and Myrmekites***

K-feldspar microveins at the boundary between quartz and feldspar in granulite facies rocks were already noted by Coolen (1980), then described in India (Hansen et al. 1984) and subsequently in a series of Russian and Finnish granulite facies rocks (Perchuk and Gerya 1992, 1993, 1995). Similar K-feldspar microtextures have subsequently been found in many other mafic granulites (Franz and Harlov 1998;



**Fig. 11.9** Post-peak metamorphic effect of  $\text{CO}_2$ : calcite + quartz symplectites (indicated by *arrows*) after wollastonite in a hedenbergite-scapolite-wollastonite-quartz rock from the Asheimvatn, Rogaland, Norway

Harlov et al. 1998; Safonov 1999; Parfenova and Guseva 2000; Harlov and Wirth 2000; Harlov and Förster 2002; Montanini and Harlov 2006; Hansen and Harlov 2007). K-feldspar in the veins often has high Ba-contents (Hansen et al. 1995; Franz and Harlov 1998) and may contain relatively high albite contents. Different microtextures occur (Harlov et al. 1998; Sukumaran and Ravindra Kumar 2000; Harlov and Wirth 2000; Harlov and Förster 2002; Montanini and Harlov 2006; Hansen and Harlov 2007) (Figs. 11.10–11.13). These include:

- Rods, blebs, and irregular patches of antiperthite in randomly scattered plagioclase grains.
- Microveins along quartz-plagioclase and plagioclase-plagioclase grain boundaries or other minerals grain boundaries (pyroxene, opaque, etc.) or transecting these (Figs. 11.10 and 11.13).
- Myrmekites, which are vermicular intergrowths with Ca-rich plagioclase along (mainly) K-feldspar grain boundaries (Figs. 11.11–11.13).
- Occasional micropertthitic discrete anhedral grains.

It is interesting to note that, despite their ubiquitous character, these intergranular microveins had remained largely unnoticed, until fluid inclusion studies indicated the possible occurrence of intergranular fluids at these deep levels.

Among all these microstructural features, myrmekites are the most common and obvious. They are also quite abundant in granites, especially when deformation has facilitated the introduction of external fluids (Simpson and Wintsch 1989). Myrmekites in granulites deserve special attention for several reasons:

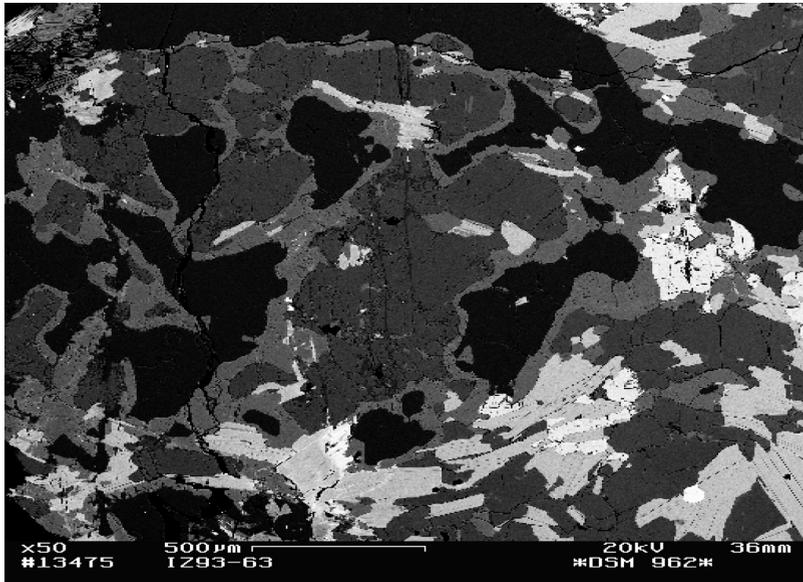


Fig. 11.10 Examples of K-feldspar microveins in granulites from a localized dehydration zone from the Seward peninsula, Alaska (Harlov and Förster 2002)

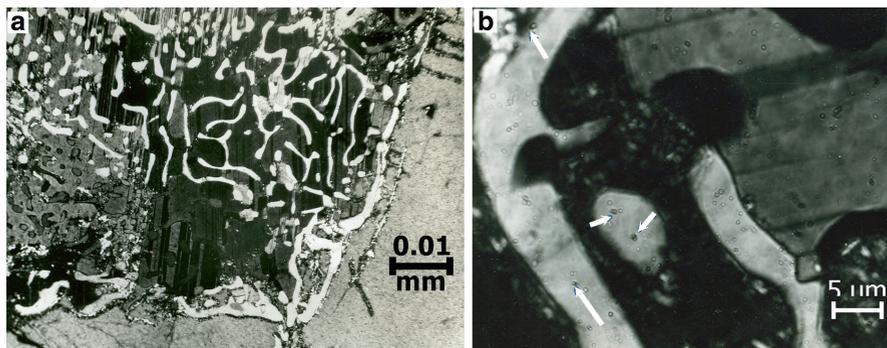
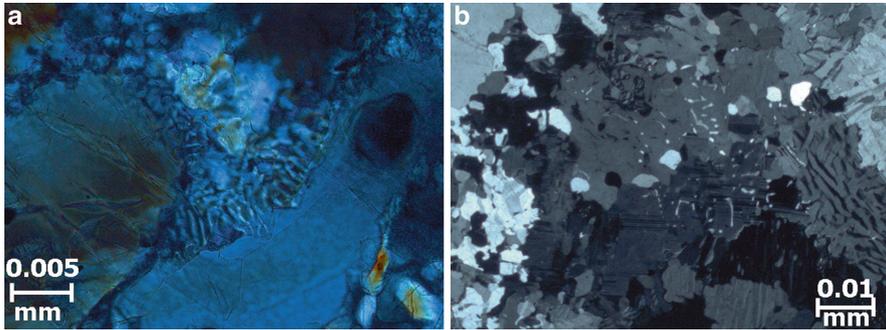


Fig. 11.11 Giant myrmekites in plagioclase at the contact with cordierite in cordierite-anthophyllite rocks from Åkland, Bamble sector, Norway (a). (b): detail, with *arrows* indicating minute brine fluid inclusions

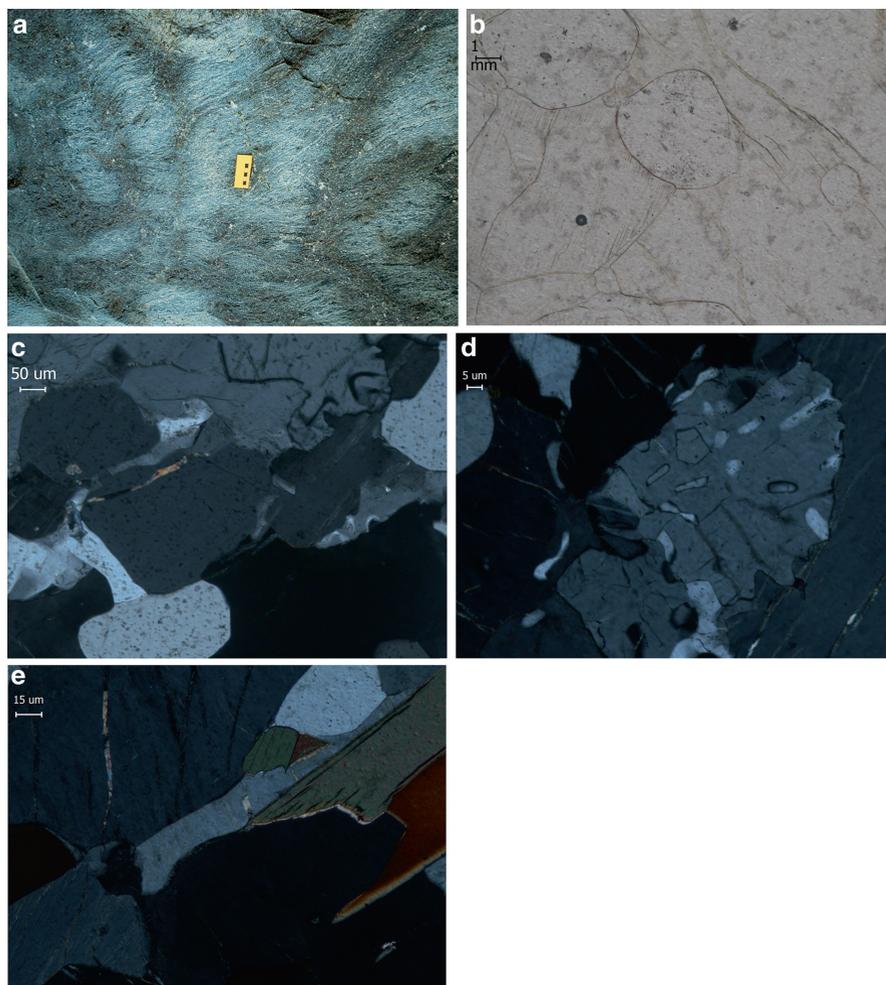
- They are best developed along the margins of large K-feldspar phenocrysts in magmatic granulites (charnockites), especially when these rocks show evidence of high temperature deformation (so-called *charnockitic augen gneisses*), e.g. Hovdefjell and Ubergsmoen in southern Norway (Figs. 11.2 and 11.12a) or Ansignan, Massif de l'Agly, French Pyrénées (Fonteilles 1970) (Fig. 11.14). In these rocks, static mineral recrystallization (*granulite texture*; cf. Vernon 2004)



**Fig. 11.12** Myrmekites in magmatic granulites (charnockites and related rocks). (a): Charnockitic augen gneiss, Hovdefjell summit, Bamble sector, Norway. Along the boundary of a large, plastically deformed K-feldspar phenocryst (augen), a continuous myrmekite layer grades into a granulated groundmass towards the *top* of the figure. Secondary muscovite (and biotite) adjacent to the myrmekite (see Fig 11.14). (b): Myrmekite (*centre*) in an entirely recrystallized, undeformed garnet granulite, commercially distributed as ornamental stone under the name Kashmir White, (India); large, undeformed mesoperthite crystals occur to the *right* and *bottom* and typical groundmass granulite texture to the *right*

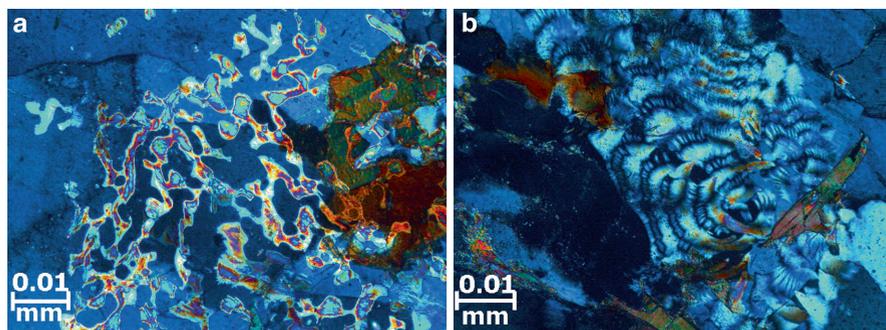
postdates high temperature plastic deformation, occurring first in the ground mass of the rocks and eventually in the augen. In such cases, any sign of plastic deformation are erased (Fig. 11.12b). This type of re-equilibrated texture can only occur in high temperature granulite facies environments, demonstrating that the formation of myrmekites in granulites occurred at high temperature, close or equal to peak metamorphic conditions. The myrmekites should be considered as part of the characteristic granulite facies mineral assemblage instead of late phenomena as often assumed, especially in granitic rocks.

- Myrmekites occur preferentially (but not exclusively) in K-feldspar bearing rocks. For instance, in the Ansignan charnockite, they are much more abundant in the garnet-bearing marginal charnockitic variety than in the orthopyroxene-bearing enderbitic core. However, they may also be found, sometimes in a most spectacular way, in rocks virtually free of K-feldspar. An example is the cordierite-anthophyllite rock (often replaced by phlogopite or actinolite) from the Bamble sector, southern Norway, shown in Fig. 11.11. These well known rock varieties, remarkable by the gem quality and the large size (up to 1 dm) of most of their rock forming minerals, may contain spectacular myrmekites at the contact between cordierite and quartz. These rocks contain great numbers of small brine inclusions, either in the cordierite or, most typically, in quartz blebs within the cordierite (Fig. 11.5c, d). The origin of these cordierite-anthophyllite rocks has been much debated, as shown later (§ 11.5.5). In the case of most possible precursor, brines are a more than a likely premetamorphic fluid, and the brines found in inclusions probably represent far remnants from these hydro-thermal fluids.
- In charnockites, charnockitic augen gneiss and related rock types, myrmekites are closely associated with feldspar microveins on the scale of single intergrain



**Fig. 11.13** Network of incipient charnockite veins crosscutting the foliation of garnet-bearing pelitic gneisses, Kurunegala, Sri Lanka. The incipient charnockite shows K-feldspar microveins and myrmekite (Sample provided by L.L. Perchuk, 96 Per 4, see Perchuk et al. 2000). (a): Field occurrence (Photo courtesy A. Kröner), with *greenish* charnockite and *light grey* pelitic gneisses. **b–e**: Microscopic features (**b** plain polarized light, **c–e** cross polarized light). (b): massive part of an incipient charnockite vein made up by mesorhites and rounded quartz crystals containing minute fluid inclusions (appearing as *black dots*). (c): myrmekites (*right*) adjacent to K-feldspar microveins (*left*) along the boundary of large mesoperthite crystal. (d): detail of myrmekite. (e): detail of K-feldspar microvein, in this case extending along biotite interfaces

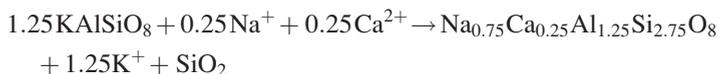
boundaries (Harlov and Wirth 2000). They occur in such a way that they have to be considered as complementary processes. A good example of this can be found in so-called *incipient charnockite* from Kurunegala, Sri Lanka (Perchuk et al. 2000), described in detail in Fig. 11.13. Here, in the middle, massive part of



**Fig. 11.14** Muscovitization of K-feldspar microveins in the Ansignan charnockite, Massif de l'Agly, French Pyrénées. (a): Network of muscovite, first partially replacing K-feldspar microveins (*upper left corner*), subsequently overgrowing the remaining mineral assemblage. (b): Detail of circular arrangement of platy muscovite crystals, possibly reminiscent of former myrmekite textures

an incipient charnockite vein contains mostly mesoperthites and quartz (Fig. 11.13b), with small amounts of orthopyroxene and biotite. Myrmekites and K-feldspar microveins occur along the same boundary of large mesoperthite crystals, at a distance of a few millimetres or less (Fig. 11.13c). Whereas myrmekites only occur at or in the mesoperthites (Fig. 11.13d), the K-feldspar microveins have developed at different mineral interfaces, either feldspar, quartz or even biotite (Fig. 11.13e). As in the case of the cordierite-anthophyllite samples, the role of brines is indicated by the fact that, when a K-feldspar microvein is in contact with or very near a quartz grain (Fig. 11.13c), the quartz contains many small, collapsed brine inclusions. If enough fluid remains available after formation of the microveins, K-feldspar may eventually be replaced by muscovite or, in relatively Fe-rich environments, by biotite. For example in the Ansignan charnockite, muscovite occurs in spectacular networks superimposed on the original mineral assemblage (Fig. 11.14a) and in circular aggregates which suggest pseudomorphism of former myrmekitic textures (Fig. 11.14b).

So much has been written on myrmekites, that any interpretation will immediately lead to some controversy. Our observations fit best with the early interpretation of myrmekites by Becke (1908), the first one ever proposed after these structures had been discovered under the microscope. Here K-feldspar is replaced by intergrowths of plagioclase + quartz under the influence of Na- and Ca-bearing aqueous solutions, according to the reaction (Simpson and Wintsch 2007):



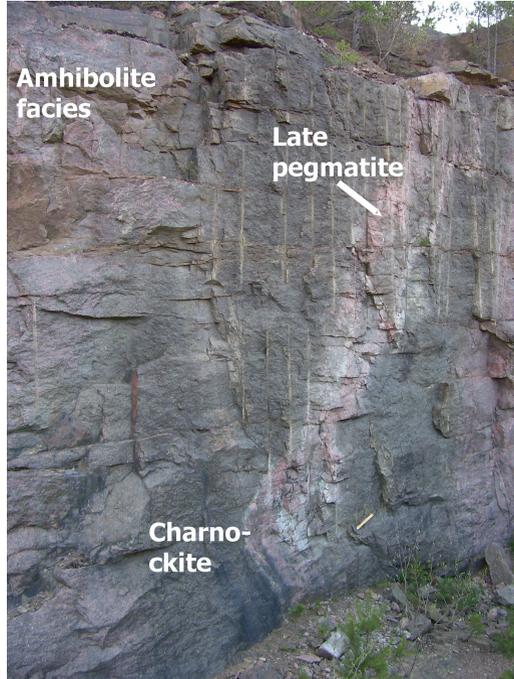
This reaction releases  $\text{K}^+$ , which immediately reprecipitates in the form of alkali feldspar, as it is less mobile in solution than  $\text{Na}^+$  or  $\text{Ca}^{2+}$ . The fact that this represents the formation of a new mineral, not, as in the case of myrmekite,

the replacement of a pre-existing mineral phase, is indicated by the fact that the K-feldspar microveins are developed between quite different minerals, including biotite (Fig. 11.13e). Silica liberated during myrmekite formation is less mobile than the K-feldspar. This is shown by the frequent occurrence of small quartz grains, all with the same optical orientation, around the myrmekite domains (Fig. 11.13c). It gives the impression of silica-rich solutions penetrating the rocks on the scale of a few mineral grains.

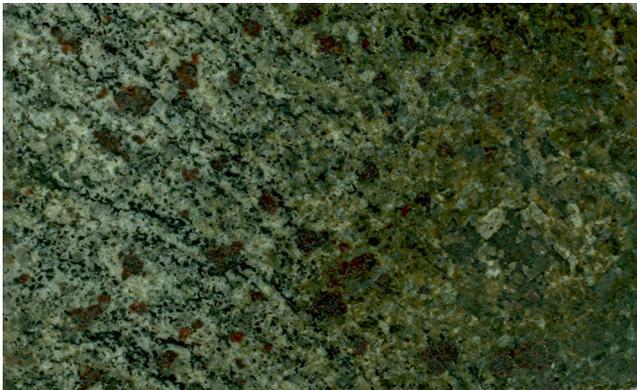
### ***11.5.3 Incipient Charnockites***

Ever since charnockite was first defined by T.H. Holland from the tombstone of Calcutta's founder Job Charnock (Holland 1900), its genesis has been debated between proponents of either a metamorphic or an igneous origin (e.g. Quensel 1952; Pichamuthu 1953; Klimov et al. 1964; Harley 1989; Newton 1992a,b; Kilpatrick and Ellis 1992; Abramov and Kurdyukov 1997; Frost and Frost 2008). As early as 1960, C.S. Pichamuthu (1960, 1961) reported the local transformation of biotite and amphibole gneisses into charnockite in Mysore state, southern India. His observation raised no marked interest until the 1980s, when it was realized that some of these incipient charnockites, in most cases irregular patches of decimetre to metre size, did not only occur in a variety of protoliths, ranging from metavolcanics to metapelites and gneisses, but also often occurred along a regular fracture network (Fig. 11.13a), suggesting that they represented fossil fluid pathways (Friend 1981). This distinguishes them from igneous charnockites occurring in anorthosite-mangerite-charnockite-granite suites. Incipient charnockites have since been found in many areas including southern India and Sri Lanka (e.g. Janardhan et al. 1979, 1982; Park and Dash 1984; Hansen et al. 1984, 1987; Raith and Srikantappa 1993), the Seward Peninsula, Alaska (Harlov and Förster 2002) and SW Sweden (Harlov et al. 2006; Fig. 11.15). Fluid inclusion data document the general occurrence of CO<sub>2</sub>-rich fluid inclusions in the charnockite only, not in the surrounding amphibolite facies rocks (e.g. Sri Lanka, Perchuk et al. 2000, SW Sweden, Harlov et al. 2006). But the other granulite fluid (brines) can also be present, especially in granitic intrusions. This is notably the case for the well-known Closepet granite in the Indian Dharwar craton (Srikantappa and Malathi 2008; see also Slaby et al. 2011). Brine-loaded incipient charnockites (Srikantappa and Arash Zargar 2009) occur in the lower part of the granite. These brines were presumably expelled from underlying massive igneous charnockites.

Data from all incipient charnockite localities, whether with a metasedimentary (metapelitic) or magmatic precursor, clearly demonstrate the progressive nature of the metasomatic transformation from precursor to charnockite (Fig. 11.16). In detail, this can be quite complicated. Knudsen and Lidwin (1996) interpreted enderbite veins on Tromøy and Havsøya in the Bamble sector, southern Norway, as intrusive, and described the formation of dehydration bands along these veins as being due to the infiltration of CO<sub>2</sub> ± N<sub>2</sub> from the veins (Fig. 11.17). Vander



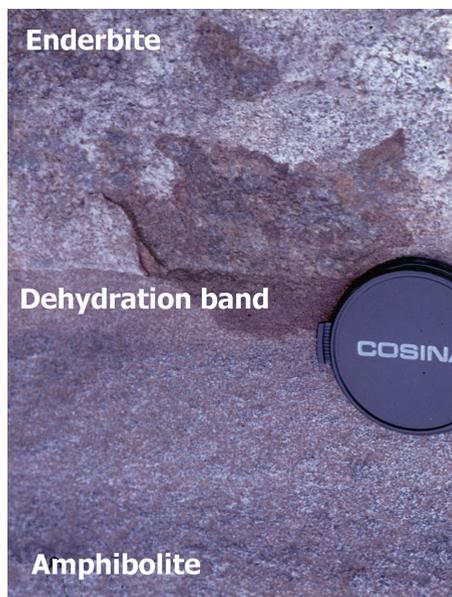
**Fig. 11.15** Incipient charnockites in the Söndrum quarry (Stenhyggeriet), Halmstad, SW Sweden (cf. Harlov et al. 2006)



**Fig. 11.16** At the scale of the hand specimen, progressive transformation of garnet-biotite gneisses (*left*) into coarse-grained, homogeneous charnockite (*right*) (*Arrested charnockite formation*; Raith and Srikantappa 1993) (Sample 04JT3, courtesy M. Raith; width of picture 8 cm)

Auwers (1993) described the formation of dehydration bands at the margins of amphibolites against charnockites, with element mobility and continuous changes in solid solution compositions over the bands. In this case, element mobility is

**Fig. 11.17** Dehydration band along the margin of amphibolite against enderbite on Tromøy, Bamble sector, Norway



considered to be a diffusional process rather than an infiltration process. The diffusional process was considered to be controlled by the chemical potential of  $\text{SiO}_2$  rather than that of  $\text{H}_2\text{O}$ .

Following Korzhinskii's (Korzhinskii 1962) notion of the importance of the chemical potential of  $\text{K}_2\text{O}$ , Perchuk and Gerya (1992) developed the concept of charnockitization of amphibolite facies rocks being dependent on the infiltration of one or both metasomatizing agents,  $\text{CO}_2$  or  $\text{K}_2\text{O}$ , a notion also proposed by Stähle et al. (1987). In the case of  $\text{CO}_2$ , this involved generalized reactions like tschermakite + quartz +  $\text{CO}_2$ -fluid  $\rightarrow$  anorthite + enstatite +  $\text{CO}_2$ - $\text{H}_2\text{O}$ -fluid, i.e. in which the  $\text{CO}_2$  component is not consumed. In case of  $\text{K}_2\text{O}$ , reactions like pargasite + quartz +  $\text{K}_2\text{O}$  (in fluid)  $\rightarrow$  K-feldspar + enstatite + diopside +  $\text{H}_2\text{O}$ -fluid containing  $\text{Na}_2\text{O}$  are involved. These reactions illustrate several important aspects regarding incipient charnockitization:

- In all cases, dehydration is accompanied by the formation of new feldspars.
- In the case of  $\text{CO}_2$ , the active agent promoting dehydration is not consumed in the reaction, but remains available, thus capable of promoting charnockitization/enderbitization elsewhere after transport.
- In many cases, actual metasomatism occurs.  $\text{K}_2\text{O}$  is introduced and consumed in the reactions, resulting in evident changes in the bulk rock compositions.

The later is not limited to  $\text{K}_2\text{O}$ . Bulk chemical analysis of incipient charnockites show that, compared to the original amphibole-bearing gneiss, the former show an increase in not only  $\text{K}_2\text{O}$  but also  $\text{Na}_2\text{O}$  and  $\text{SiO}_2$ , and a decrease in  $\text{MgO}$ ,  $\text{FeO}$  and especially  $\text{CaO}$  (Hansen et al. 1987; Stähle et al. 1987). The loss of  $\text{CaO}$ , probably

due to the incongruent dissolution of hornblende (Newton pers. com. 2011) and may be explained by its high solubility in concentrated NaCl solutions (Newton and Manning 2007).

For those interested in the history of geology and its concepts, it is tempting to speculate how the recent results regarding to the formation of feldspars discussed above would have figured in the great debate in the 1940s and 1950s between the so-called *soaks* and *pontiffs* (Young 2002). The soaks, a nickname coined by N.L. Bowen, favouring formation of granites by diffusion in the solid state (Bugge 1945), denominated as granitization (Read 1957), the pontiffs headed by E. Niggli a magmatic origin. A major difference with the current hypotheses is, however, that whereas true solid state diffusion must be extremely limited to a scale not exceeding few millimetres at most, element transport resulting in charnockite formation in the amphibolite to granulite transition zone must occur on a much longer scale on the order of metres to kilometres. This type of K-feldspar formation may not be limited to incipient charnockites. Baltybaev et al. (2010) presented evidence for the regional formation of large K-feldspar crystals in migmatitic quartz-feldspar leucosomes after solidification and deformation of these leucosomes.

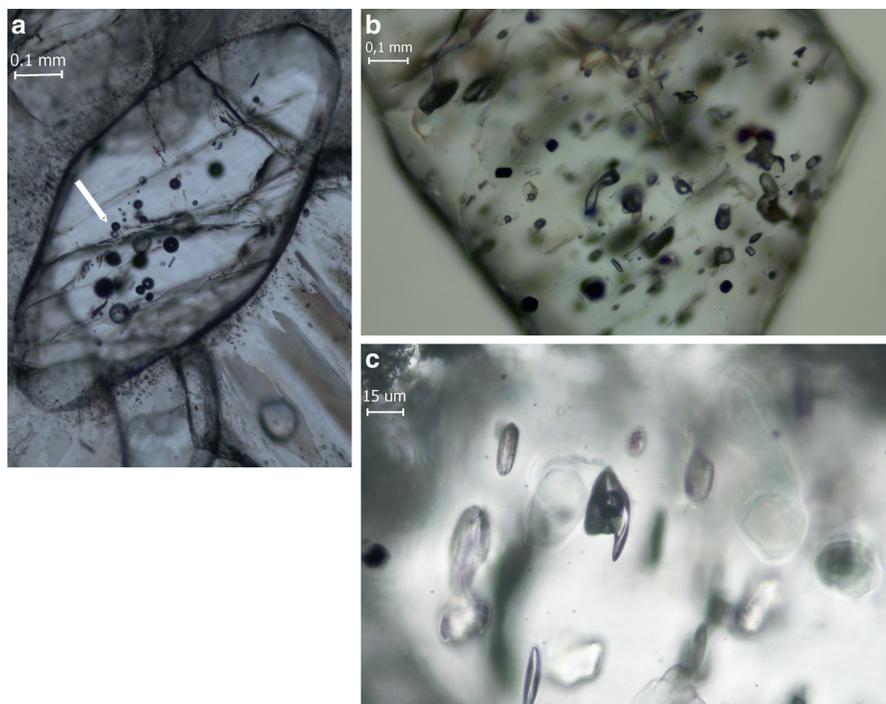
The metasomatic origin of incipient charnockites does by no means imply that they have systematically escaped melting. In a very detailed study of the Kurunegala (Sri Lanka) occurrence shown in Fig. 11.13, Perchuk et al. (2000) concluded that gneisses had been metasomatically altered along shear zones with minimum melting for the post metasomatic composition (700–750°C, 6 kb,  $a_{\text{H}_2\text{O}}$  0.52–0.59) due to the influx of an external fluid with both CO<sub>2</sub> and a supercritical brine component. These observations would imply that in this case supposedly magmatic charnockites can then be produced in situ, without showing any sign of restitic material along the edge of the molten zone.

#### ***11.5.4 Metasomatic Redistribution of Elements Traditionally Considered as Immobile***

Until a decade ago, widespread opinion considered REE, Th and U-bearing accessory minerals in high grade rocks, notably phosphates and zircon, to be non-reactive and recording magmatic phenomena only. For this reason, they are widely used for dating, each successive layer, if occurring, being ascertained to successive episodes of magmatic growth. This is in marked contrast to observations that minerals like monazite and xenotime may be formed at sub-greenschist facies conditions (Sestri-Voltaggio zone, Liguria, Italy, Cabella et al. 2001; Bukowiec, Carpathians, Poland, Budzyn et al. 2005; Malopolska block, Poland, Stanislawska and Michalik 2008) or even in diagenetic environments (Welsh basin; Evans and Zalasiewicz 1996). Meanwhile, it has been known for a long time that many granulite zircons show rounded, almost eroded shapes. These are often explained by multiple resorption and interaction with extracted melts (Tichomirova et al. 2005). However, several

studies have shown that in many cases Zr-bearing minerals can be quite mobile during granulite facies metamorphism, through dissolution-reprecipitation processes (Harlov and Dunkley 2010, Schutesky della Giustina et al. 2011). Some observations show that distances of mobility can reach at least the outcrop scale, such as the case of ilmenite-zircon veinlets occurring in granulite facies metamorphosed pegmatite in Rogaland, south Norway (Wielens 1979).

Also in Rogaland, Bingen (1989) demonstrated that granulite facies metamorphism may result in the redistribution of supposedly immobile elements, manifest by a regional allanite-out isograd; over the amphibolite to granulite facies transition zone, REE, U and Th are redistributed between accessory phases (Bingen et al. 1996). Similar observations with regard to the redistribution of REE in minerals as a function of metamorphic grade have been made along a traverse of amphibolite to granulite facies Archean lower crust in the Dharwar craton, southern India (Hansen and Harlov 2007). During the last decade, it has become clear that dissolution-reprecipitation processes, assisted by brines, might affect accessory phases under P-T conditions relevant to granulite facies metamorphism (Harlov et al. 2002, 2005; Harlov and Förster 2003). However, these do not explain typical zonation textures in for example monazite and zircon with respect to Th, U and Pb. Recent experiments at Potsdam (Harlov and Hetherington 2010; Harlov et al. 2011) have shown that at the relevant P-T conditions (900°C, 1,000 MPa and 600°C, 500 MPa, respectively), alkali-bearing fluids are able to explain these textures, resulting in pseudomorphic (partial) replacement and redistribution of elements by coupled dissolution-reprecipitation processes, the grain boundary fluid essentially acting as catalyst. The metasomatized monazite is apparently completely recrystallized, with a pristine appearance. This mechanism provides an alternative explanation to melt extraction. Experimental fluids consisted of NaOH, KOH, a water glass-like compound  $\text{Na}_2\text{Si}_2\text{O}_5$  or NaF, with addition of a few more 'traditional' fluid components (HCl,  $\text{CaCl}_2$  and  $\text{Ca}(\text{OH})_2$ ) in case of zircon (Harlov and Dunkley 2010). The exact nature of these fluids is still an open question. As a matter of fact, they represent a key problem with regard to fully understanding processes occurring in the lower crust. Their presence would, however, have far reaching consequences. Not only does it explain the remarkable abundance of monazite in some granulite terrains, – monazite-bearing sands are a major source of thorium along the southern coast of Madagascar-, but it again implies that dating the external layer of a monazite, xenotime or zircon crystal does not necessarily relate to the last magmatic event, but might well reflect metasomatic action. Another interesting consequence of such fluids would be their high capability to transport silica. Metasomatic granulite veins, like in the Seguin subdomain of the Grenville province, for which granulite facies infiltration of  $\text{SiO}_2$ ,  $\text{Na}_2\text{O}$  and  $\text{Al}_2\text{O}_3$  has been documented (Zaleski and Pattison 1993) might be an interesting case for future research. These fluids have not yet been found in fluid inclusions, though, as indicated at the beginning of this paper, it can not be excluded that they correspond to some 'low salinity' aqueous inclusions occasionally found in granulites, but generally considered not to be related to granulite facies metamorphism because of their incompatibility with the granulite mineral assemblage.



**Fig. 11.18** Fluid inclusions in carbonatites: (a): CO<sub>2</sub>-inclusions in apatite from Kovdor, Kola peninsula, Russia. (b): Overall view of an apatite crystal from the Kerimasi carbonatite, completely crowded with primary inclusions. These correspond to three immiscible fluid/melts: carbonatite melt, Mg- and Fe-rich peralkaline silicate melt, and (*arrow*) C-O-H-S alkaline fluid (Guzmics et al. 2011). (c): detail of the biphase (liquid-vapour) alkaline fluid (C-O-H-S- Na and K components, T. Guzmics pers. com. 2010)

Though synmetamorphic alkaline intrusions in granulite facies terrains have not been documented, it is interesting to note that comparable fluids (alkali-bearing and alkaline aqueous solutions) occur in some carbonatites, notably at the transition between calcium- and natrocarbonatites (Fig. 11.18). The inclusions shown in Fig. 11.18, in apatite from the Kerimasi (Tanzania) calciocarbonatite contain a C-O-H-S alkali-bearing fluid (Guzmics et al. 2011), only partly analyzed at present. However, at first sight, estimated compositions are in broad agreement with fluids used by D. Harlov and co-workers in their experiments. As observed by Guzmics et al. (2011), these alkali-bearing aqueous fluids occur in the Kerimasi carbonatite at an early magmatic stage as a result of immiscibility from a Mg- and Fe-peralkaline silicate melt, itself the result of unmixing from a Ca-rich, P-, S- and alkali-bearing carbonatite melt (Fig. 11.18). These types of multiple immiscibilities, only detectable by a thorough investigation of melt and fluid inclusions, are probably much more widespread than commonly realized. They are likely to be a major source of melt and fluid production in the mantle. It is interesting in this respect to note that

highly siliceous alkali-bearing fluids have been produced experimentally in synthetic fluid inclusions (Wilkinson et al. 1996).

Southern Madagascar, a well known, classical granulite province, is a good example to illustrate U, Th and REE-mobility under granulite facies conditions. It has been metamorphosed about 580–600 Ma ago, at the same time as the larger part of east Africa, Sri Lanka, and the most southern part of India (Pan-African orogeny, e.g. Paquette et al. 1994). The fact that all these regions are among the world's largest producers of gemstones is certainly not a coincidence (e.g. Dahanayake and Ranasinghe 1981). It indicates that typical granulite-facies P-T conditions (high temperature, relatively low pressure, low water activity fluids) are ideal for creating gem varieties of ordinary minerals like zoisite (tanzanite) and garnet (tsavorite). In the region of Tranomaro, in the southeasternmost part of Madagascar, a number of carbonate-rich metasedimentary layers occur, partly transformed into pyroxene-rich rocks. These pyroxenites also contain garnet; therefore, they are commonly described as skarns (Moine et al. 1985). However, like in the Arendal region of the Bamble sector, southern Norway, they are not a product of contact metamorphism due to a granitic intrusion, but the result of regional granulite facies metamorphism, at a temperature of about 800–850°C and a pressure of 4–5 kb (Moine et al. 1985). Some marbles and, to a lesser extent, skarns are enriched in U, Th, Zr and REE. Locally, they form deposits of economical value, being the source of urano-thorianite minerals abundant in placers along the southeastern coast of Madagascar (Rakotondrazafy et al. 1996). Boulvais et al. (2000) showed how, across a meter-scale profile at the marble-pyroxenite contact, REE-contents increase markedly towards the skarns, e.g. La from 19.8 ppm in the most distant sample to 129 ppm in marble at the pyroxenite contact. These authors concluded that this element distribution was caused by the infiltration of a fluid at peak metamorphic conditions. Fluid inclusion studies show this fluid to be CO<sub>2</sub>-rich ( $X_{\text{CO}_2} > 0.8$ ; Ramambazafy et al. 1998). It is interesting to note that fluid infiltration was not recognizable from C- and O isotopic signatures, because of the persistence of large isotopic variations predating granulite facies metamorphism.

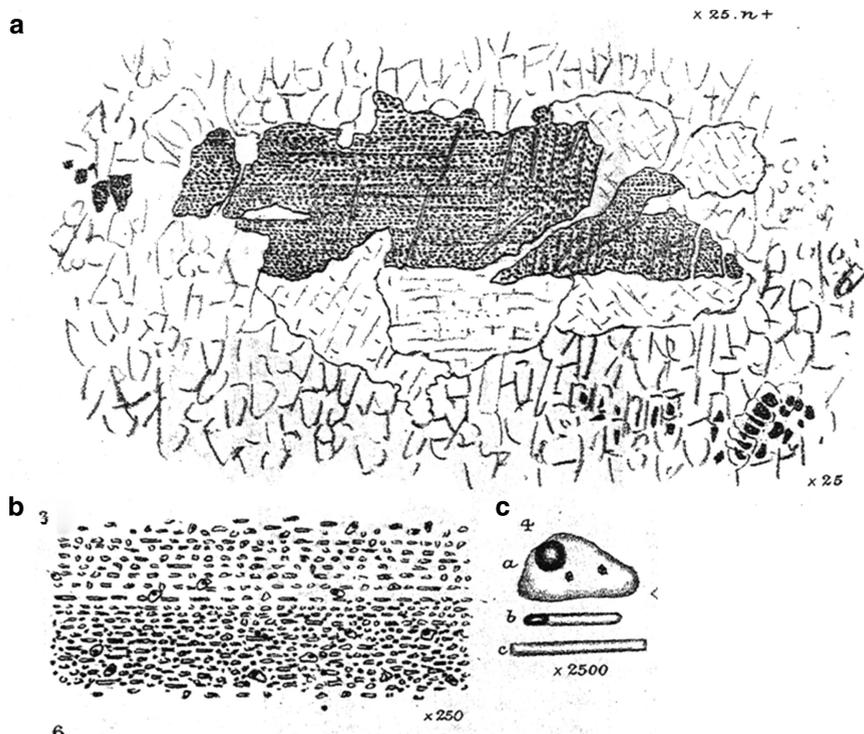
### ***11.5.5 Peak Metamorphic to Retrograde Bulk Chemical Processes (Scapolitization, Albitization)***

Lower crustal granulites now exposed at the Earth's surface must have experienced a complicated uplift history, during which a number of retrograde features may have happened. If still hot granulites are thrust on low-grade, H<sub>2</sub>O-rich metamorphic terranes, it may lead to widespread retrogradation and various types of retrograde isograds (e.g. orthopyroxene to anthophyllite), as described by D. Van Reenen and co-workers in the Limpopo complex of southern Africa (Van Reenen 1986; Van Schalkwyk and Van Reenen 1992; Van Reenen et al. 2011). In general, however, both peak and retrograde metamorphic evolution of

amphibolite and granulite facies terranes involve regional scale metasomatic processes, provoked by the infiltration of notably Na ( $\pm$  Cl)-rich fluids (albitization and scapolitization), K-rich fluids or possibly Mg-rich fluids, giving rise to the formation of particular lithologies. Mg-rich cordierite-orthoamphibole rocks have already been mentioned. These high temperature analogues of the high pressure orthoamphibole-kyanite-talc schists (*whiteschists*, Schreyer 1974) occur in various settings within regional amphibolite-granulite facies terrains and high temperature contact metamorphic aureoles. The question whether synmetamorphic metasomatism is involved is still debated. An origin due to Mg metasomatism has been proposed for these rocks by several workers (Eskola 1914; Tilley 1937; Oen 1968). Alternative origins proposed include low temperature alteration of mafic volcanics (Vallance 1967; Reinhardt 1987; Smith et al. 1992; Visser 1995; Hinchey and Carr 2007), sediments derived from altered or weathered volcanics (Robinson and Jaffe 1969), weathered greywacke (Owen and Greenough 1995), meta-evaporites (Touret 1979; Nijland et al. 1993), products of metamorphism at very high oxygen fugacities (Cameron and Hattori 1994) or retrogressed granulites (Van Reenen 1986). Probably, different genetic types exist.

Scapolitization and albitization, on both a local as well as a regional scale, have been observed in several amphibolite and granulite facies terrains (for a more detailed discussion, see Chaps. 4 and 7). Both processes clearly involve chemical mass transfer by fluids, notably brines. Both processes may be contemporaneous. This is, for example, the case in the amphibolite facies Mary Kathleen fold belt, Mt. Isa inlier, Queensland, Australia where albitization and scapolitization of calc-silicate rocks occur at peak metamorphic conditions (560–630°C, 3–4 kb; Oliver and Wall 1987; Oliver et al. 1990, 1994; Oliver 1995). In other areas, they show close spatial relationships, but follow each other in time. This is the case in the amphibolite to granulite facies transition zone of the Bamble sector, southern Norway (Putnis and Austrheim 2010; Engvik et al. 2011). Albitization and scapolitization occur in metasedimentary complexes dominated by quartzites intruded by gabbroic magmas (the Nidelva Quartzite Complex in the Froland area and Coastal Quartzite Complex in the Kragerø area). However, whereas scapolitization likely occurred at or near peak metamorphic conditions (Visser et al. 1999, Engvik and Austrheim 2010), spatially associated albitization has occurred during cooling and uplift.

In the Bamble sector, scapolitization in the form of Cl-rich scapolite mainly affects the gabbros and related amphibolites in the amphibolite facies portion of the amphibolite to granulite facies transition zone (Brøgger and Reusch 1875; Brøgger 1934; Green 1956). The role of fluids during the transformation of plagioclase to scapolite has already been shown in 1899 by W.J. Judd in a paper inspired by H.C. Sorby, which contains one of the first microscopic descriptions of fluid inclusions in rocks (Fig. 11.19). Judd was not able to identify precisely the fluid composition, but the strong contrast between liquid and vapour bubble in his drawings (Fig. 11.19c), as well as the occurrence of minute cubic solids in the inclusions clearly indicate that he had found a highly saline aqueous solution. Another element frequently found in or associated to brines is boron; its presence is suggested by the frequent

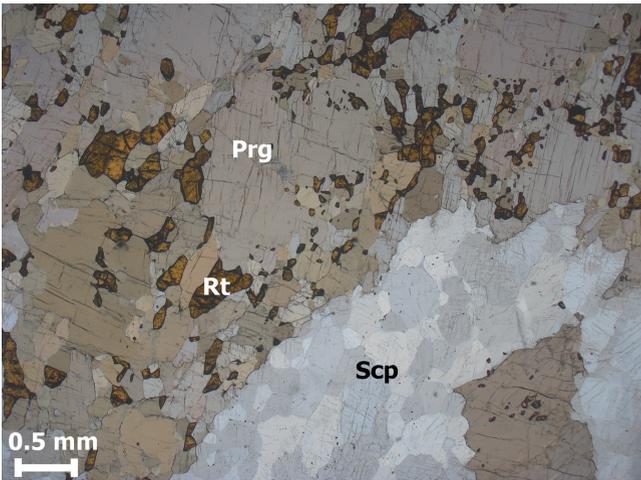


**Fig. 11.19** Judd's (1889) '*.. plagioclase feldspar converted into scapolite*'; (a): cloudy plagioclase (grey) partly surrounded by scapolite (clear). (b): plagioclase containing myriads of fluid inclusions. (c): detail of the inclusions

occurrence of tourmaline in scapolite veins (Fig. 11.20). These veins contain the rare tri-octahedral Al-rich sodium mica preiswerkite (Visser et al. 1999), a mineral usually found in HP metamorphic rocks (e.g. Godard and Smith 1999; Wang et al. 2000), but in this case indicative of high  $\text{Na}^+$  activity in the fluid. Scapolitization in southern Norway is generally accompanied by the formation of local apatite-apatite-phenogopite veins (Liefink et al. 1994; Engvik et al. 2009). This illustrates the economic importance of metasomatism. At one time, the Ødegårdens Verk deposit in the Bamble municipality constituted the largest phosphate mine in Europe, exploited by the Compagnie Française de Mines de Bamle and the Norwegian Bamle Apatit AS (Bugge 1965, 1978). Associated with scapolitization are also significant enrichments of rutile (Korneliussen et al. 1992; Fig. 11.21). Other local metasomatic effects accompanying scapolitization include the formation of sapphirine in scapolitized gabbro (Engvik and Austrheim 2010; Engvik et al. 2011). The widespread formation of Cl-rich scapolite (up to  $X_{\text{Cl}}$  up to 0.94; Liefink et al. 1993), which itself contains brine inclusions (Fig. 11.5f), demonstrate the movement of saline fluids. Chlorapatite and Cl-bearing amphibole formed by (exchange



**Fig. 11.20** Example of preiswerkite and tourmaline-bearing scapolite veins at Blengsvatn, Bamble sector, Norway (cf. Visser et al. 1999)



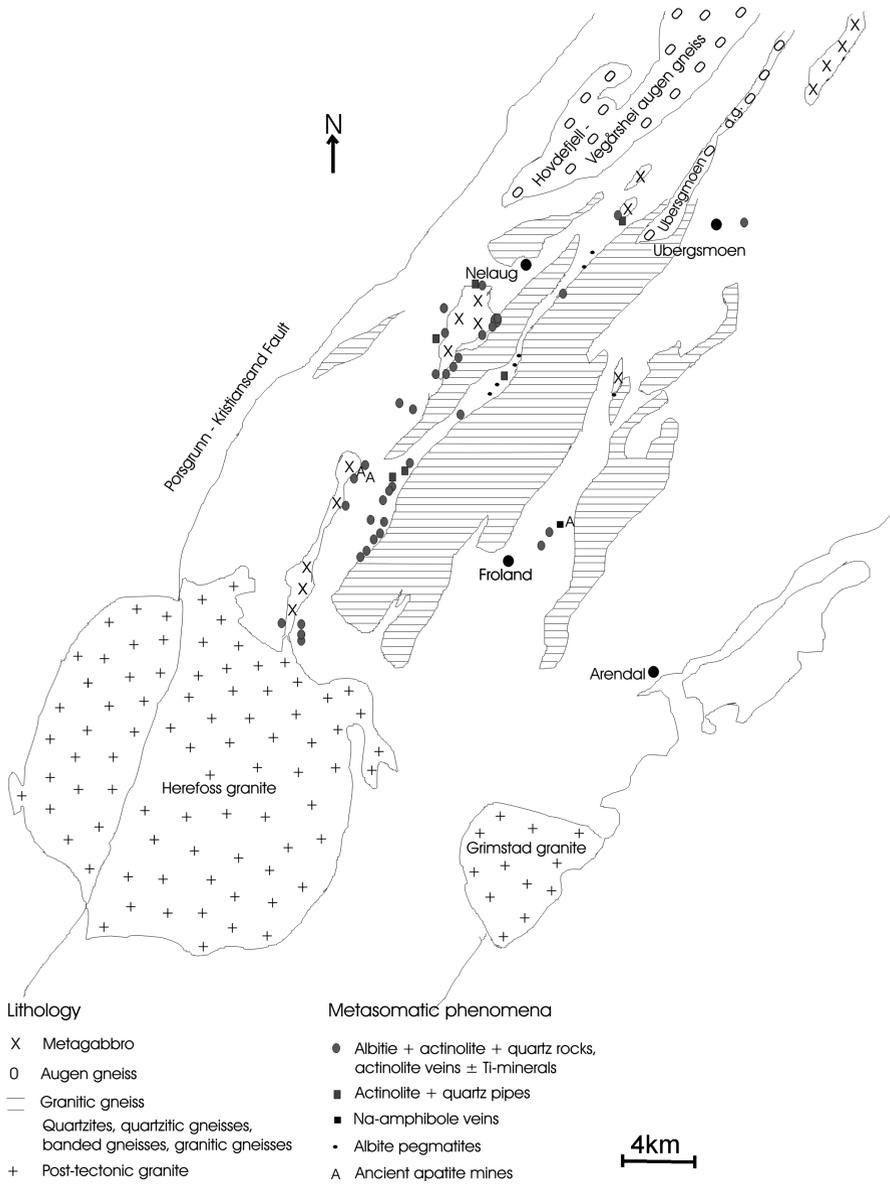
**Fig. 11.21** Rutile-scapolite-pargasite rock from Ødegårdens Verk, Bamble sector, South Norway, illustrating the enrichment in rutile associated with scapolitization

with) these fluids have  $\delta^{37}\text{Cl}$  isotopic signatures comparable to modern sea water (Eggenkamp and Schuiling 1995).

Albitization is well known as a diagenetic phenomenon in sedimentary basins (e.g. Boles 1982), as a subsolidus alteration of granites (e.g. Baker 1985; Boulvais et al. 2007), or as alteration of basalts (e.g. Rehtijärvi and Saastamoinen 1985). Regional scale albitization during cooling and uplift is prominent in several terrains. Besides from the Proterozoic Mary Kathleen fold belt, Australia, it is known for example from the Proterozoic Kongsberg (Jøsang 1966; Munz et al. 1994, 1995) and Bamble sectors of southern Norway (Brøgger 1934; Bugge 1965; Elliott 1966; Nijland and Touret 2001; Engvik et al. 2008). In the latter, albitization occurs on a regional scale (Fig. 11.22). Though called albitization, in many cases spectacular albite + actinolite (hedenbergite)  $\pm$  quartz assemblages are formed, for example in the Bamble sector. In many cases, albitization spreads from central veins that are filled with actinolite ( $\pm$  hedenbergite) (Fig. 11.23). Albitization is accompanied by enrichment in Ti, in the form of abundant titanite and rutile (Watson 1912; Nijland and Touret 2001). The presence of an albite-component in fluids enhances the solubility of rutile (Hayden and Manning 2011). Initial stages often preserve original sedimentary or igneous textures on the meso-scale, as illustrated for example by a graphic pegmatite from Mjåvatn, Norway (Nijland and Touret 2001). Here, plagioclase is replaced by albite, and quartz by actinolite/clinopyroxene intergrowths, whilst the original graphic texture is apparently preserved (Fig. 11.24). On the microscale, however, the original grain boundaries are not preserved. In this case, albitization is evidently accompanied by the leaching of silica. The comparable process of episyenitization, a term originally coined by Lacroix (1920) to describe dequartzification, i.e. the subsolidus leaching of quartz from granitic rocks, has been documented in several cases under greenschist to lower amphibolite facies conditions. In most cases, dequartzification is accompanied by albitization (e.g. Cathelineau 1985, 1986; Petersson and Eliasson 1997; Boulvais et al. 2007). Interestingly, in the Bamble sector, albitite occurrences are often accompanied by the formation of hydrothermal quartz pipes up to several metres in diameter, consisting of very clear quartz and up to several decimetres long (by a 0.5–1 mm diameter) actinolite crystals.

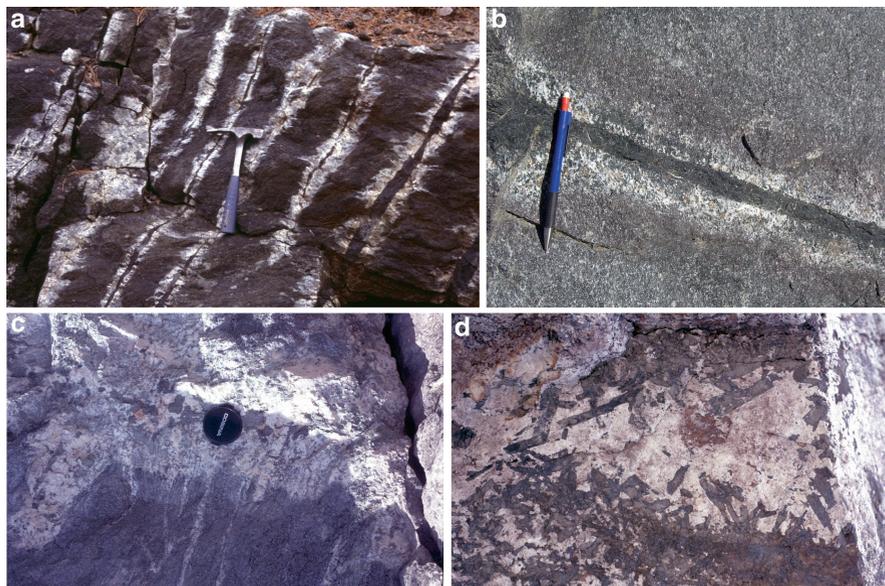
Restoration of volatiles in granulites requires the formation of a new metamorphic plumbing network in response to deformation, including processes such as strain partitioning channeling fluid flow and fluid overpressure initiating fracturing (Etheridge et al. 1983; Yardley 1997; Oliver and Bons 2001). Munz et al. (1995) studied fluid inclusions in albitized rocks from the Modum amphibolite facies terrain, south Norway, which show methane-dominated hydrocarbons and low K, Na-Ca-Cl brines with  $\text{Ca}/\text{Na} < 1$  and  $\text{Br}/\text{Cl}$  ratios comparable to modern seawater. These authors interpreted these as evidence for deep infiltration of fluids from sedimentary basins.

In the case of Mjåvatn, mentioned above, fluid inclusions in the albitite-actinolite-clinopyroxene assemblage show brines with c. 25 wt% NaCl (Nijland and Touret 2001). Ordinary amphibolitized metagabbro (i.e. not affected by the formation of albite-actinolite assemblages) from the area show, besides high density  $\text{CO}_2$ -



**Fig. 11.22** Regional distribution of metasomatic and magmatic albitites in the Froland area of the Bamble sector, South Norway

N<sub>2</sub> and low density N<sub>2</sub> fluid inclusions, NaCl brine inclusions as well (Dam 1995). This might suggest that infiltration of brines starts prior to the formation of albitite-actinolite assemblages. Albitite-actinolite assemblages are generally considered to be formed at 350–450°C, but albitization may actually start at higher temperatures.



**Fig. 11.23** Examples of albitization in the Bamble sector, Norway: (a, b): spreading from central veins filled by actinolite (a): Barlindåsen and (b): Kleivane). (c, d): progressive albitization of gneisses, finally resulting in coarse grained albite-clinopyroxene rock (Mjåvatn); both pictures are from the same, continuous outcrop



**Fig. 11.24** Replacement of graphic pegmatite by albite + actinolite/clinopyroxene from Mjåvatn, Bamble sector, Norway (cf. Nijland and Touret 2001)

TEM-studies show that actinolites from metasomatic albitite rocks in the Bamble sector exhibit cummingtonite exsolution lamellae, indicating formation above the cummingtonite solvus, ca. 620°C at  $P_{\text{Fluid}} = 2 \text{ kb}$  (Ter Haar 1988). This relatively

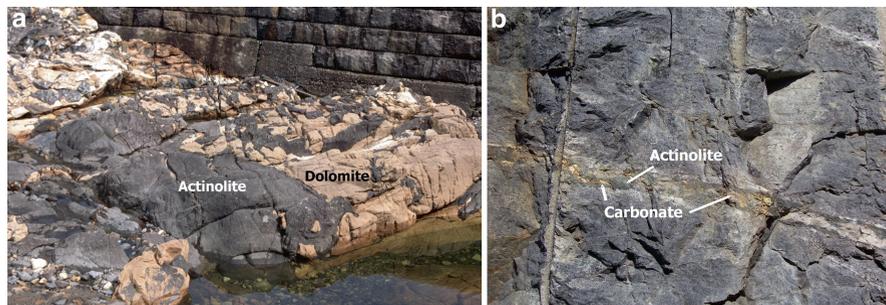
high temperature initiation for the albitization process may be supported by the fact that metasomatic albitites are accompanied by magmatic-hydrothermal pegmatitic dykes or veins, like clinopyroxene-bearing albite pegmatites in the Bamble sector (Bodart 1968; Morshuis 1991) or magmatic-hydrothermal albite-actinolite-apatite rocks in the Mary Kathleen fold belt, Australia (Mark and Foster 2000). In the Froland area of the Bamble sector, where albitization is best developed, it affects both metasediments and metagabbros, in particular along their contacts. In addition, it occurs along late joints. It may be speculated that synmetamorphic saline fluids were collected at the lithological contacts during cooling and uplift.

From an economic geological point of view, it is worth noting that albitization (and dequartzification) is, in several cases, accompanied by the U mineralizations (e.g. Hoeve 1974; Hålenius and Smellie 1983; Lobato et al. 1983; Poty et al. 1986; Porto da Silveira et al. 1991; Respaut et al. 1991), Cu and Au (Frietsch et al. 1997), Fe (Nuutilainen 1968), Ti (e.g. Watson 1912; Force 1991; Korneliusen et al. 1992) or apatite (Bugge 1922; Nijland and Maijer 1991).

### ***11.5.6 Long Distance Action of Granulite Fluids***

If granulite fluids leave behind a wealth of traces on the single mineral or hand specimen scale, their long distance action, namely the possibility to transport elements without noticeable modification of the rock structure, is much more difficult to assess. Besides localized evidence of granulite facies fluids, as discussed above, a number of arguments suggest that these fluids could scavenge some elements at peak metamorphic conditions, subsequently transporting and redistributing them during retrograde stages. Such a hypothesis is notably supported by the fact that many medium to high grade metamorphic areas, like India, Madagascar and Norway, contain shear zones of the regional scale, typically over 100 km by 10 km (Newton 1990; Pili et al. 1997, 1999), in which up to 20% of the country rocks are replaced by massive calcite and/or dolomite (e.g. Dahlgren et al. 1993; Newton and Manning 2002; Fig. 11.25). Carbonate formation occurred at 500–700°C and the uniform carbon isotopic signature ( $\delta^{13}\text{C}$  about  $-7\%$ ) clearly points to a primary mantle origin. A great number of gold deposits are associated with this type of environment (Archean epigenetic gold deposits, e.g. Eisenlohr et al. 1989). With this respect, it is interesting to note that many granulite terranes, on a regional scale, appear to be strongly depleted in gold (e.g. Bamble, Norway and the Lewisian, Scotland; Cameron 1989a,b, 1994). One may speculate that brines have scavenged some elements during granulite facies metamorphism, notably Au or U (Cameron 1988; Chi et al. 2006; Cuney and Kyzer 2009) and transported them, such that they finally end up being deposited in late shear zones.

A similar mechanism had been proposed to scavenge all LILE from the lower crust. This is based on the assumption that the lower crust was depleted in heat producing elements, notably K or Rb, a notion first put forward by Ramberg (1951, 1952) and Heier (1965, 1973). As a matter of fact, this supposed depletion had been



**Fig. 11.25** (a): Dolomite-actinolite rocks at Kammerfoss, Bamble sector, Norway (cf. Dahlgren et al. 1993; note concrete blocks for scale). (b): carbonate-actinolite vein at Kleivane, Bamble sector, Norway (width of vein about 6–8 cm)

a key argument for the vapour-absent melting model of granulite metamorphism, i.e. LILE being supposedly removed by anatectic melts (Fyfe 1973). More detailed studies have shown that the picture is much more complicated (Touret 1996). LILE depletion is not systematic but rather variable within granulite facies terranes (e.g. Moine et al. 1972; Rudnick et al. 1985). In some granulites, many minerals are actually LILE-enriched (Villaseca et al. 2007). Some, notably associated magmatic rocks, such as charnockites, may also be LILE-enriched. In some areas where this depletion exists, like the southern part of the Bamble sector, once taken as the most typical and extreme example of granulite facies LILE-depletion (Field et al. 1980), it has been shown that this depletion is related to the initial whole rock composition. Whereas several supracrustal rocks are not depleted at all (Moine et al. 1972), LILE-depletion of igneous rocks is a premetamorphic feature preserved during high grade metamorphism (Knudsen and Andersen 1999). Similar igneous, premetamorphic LILE-depletion was proposed by Rollinson and Tarney (2005). However, several other studies have demonstrated LILE and HFSE mobility during incipient charnockitization (e.g. Raith and Srikantappa 1993), which may affect granulite facies terrains in general (Hansen and Harlov 2007).

It seems finally that the only elements which can be transported over long distances outside of the granulite domain are those which are relatively soluble at granulite facies conditions and may remain in solution over a wide range of P-T conditions. Gold, U and some REE are good candidates, in contrast to alkali (K or Na) that are rapidly redistributed in the host rocks. Major minerals deposited from fluids during retrogradation are silica (quartz) and carbonates, in such quantities that the fluid source at peak conditions must have been very large indeed. It is likely that fluids remaining after mineral deposition may even reach the hydrosphere and atmosphere, with important climatic consequences (Touret and Huizenga 2012). The compositional similarities between these fluids and those occurring at peak conditions indicate a common source, namely late fluids collected in shear zones from peak metamorphic fluids under decreasing P-T conditions. The extent of these retrograde phenomena shows indeed that amount of *water undersaturated* fluid at

peak metamorphic P-T conditions must have been very great indeed, orders of magnitude more than the minute remnants now preserved in fluid inclusions.

## 11.6 Conclusions

The metamorphic transition from amphibolite to granulite facies is traditionally regarded as a transition from wet to dry rocks. In reality, granulites are all but fluid-free. Research over the past decades has shown two free fluid phases being present in granulites, i.e. CO<sub>2</sub> and highly saline brines. The transition from amphibolite to granulite facies does not only reflect an increase in temperature, but also a transition from a H<sub>2</sub>O-dominated metamorphic facies to a CO<sub>2</sub> and brine-dominated one. The two fluids present in granulite facies rocks are immiscible under granulite facies P-T conditions (Bowers and Helgeson 1983; Johnson 1991; Shmulovich and Graham 2004). They also have rather different transport properties and effects on the solubilities of minerals and elements. Their presence, however, implies that fluid-assisted reactions may occur, including removal and introduction of specific elements (metasomatism). Discussing the origin of these fluids is outside of the scope of this current review. However, it has been recently discussed at some length by Touret (2009) and Touret and Huizenga (2012), who basically conclude that CO<sub>2</sub> is dominantly, if not exclusively, mantle-derived. As deduced from coupled He-CO<sub>2</sub> fluxes, CO<sub>2</sub> flux from the mantle is too low to generate a regional scale amphibolite to granulite facies transition (O’Nions and Oxburgh 1988). The other granulite facies fluid, brines, is an essential companion. The question of their origin is more complicated. Some brines are probably locally derived, remnants of original sedimentary pore waters (Yardley and Graham 2002), which may have been possibly progressively concentrated during metamorphism (Kullerud 1996; Markl et al. 1998). Others, like CO<sub>2</sub>, may have been introduced from the underlying mantle by intrusions at peak metamorphic conditions (e.g. Hansen et al. 1995). These may ultimately be derived from remnants of subducted crust (Newton et al. 1998). Like for CO<sub>2</sub>, the carbonatite connexion cannot be excluded for brines. Brines and carbonatite melts are among the most typical inclusions found in diamonds (Klein-BenDavid et al. 2004; Araujo et al. 2009), in which part of the brines may be of juvenile mantle origin instead of oceanic crust incorporated in the mantle (Burgess et al. 2009). If CO<sub>2</sub> derives from the breakdown of metasomatic carbonate melts in the mantle, akin to carbonatites (Touret 2009), it seems probable that at least some brines also have this origin. In all cases, it should be realized that fluids, whether locally generated or introduced, may mix or change composition by reactive transport.

The widespread occurrence of metasomatic features does not preclude that, in the same areas and often within close range, pre-metamorphic features, such as whole rock chemistry reflecting surficial weathering like lateritization (Bol et al. 1989) or premetamorphic albitization (Owen et al. 2003), sedimentary structures (Nijland et al. 1993) and/or regional pre-metamorphic carbon and oxygen isotopic

trends in carbonates and graphites from metasediments (Broekmans et al. 1994; Bol et al. 1995; Boulvais et al. 1998) are still present, having survived both devolatilization and the action of granulite facies fluids. They have also survived melt extraction, another important process occurring widely in the middle and lower continental crust (Brown and White 2008). Overall, the lower continental crust appears to be a rather complicated domain, in which highly transformed and well-preserved rocks occur side by side, sometimes on the scale of the outcrop or even the hand specimen. Tracing the contributions from the different processes relating to granulite genesis and metasomatism during the transition from amphibolite to granulite facies will be a great challenge for future work.

**Acknowledgements** Successive versions of this paper have benefited from comments and/or discussions and/or (in)formal reviews by H. Austrheim T. Andersen, M. Cuney, D.E. Harlov, W.L. Griffin, R.C. Newton, D. Rumble and O. Safonov, as well as by careful editorial work by the editors of this volume. The senior author wants to acknowledge the constant support and inspiration he has got from Bob Newton during more than 40 years, as well as the technical support of the ENS team (C. Chopin, E. Charon) for a number of microphotographs.

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