

Diffusion-controlled biotite breakdown reaction textures at the solid/liquid transition in the continental crust

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Received: 2 January 2007 / Accepted: 17 May 2007 / Published online: 6 June 2007
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Abstract Two-phase quartz intergrowths with garnet, cordierite and tourmaline occur commonly in prograde high-temperature migmatites, granulites, as well as in the last crystallization stages of biotite granites. Structural, microtextural and mineralogical data show that they result from the breakdown of biotite in the presence of a melt phase associated with incongruent dissolution of feldspars into the melt and silica release (giving quartz in silica saturated rocks). Biotite breakdown and growth of Al-rich ferromagnesian minerals, occurring at the solid–liquid transition in the crust (early melting or final crystallization), is kinetically controlled by Fe and Mg mass transport, the network-forming cations Si and Al being locally compensated for by feldspar dissolution/crystallization. This process leads to significant changes with respect to equilibrium dehydration-melting reactions wherein quartz is a reactant and K-feldspar a reaction product. Therefore, quartz inclusions commonly occurring in garnets from granulite-facies metapelites and metagraywackes are not simply grains passively included during garnet growth. They may also correspond to newly crystallized phases. Resorption of feldspar may lead to more alkaline melt and to crystalline residue richer in Al than expected under equilibrium conditions. Hence, excess alumina in granulite-facies rocks is not necessarily related to initial alumina-rich whole-rock compositions (as currently considered), but may be due, at least partly, to kinetics of melting.

Keywords Chemical diffusion · Intergrowths · Garnet · Cordierite · Tourmaline

Introduction

Biotite, a common mineral of granites, is a fundamental phase in crustal anatexis and differentiation of the continental crust (Brown and Fyfe 1970). It has been shown that the amount of water available in the deep continental crust is unlikely to account for the large volumes of granite emplaced in the middle and upper continental crust (e.g. Clemens and Vielzeuf 1987), with consequence that much granitic magmas are initially water-undersaturated (Clemens 1984). Biotite breakdown and melt water-undersaturation have been accounted for by the two well-known models of “carbonic metamorphism” and “dehydration melting” (see Vielzeuf and Vidal 1990). Carbonic metamorphism (Newton et al. 1980; Touret 1971, 1989, 1992; Touret and Hartel 1990) involves the breakdown of OH⁻ bearing phases, notably biotite, in response to water activity change of the fluid phase. This change results from dilution by a CO₂-rich fluid either formed in situ by mineral reactions or streaming from deeper levels (e.g. Friends 1981; Janardhan et al. 1982; Raith et al. 1990; Touret and Hansteen 1988). Dehydration melting (Burnham 1967; Thompson 1982; Clemens 1984; Grant 1985; Vielzeuf and Schmidt 2001) involves the breakdown of OH⁻ bearing phases to produce water-undersaturated melt and anhydrous solids. Experimental investigations for various starting rock compositions (Le Breton and Thompson 1988; Vielzeuf and Holloway 1988; Rushmer 1991; Vielzeuf and Montel 1994; Patiño Douce and Beard 1995, 1996; Rapp and Watson 1995; Skjerlie and Johnston 1996) have shown the power of this model to understand both the

Communicated by J.L.R. Touret.

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formation and differentiation of the crust, as it accounts (i) for the overall mineralogy and geochemistry of granulites and granites; (ii) for the water-undersaturated nature of granitic magmas; (iii) for the production of significant amounts of melt; and (iv) for the generation of both granulites and granites in a unique process of crustal differentiation (Clemens 1991). Some consider that these two models are more complementary than exclusive (Touret and Dietvorst 1983; Touret 1989; Nair and Chacko 2002), whereas others contend that the properties of granitic magmas imply that their generation occurred in the absence of excess pervasive fluid (Clemens and Watkins 2001). Lastly, one has to remember that biotite breakdown can also occur under water-saturated conditions (e.g. Vielzeuf and Schmidt 2001; Fig. 1).

Whatever the conditions (hydrous, water-undersaturated, fluid-absent) partial melting, and magma crystallization as well, are generally thought in terms of equilibrium phase relationships and element partitioning, reaction kinetics being generally not taken into account. However, petrological and geochemical studies have shown that granitic melts formed either in lava xenoliths (Pushkar and Stoesser 1975; Kaczor et al. 1988) or in migmatites (Barbey et al. 1989; Sawyer 1991; Zeng et al. 2005) do not follow the expected isotopic or chemical equilibrium trends. Garnet microtextures in partially melted granulite-facies rocks (Waters 2001) and cordierite microtextures in granites (Barbey et al. 1999) also suggest a strong kinetic control of reaction processes. Moreover, melting experiments of F-phlogopite with quartz or plagioclase (Hammouda and Pichavant 1999, 2000) suggest

that for micas, rapid interface reactions may lead to chemical disequilibrium, due to dissolution rates faster than chemical exchange between crystals and melt. Disequilibrium may be preserved if melt extraction is faster than diffusive equilibration in crystals. Therefore, one may wonder whether diffusion-controlled reaction processes involving biotite are only limited to a few atypical situations, or whether they occur at larger scale in the crust in response to high degree of overstepping, then expressed by specific phase assemblages, textures and reaction processes.

This study deals with nodular to dendritic, quartz-bearing intergrowths involving either garnet, or cordierite or tourmaline. In addition to a review of the literature, we present new observations made from high-grade hornfels (Mauritania), metapelitic granulites (Finland, Cameroon, Corsica), high-temperature migmatites and granites (Velay massif in France, Mauritania). Microtextural and mineralogical data show the basic role of melt and feldspar in the formation of two-phase quartz intergrowths with an Al-rich ferromagnesian mineral. Then, we discuss the role of network-forming cations in biotite breakdown reactions at the liquid–solid transition and some possible implications.

Biotite breakdown reactions

Among the many reactions involving the breakdown of biotite (Fig. 1), a classical melt forming reaction in quartz-saturated metapelites and metagraywackes is $Bt + Pl + Als + Qtz = Opx \pm Grt + Kfs + Melt$ (Le

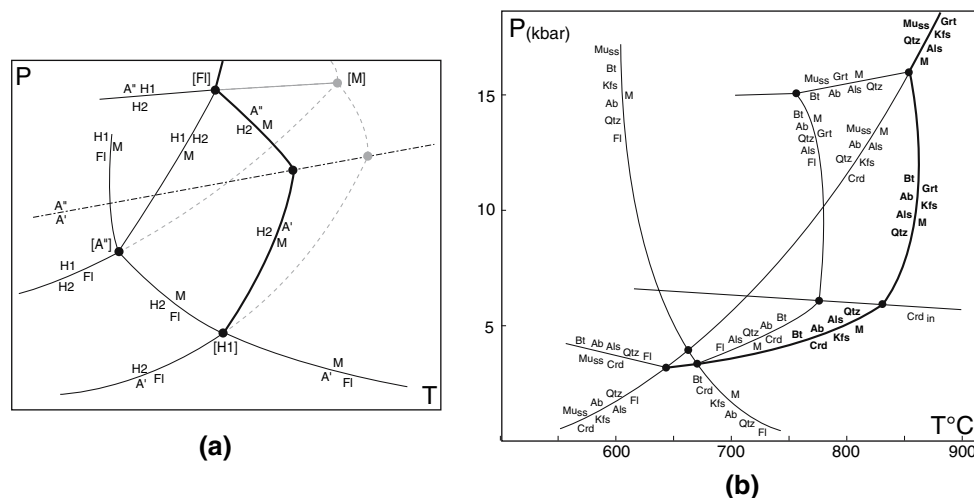


Fig. 1 **a** P-T diagram showing the topology of dehydration, hydrous melting (light lines) and dehydration melting (heavy lines) reactions for a binary system containing two hydrates (H1 and H2) and two anhydrous solids (A' and A''). Redrawn from Vielzeuf and Schmidt (2001). **b** Illustration of the same topology as in **a** in the case of quartz-saturated metapelites (KNFMASH system), showing some

classical reactions involving muscovite, biotite, cordierite and garnet (dehydration melting reactions with bold characters and heavy lines); note the location of quartz and K-feldspar on the low-T and high-T sides of the dehydration melting reactions, respectively. Redrawn and simplified from Vielzeuf and Holloway (1988) and Le Breton and Thompson (1988)

Breton and Thompson 1988; Vielzeuf and Holloway 1988). This example shows that a distinctive feature of the dehydration-melting reactions is that quartz is a reactant, whereas K-feldspar is a reaction product. Nevertheless, it has been emphasized that K-feldspar is not systematically present in experimental run products, depending on the biotite content of the starting material (Vielzeuf and Holloway 1988), or on the fact that the orthoclase component may be incorporated in plagioclase (Vielzeuf and Montel 1994). We shall see below that reaction kinetics may lead to reverse situations in which not only K-feldspar is a reactant, but also quartz is a reaction product, suggesting significant departure from the equilibrium dehydration-melting model.

Quartz-bearing intergrowths: textural and mineralogical data

Garnet and cordierite form frequently two-phase intergrowths with quartz (Barbey et al. 1999; Waters 2001). They occur in prograde high-temperature migmatites, in granulites and in the last crystallization stages of biotite granites. We summarize below the main data and provide new observations, which allow the reactions to be more tightly defined and generalized.

Garnet-quartz intergrowths

Waters (2001) described garnet-quartz intergrowths in granulite-facies pelitic and semi-pelitic migmatites from Namaqualand. Quartz systematically occurs in leucosome garnets as lobate or ovoid inclusions. The only quartz present in leucosomes may be in garnet. In aluminous rocks, garnet may have a core containing sillimanite and an outer part with large quartz inclusions. In some cases, optical continuity of quartz inclusions suggests the concomitant growth of quartz and garnet. Mass balance calculations (Waters 2001) show that the breakdown of biotite into garnet + quartz occurs at constant Si and almost constant Al, with loss of Na and K, and gain mainly in Fe. The garnet-quartz ratio in intergrowths indicates Al_2O_3/SiO_2 ratio of 0.3 close to the mean composition of the rock.

Garnet-quartz intergrowths from a migmatite in a contact aureole around a granite intrusion (Aguelt Nebkha, Reguibat Rise, Mauritania) bring further information on the exact nature of the reaction. A sample collected a few meters from the contact with the granite, corresponds to a fine- to medium-grained, Al-silicate-free ($A/CNK = 1.1$), felsic hornfels (metatuf). It consists of a matrix of K-feldspar, plagioclase (An_{17-33}), quartz and biotite ($Al_{tot} = 3.2-3.4$ apfu, $X_{Fe} = 0.67-0.75$), containing cm-sized garnet poikiloblasts ($X_{Alm} = 0.83$, $X_{Prp} = 0.11$) surrounded

by a thin, mm-thick, biotite-free halo. Garnet contains rounded quartz grains, which include minute rounded feldspar relics (Fig. 2c). In a same quartz grain, several feldspar inclusions may be in optical continuity, suggesting that they were once a single crystal. Isocon representation (Fig. 3a) shows that the breakdown of biotite into garnet + quartz occurs at almost constant Si and Al, with loss of Na and K.

Similar microtextures involving residual feldspars included in quartz of garnet are common in granulite-facies terrains, and express a process of broader interest than considered so far. A few examples of garnet-quartz intergrowths are presented here, namely the Palaeoproterozoic Svecofennian and Lapland granulites (Fig. 2d–f), the Pan-African Yaoundé granulites (Fig. 2g) and the Variscan Solenzara-Faoutea granulites (Lardeaux et al. 1990; Fig. 2h).

Interestingly, rounded quartz inclusions also occur in garnets formed during fluid-absent melting experiments on biotite-bearing metapelites and metagraywackes (Vielzeuf and Holloway 1988; Vielzeuf and Montel 1994; N. Le Breton, personal communication), showing that quartz inclusions are indeed produced during garnet growth.

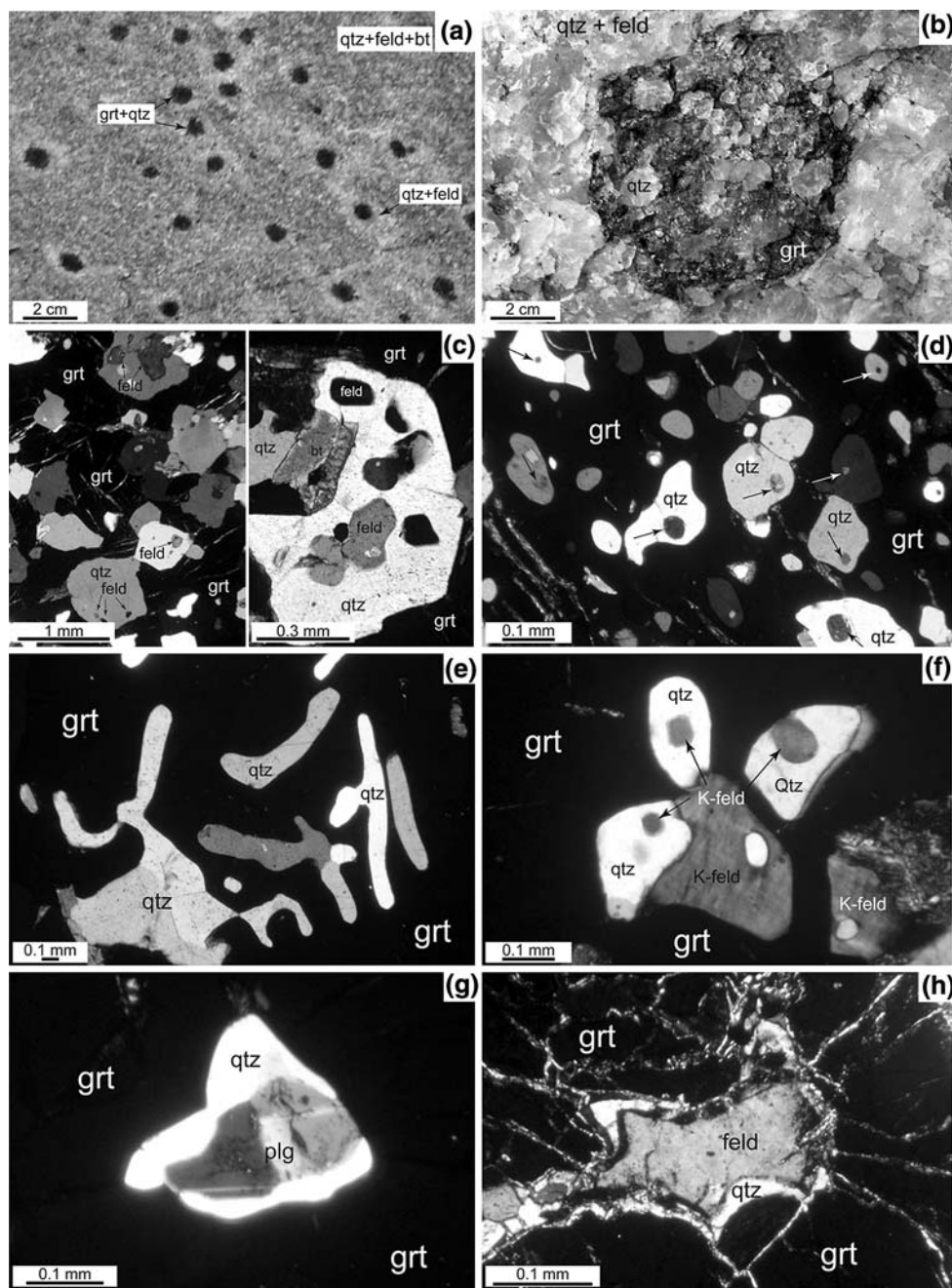
Cordierite-quartz intergrowths

Barbey et al. (1999) described cordierite-quartz intergrowths in high-T migmatites and granites from the Velay massif (France). Intergrowths form nodules and dendrites, which occur in cm-thick biotite-free halo in granite (Fig. 4a), or in migmatite leucosomes (Fig. 4b). The halo consists of quartz, euhedral to subhedral plagioclase and subhedral to interstitial K-feldspar. In both migmatites and granites ($A/CNK = 1.1$), nodules occur in the absence of sillimanite. Structural relationships between the nodular intergrowths and the granite fabric show that they grew before full consolidation of the host matrix. In the nodules and dendrites, both feldspars, when present, occur as resorbed grains locally rimmed with quartz (Fig. 4c). However, in most cases feldspars are totally resorbed, so only lobate quartz subsists (Fig. 4d). Optical continuity of quartz inclusions are common suggesting the concomitant growth of quartz and cordierite. Mass balance calculations made on cordierite-quartz intergrowths from granite show that their formation involves almost constant Si, Al and Fe, loss of Na, K and Ca, and gain in Mg (Fig. 3b). The radial structure of intergrowths, their occasionally highly dendritic shape and the presence of a biotite-free halo suggest that the reaction involving biotite breakdown is diffusion-controlled.

Tourmaline-quartz nodular intergrowths

Tourmaline-quartz nodular intergrowths, though commonly reported in granites (Nemec 1975; Samson and

Fig. 2 Examples of garnet-quartz intergrowth textures. **a–b** Intergrowths in prograde granulite-facies rocks, respectively: partially melted biotite metagranite and leucosome from a metapelitic granulite (Turku, Finland). **c** Garnet in a leucosome from a migmatitic metatuf (Reguibat Rise, Aguelit Nebkha, Mauritania), containing rounded quartz grains with minute corroded feldspar inclusions (crossed polars). **d** Rounded plagioclase and K-feldspar inclusions (arrowed) in quartz grains within a garnet porphyroblast (Svecofennian granulites, Anttola, southern Finland). **e** Lobate quartz crystals probably representing former rims at grain boundary intersections (Lapland granulites, Koppelo, northern Finland). **f** Rounded K-feldspar grains included in quartz; all feldspar grains belong to the same crystal (Lapland granulites, Inari, northern Finland). **g–h** Feldspars rimmed with quartz in garnet porphyroblasts from the Yaoundé granulites (Olembé, Cameroon) and Solenzara-Faoutea granulites (Anse de Tarcu, southeastern Corsica), respectively. *qtz* quartz, *feld* feldspar, *Kfs* K-feldspar, *plag* plagioclase, *grt* garnet

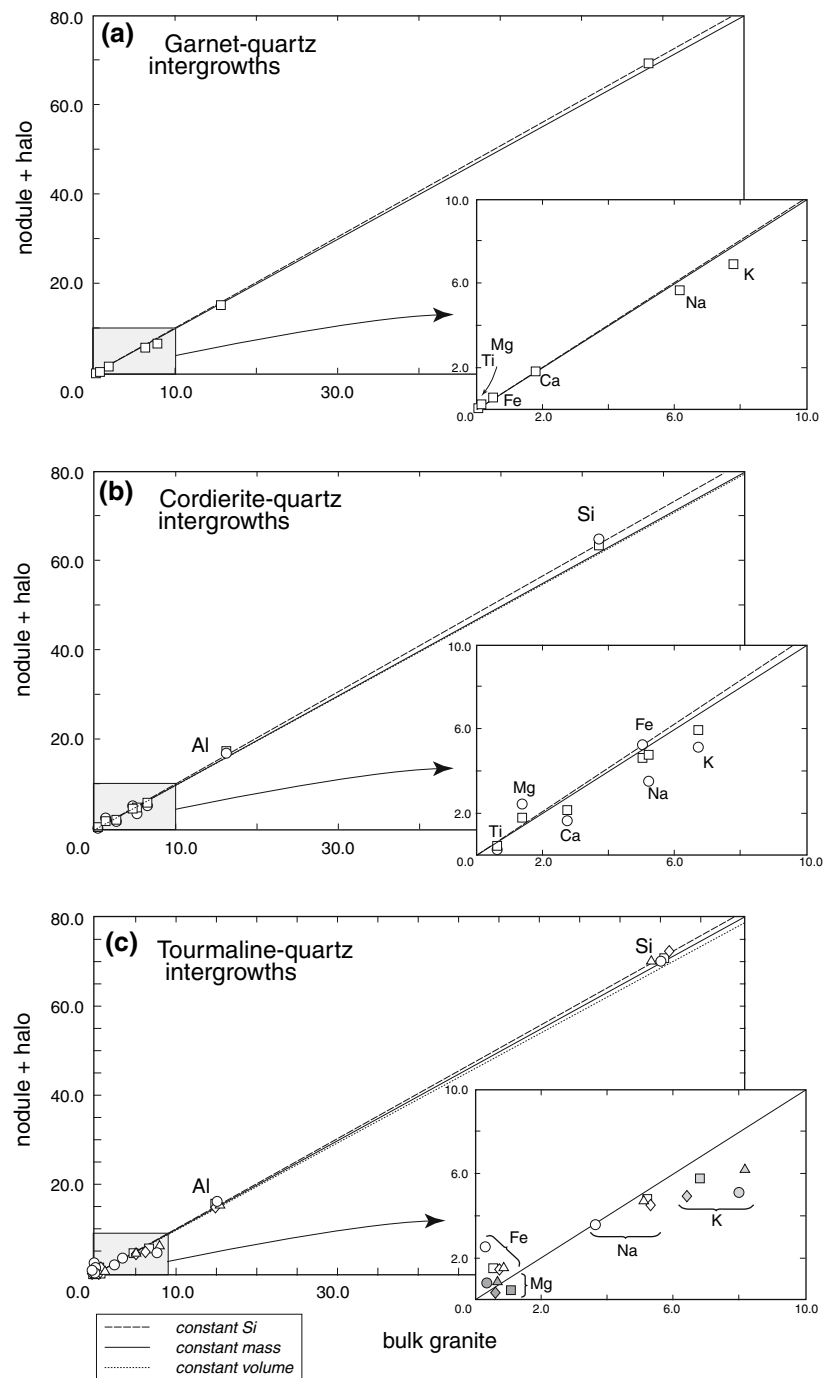


Sinclair 1992; Sinclair and Richardson 1992; Rozendaal and Bruwer 1995), also occur in high-T migmatites (Weber et al. 1985; Acosta-Vigil et al. 2001). For instance, in the Velay migmatites (French Massif Central) biotite-free leucosomes contain nodules consisting of tourmaline-quartz or cordierite-quartz intergrowths (Fig. 4b). Locally, tourmaline may be associated with cordierite in single nodules. In granites, intergrowths commonly occur as nodules scattered within granite free of pegmatite vein or of hydrothermal alteration (Fig. 4e). Tourmaline-quartz nodules are surrounded by a biotite-

free halo. Nemeč (1975) shows that the radius ratios of nodule:(nodule + halo) average 1:1.5. Residual corroded feldspar grains occur in quartz from intergrowths (Fig. 4f). Mass balance calculations using data from Nemeč (1975) show that quartz-tourmaline nodules grow at constant Si and Al, with loss of Na and K, and gain in Fe (Fig. 3c).

It must be noticed that tourmaline-quartz intergrowths occur also in association with pegmatites and greisen (Nemeč 1975; Sinclair and Richardson 1992; Rozendaal and Bruwer 1995). In that case, cores of nodules may

Fig. 3 a–c Isocon diagrams (Grant 1986) for garnet-quartz, cordierite-quartz and tourmaline-quartz intergrowths compared to their host-matrix. Data from Barbey et al. (1999), Nemeč (1975) and this study (see “Appendix”). Elements are given as atomic weight percent. Each *symbol* corresponds to an intergrowth



have miarolitic textures, with feldspars replaced by quartz-free tourmaline stringers, or are made of massive tourmaline. Note that in the Seagull batholith, nodules have lower tourmaline content at depth and lack miarolitic cavities, which are restricted to the top of the batholith (Sinclair and Richardson 1992). Therefore, tourmaline-quartz intergrowths are likely to develop under both magmatic and hydrothermal conditions (see Touret et al 2007).

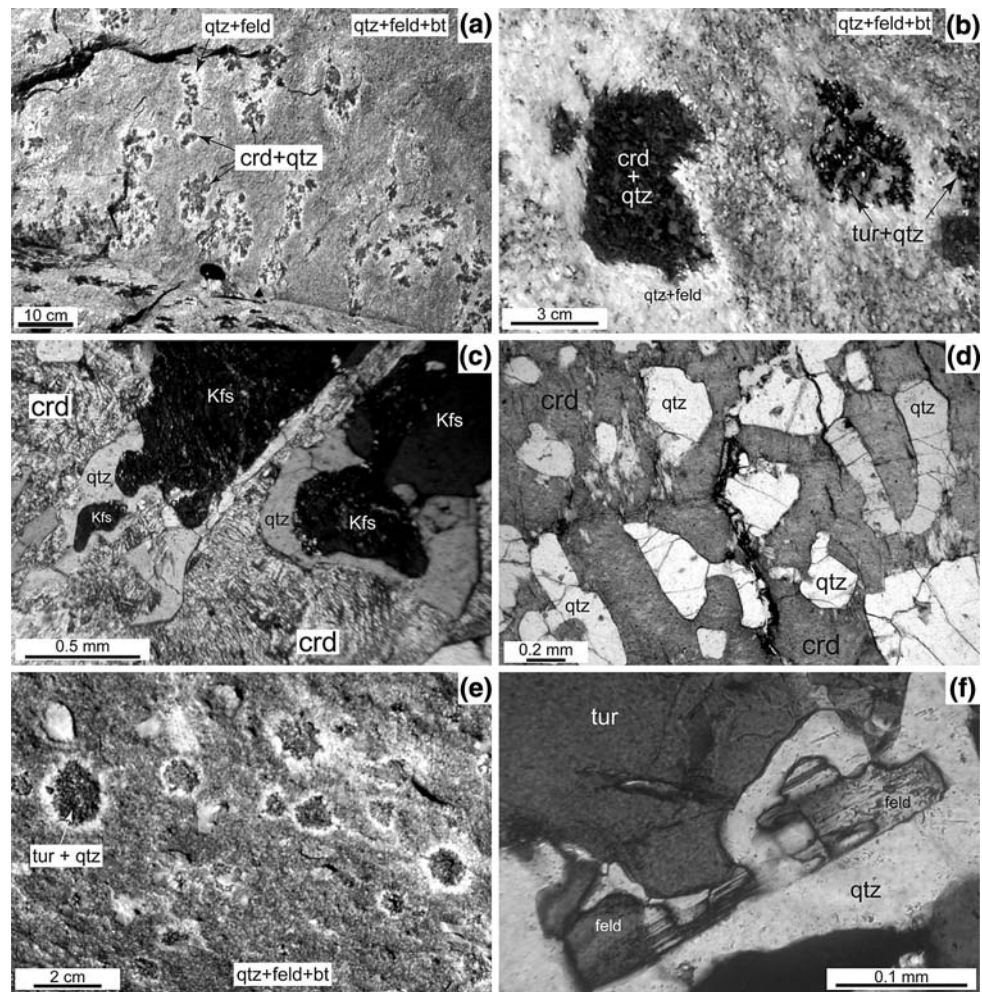
Evidence for the presence of a melt phase

The two-phase, garnet-, cordierite- and tourmaline-quartz intergrowths share common features: (i) occurrence in high-temperature, above solidus, biotite-bearing rocks regardless of the presence of Al-silicate minerals; (ii) radial symmetry with nodular to dendritic growth morphologies; (iii) shape-preferred orientation in deformed granites without evidence of plastic deformation; (iv) biotite-free

Fig. 4 Examples of quartz-bearing intergrowth textures with cordierite (Burzet, Velay dome, French Massif Central) and tourmaline (Aguelt Nebkha, Reguibat Rise, Mauritania; 25°35'568/7°24'423).

a Dendritic cordierite in a biotite granite. **b** Cordierite-quartz and tourmaline-quartz nodules within leucosomes in a biotite-bearing migmatitic orthogneiss. **c** Nodular cordierite (crossed polars, cordierite pinitized) containing residual feldspars rimmed with quartz. **d** Lobate quartz in cordierite probably representing former rims. **e** Tourmaline-quartz nodules in a fine-grained biotite granite. **f** Thin section (crossed polars) showing a corroded feldspar lath rimmed with quartz in a tourmaline-quartz intergrowth.

Abbreviations same as in Fig. 2



quartzofeldspathic halo; (v) presence of resorbed feldspars rimmed with quartz; and (vi) mass balance data suggesting quasi-constant Si and Al, with loss of Na–K and gain mainly in Fe or in Mg depending on the growing ferromagnesian mineral. These data point to a common kinetically controlled process involving a melt phase. The fact that garnet-, cordierite- and tourmaline-quartz intergrowths were made in the presence of a melt phase is supported by several observations.

- (1) Intergrowths occur within migmatite leucosomes formed under high-T conditions, well above the melting point of the granite system. In Namaqualand granulite-facies migmatites, garnet-quartz intergrowths formed at P–T conditions estimated at 800°C and 5 kbar (Waters 1997). Cordierite-bearing migmatites and granites in the Velay massif are considered to be formed at temperatures ranging from 760 to 850°C and pressures around 4 kbar (Montel et al. 1992).
- (2) The growth of quartz-bearing garnets in fluid-absent melting experiments (Vielzeuf and Holloway 1988;

Vielzeuf and Montel 1994; N. Le Breton, personal communication) is a fundamental observation, which argues in favour of melt-present reactions.

- (3) Evidence of synmagmatic deformation, without mineral plastic strain, of cordierite-quartz intergrowths in the Velay granite also indicates that they grew before full consolidation of host-granite (i.e. nodules elongated along the fabric of the host migmatite or granite, nodules within synmagmatic shear-zones, magmatic layering deformed around nodules, concentrations of residual melt in strain shadows of nodules; Barbey et al. 1999).

Quartz-bearing associations, and more specifically tourmaline-quartz intergrowths, are commonly attributed to hydrothermal solutions and base-cation leaching (e.g. Vernon et al. 1987; Rozendaal and Bruwer 1995). However, several data show that tourmaline growth can be magmatic, in pegmatites included. Firstly, field and experimental data on tourmaline stability in silicic magmas (Pichavant et al. 1988; Benard et al. 1985) show that tourmaline can be a restitic or early magmatic phase. Wolf

and London (1997) and London (1999) consider that tourmaline is likely to form during partial melting if the melt fraction remains low due to the high solubility of B in granite melts. Secondly, Touret et al. (2007) report, from fluid inclusion studies, a magmatic/hydrothermal transition in tourmaline-bearing miarolitic pegmatites. Moreover, even though the presence of miarolitic cavities in the core of nodules near the top of the Seagull batholith (Sinclair and Richardson 1992) implies the presence of an exsolved hydrothermal fluid, their disappearance at depth suggests a transition from purely magmatic (nodules) to fluid-saturated (miarolitic nodules) conditions of genesis. Thirdly, one argument used to support the hydrothermal origin of cordierite-quartz and tourmaline-quartz nodules is their alignment along what appears to have been fractures (e.g. Didier and Dupraz 1985; Rozendaal and Bruwer 1995), but brittle deformation is known to occur in the presence of a melt phase (e.g. Hibbard and Waters 1985; Bouchez et al. 1992; Vigneresse et al. 1996). Lastly, the occurrence of tourmaline-quartz intergrowths in migmatite leucosomes in association with cordierite further supports such a magmatic origin.

Discussion

Reaction controlled by chemical diffusion in melt

Diffusion-controlled transformation involving biotite breakdown is suggested by the presence of a biotite-free halo, as well as by the radial structure of intergrowths and their occasionally highly dendritic shape. As the intergrowth-forming phases are quartz and an Al-rich ferromagnesian mineral, the mobility of Si and Al is necessarily limited, in agreement with mass balance data and the phase rule in open systems (Korzinski 1959). Within nodules, plagioclase and K-feldspar are generally absent. When present, both feldspars are partially resorbed and surrounded by quartz rims, indicating that they are involved in the transformation. However, their stable association with biotite in the host rock, and with garnet, cordierite or tourmaline in the intergrowth, shows that feldspars are not directly involved in the biotite breakdown reaction, but more likely that they dissolve incongruently in the silicate melt, concomitantly with the reaction. This behaviour is a major difference with classical dehydration melting reactions where K-feldspar is a product, not a reactant. These observations further suggest that generation of the two-phase intergrowths cannot be ascribed to eutectic crystallization, but requires two coupled reactions. Biotite breakdown releases Fe and Mg, which diffuse in the melt and lead to growth of either garnet, or cordierite, or tourmaline. Feldspar incongruent

dissolution provides the necessary Al, releasing Na, K, Ca and Si into the melt, producing quartz at melt silica saturation. Deficit in Na–K–Ca in nodules and in Fe–Mg in the quartzofeldspathic halo indicates inter-diffusion of Fe–Mg and K–Na–Ca in the melt phase. The habit of garnet, cordierite and tourmaline is probably partly due to growth at the interface with the interstitial melt in a highly crystalline mush. However, this does not rule out the possible role of undercooling for tourmaline growth (Swanson et al. 2001), or of overstepping equilibrium for the strongly dendritic cordierites formed at high-T decompression (Barbey et al. 1999).

Reaction process at the solid–liquid transition

If all intergrowths are assumed to occur in the presence of a melt phase, the different types have to be related to different environments. Cordierite-quartz intergrowths in granites and migmatites result from decompression at high-T in a partly crystallized mush, or in partially melted gneisses (Barbey et al. 1999). Garnet-quartz intergrowths from migmatites and granulites are considered to result from prograde biotite dehydration melting reactions (Waters 2001). Tourmaline-quartz intergrowths in granite are currently related to cooling in a crystallizing granitic melt (e.g., Sinclair and Richardson 1992; Touret et al. 2007). Therefore, all three types of intergrowths correspond to reactions occurring at the solid–liquid transition, though resulting from distinct P-T paths.

Due to the very slow diffusivity of ionic species in solids, it is likely that diffusion is restricted to the melt phase, and that garnet, cordierite and tourmaline grow at the melt interface. Therefore, the presence of an interconnected melt network in the crystalline mush seems to be necessary to Fe–Mg and K–Na–Ca interdiffusion, at least close to the nodule and its quartzofeldspathic halo. Conservation of network forming cations Si and Al, and evidence for local feldspar dissolution with quartz growth suggest that there is no long range transfer of Si and Al, in agreement with their slow diffusivity in silicate melts (Liang et al. 1996). Nevertheless, partitioning of Fe and Mg between biotite and the growing Al-rich ferromagnesian minerals seems to follow equilibrium, with biotite X_{Mg} lower than that of cordierite, but higher than that of garnet and tourmaline (see for instance Thompson 1976; Wyborn et al. 1981; Clemens and Wall 1988). In a same way, growth of either garnet, or cordierite, or tourmaline also follows their mutual stabilities relative to biotite, which implies some sort of equilibrium.

We suggest to consider the bulk transformation as the combination of two reactions, one at the site of biotite resorption (1), the other at the site of growth of the Al-rich ferromagnesian mineral (2). For example, for garnet in a

- (ii) The slow diffusivity of Al and Si introduces significant modifications with respect to the classical dehydration melting reactions investigated experimentally, in which quartz is one of the reactants and K-feldspar a reaction product. The behaviour of network-forming cations explains, among others, why K-feldspar does not systematically appear as a reaction product both in experimental runs and natural examples, and why garnets (cordierites) from high-temperature migmatites (granites) are commonly riddled with lobate quartz inclusions. A rapid survey of a few granulite-facies rocks from different areas and ages show that the transition to granulite-facies conditions is kinetically controlled, and that the model dehydration melting reactions do not always provide an exact picture of what happens at the amphibolite/granulite facies transition.
- (iii) Resorption of feldspar induces release of alkalis into the melt, which leads to more alkaline melt than expected under equilibrium conditions. Symmetrically, it gives rise to crystalline residue richer in Al, as alumina for garnet or cordierite is not provided by Al-silicates but by feldspars. Therefore, excess alumina in granulite-facies rocks is not necessarily related to initial alumina-rich whole-rock compositions (as currently considered), but may be due, at least partly, to kinetics of melting. Our present data do not allow the extent of the modification induced by disequilibrium resorption of biotite to be quantified. However, the widespread presence of quartz-bearing garnet in semi-pelitic granulites suggests that it should not be negligible.

Acknowledgments Thanks to M. Pichavant for thoughtful comments on an early version of this manuscript, N. Le Breton for discussion on dehydration melting reactions, and G. Libourel for providing me with samples of Solenzara granulites. Thoughtful comments and suggestions by an anonymous reviewer and J. Touret are gratefully acknowledged.

Appendix

Major element composition of garnet-quartz intergrowth and related matrix from the Reguibat Rise, Mauritania (Aguelt Nebkha area, 25°01'929/7°12'448).

SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	TiO ₂	P ₂ O ₅	LOI	Total
73.05	14.58	0.37	–	0.15	1.25	4.12	4.68	0.06	0.17	0.68	99.11
Garnet-quartz intergrowth											
74.67	14.58	0.38	–	0.19	1.24	3.84	4.18	0.01	0.16	0.69	99.94

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