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Fluid-assisted granulite metamorphism: A continental journey

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ABSTRACT

Lower crustal granulites, which constitute the base of all continents, belong to two series: high-pressure granulites generated by crustal thickening (subduction) and (ultra)high-temperature granulites associated with crustal extension. Fluid inclusions and metasomatic features indicate that the latter were metamorphosed in the presence of low-water activity fluids (high-density CO_2 and brines), which have invaded the lower crust at peak metamorphic conditions (fluid-assisted granulite metamorphism). High-pressure and (ultra)high-temperature granulites commonly occur along elongated paired belts. They were formed, from the early Proterozoic onwards, during a small number of active periods lasting a few hundreds of m.y. These periods were separated from each other by longer periods of stability. Each period ended with the formation of a supercontinent whose amalgamation coincided with low- to medium pressure (ultra)high-temperature granulite metamorphism, immediately before continental break-up. It is proposed that large quantities of mantle-derived CO_2 stored in the lower crust at the final stage of supercontinent amalgamation, are released into the hydro- and atmosphere during breakup of the supercontinent. Fluid-assisted granulite metamorphism, therefore, appears to be an important mechanism for transferring deep mantle fluids towards the Earth's surface. Possible consequences were, for example, the sudden end of Proterozoic glaciations, as well as the post-Cambrian explosion of life.

Contents

1. A granulite lower crust

In the 1960s, Russian scientists, including V.S. Sobolev at Novosibirsk and V.V. Belousov at Moscow, realised that under a thin sedimentary cover continents were not a homogeneous mass of granites as was initially proposed by Edward Suess (Suess, 1885–1909). It appeared that, similar to the oceanic crust, the continent crust is stratified (e.g., Ramberg and Smithson, 1975) comprising three metamorphic zones of roughly the same thickness: low- to medium-grade metamorphites in the upper crust, granitic migmatites in the middle crust, and granulites in the lower crust. Magmatic rocks constitute indeed a significant part of the two lower zones, but they do not form continuous layers. Rather,

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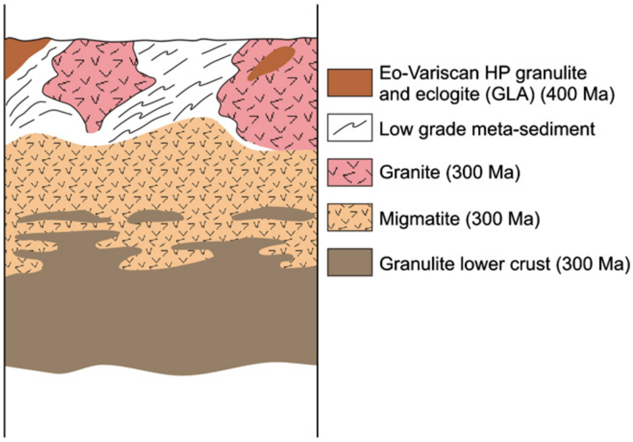

Fig. 1. Simplified crustal section for the Massif Central (France) (Touret, 2009). See text for further explanation.

they comprise discrete bodies of S-type granites and mantle-derived gabbroic intrusions, which become more abundant near the crust–mantle boundary (Bohlen and Mezger, 1989).

Fig. 1 shows a typical example of the Variscan continental crust under the French Massif Central, which forms part of the north Gondwana margin (e.g., Faure et al., 2009). Two types of granulite-related rocks occur in the Massif Central (Pin and Vielzeuf, 1983): (1) high-pressure (HP) granulites and eclogites (P = 1.4–1.6 GPa, T = 700–830 °C) (Gardien and Lardeaux, 1991; Bellot and Roig, 2007), belonging to the so-called "Groupe Leptyno-Amphibolique" (GLA) that occur at the surface. These GLA rocks have been dated at about 400 Ma (Eo-Variscan) and were formed as a result of the closure of the ocean between Laurasia and Gondwana (Matte, 1986), and subsequently exhumed through a complicated system of near horizontal thrust planes and nappes (e.g., Ledru et al., 2001). (2) High-temperature (HT) and low-pressure (T = 760–850 °C, P = 0.4–0.5 GPa) granulites (Montel et al., 2007) that constitute the present-day lower continental crust and occur at the surface as xenoliths in younger volcanic rocks (Dupuy et al., 1977). The age of the xenoliths protolith is Proterozoic (700–1000 Ma, Downes and Leyreloup 1986), and the age of granulite metamorphism is ~100 Ma younger than for GLA (Pin and Vielzeuf, 1983) (Fig. 1). This Carboniferous (~300 Ma) age corresponds also to the widespread emplacement of granites into the Massif Central middle crust (Ledru et al., 2001). Subsequent studies on the composition and structure of the continental crust (e.g., Rudnick and Fountain, 1995) have demonstrated that the Massif Central can be used as a general example for the continental crust composition worldwide. Granulites, once considered as a petrographic curiosity, are the dominant, if not unique, constituent of the lower continental crust.

2. Granulite metamorphism: fluid-absent or fluid-assisted?

The granulitic character of the lower crust has led to a debate that is still continuing amongst metamorphic petrologists. The fact that granulites are metamorphosed above the minimum granite-melting temperature, together with a wealth of experimental data and paragenetic analyses, has led to the concept of fluid (or vapour)-absent granulite metamorphism (Thompson, 1983). In this model, the anhydrous mineral phases (orthopyroxene, garnet) that characterise the granulite mineral assemblages result from dehydration melting (LeBreton and Thompson, 1988; Clemens, 1990): water-bearing mineral phases, including micas and amphibole, are progressively broken down, producing a granitic melt that migrates towards the middle or upper crust. Fluids liberated during this process are dissolved in the melt, leaving a dry, anhydrous granulite residue. In other words, H_2O cannot exist as a free fluid phase in the rock system; it is either dissolved in melts or bound in mineral structures.

The discovery of abundant fluid inclusions in granulite anhydrous mineral phases (for the first time described in Southern Norway by Touret in 1971) in many granulite terranes worldwide (Fig. 2), has led to a different idea, that we will refer to as the "fluid-assisted model". Here, low H_2O-activity fluids, i.e. CO_2 and aqueous brines (e.g., Newton

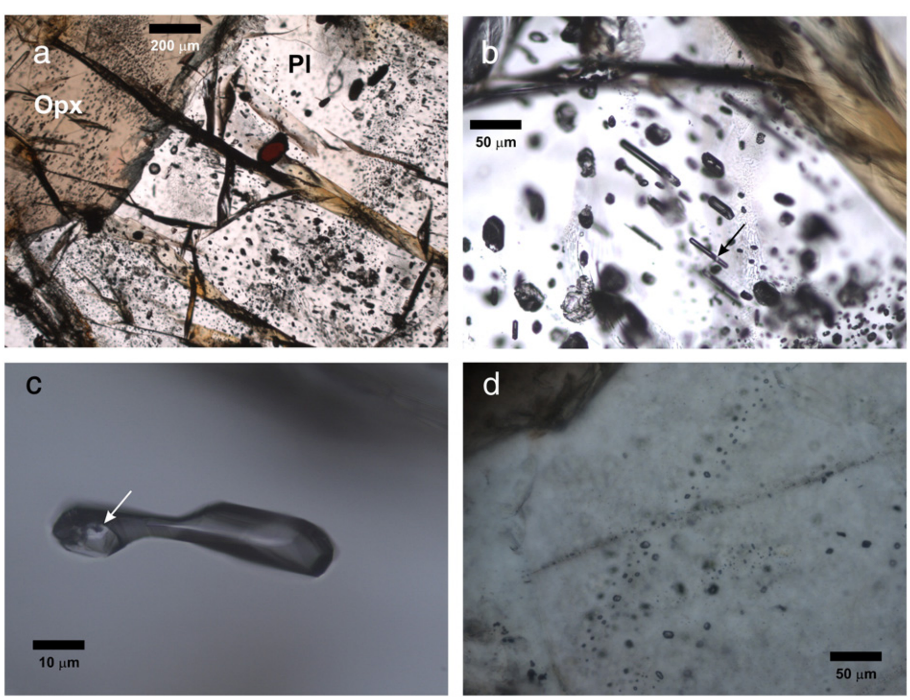

Fig. 2. Examples of CO_2 inclusions in sapphirine-bearing granulites from central Sri Lanka (Bolder-Schrijver et al., 2000) (a–c) and the Southern Marginal Zone of the Limpopo Complex (d) (Van den Berg and Huizenga, 2001). (a) Abundant primary fluid inclusions in orthopyroxene and plagioclase. (b) Primary fluid inclusions in garnet. Arrow: dolomite daughter mineral. The relative constant size of the dolomite daughter mineral in the fluid inclusions indicates that the fluid was a homogeneous mixture of CO_2 and carbonate (c) High-density CO_2 inclusion in garnet. Arrow: dolomite daughter mineral. (d) Typical trail of CO_2 inclusions in quartz. All photographs are taken in plane polarised light.

et al., 1998), coexist with granulite facies mineral assemblages at peak metamorphic temperatures, typically between 700 and 1000 °C. Fluid-absent versus fluid-assisted granulite models have led to a vivid controversy which, after more than 40 years, is not completely over yet (e.g., Newton, 2011; Rigby and Droop, 2011). As this issue is discussed in detail in other papers (Touret and Huizenga, 2011; Touret and Nijland, in press), we will only recall here the main arguments which, in our view, support the fluid-assisted model including high-grade metasomatic features, fluid inclusions, and retrograde fluid-related alteration.

Oddly enough, although metasomatism was theoretically predicted by D.S. Korzhinski about 50 years ago (Korzhinski, 1962), its consequences were ignored or not observed for years in many granulites, at least outside the former Soviet Union, although they are quite common (see review by Touret and Nijland, in press). The most spectacular evidence of metasomatism are the incipient charnockites (e.g., Santosh et al., 1990; Newton, 1992; Harris et al., 1993), first found in India and Sri-Lanka, but later also observed in many other regional granulite terranes (e.g., Santosh and Omori, 2008a, see details in Touret and Nijland, in press). The transformation of a variety of lithologies (metapelites, granitic or granodioritic gneisses) into granulites or charnockites may occur along brittle fractures which serve as pathways for the incoming fluids. Melting may have occurred (e.g., Newton, 1992), especially in the case of the patchy charnockites (Harris et al., 1993). But even then, the lack of restitic material indicates that melting has been preceded and was accompanied by metasomatic changes (Perchuk and Gerya, 1992, 1993). Incipient charnockites are so obvious and spectacular that one can wonder why, after the initial discovery by S. Pichamutu in 1961, they were almost forgotten for about 20 years before being suddenly rediscovered when the role of fluids became a critical issue in granulite metamorphism.

Other metasomatic evidence is more difficult to observe as it occurs only along grain boundaries. Firstly described in Southern India and in territories belonging to the former Soviet Union (Perchuk and Gerya, 1992), it was later found to be widespread within most granulite terranes like, for example, the Ivrea-Verbano zone, Northern Italy (Harlov and Wirth, 2000, Harlov and Förster, 2002), or the Limpopo Complex in Southern Africa (Touret and Huizenga, 2011). It includes quartz–plagioclase intergrowths (myrmekite) occurring along the borders of K-feldspar (Fig. 3a–c) and K-feldspar microveins at intergrain boundaries (Fig. 3a,d). Myrmekites, notably, can be exceptionally abundant and large in some granulites, particularly in cordierite-bearing varieties (e.g., giant myrmekites, Touret and Huizenga, 2011). Both myrmekites (Fig. 3a–c) and K-feldspar microveins (Fig. 3a,d) occur commonly near each other, implying that they can be interpreted as complementary processes (Touret and Nijland, in press). We consider that myrmekites are formed by the well-known reaction proposed by Becke (1908) (interestingly enough the first interpretation ever proposed amongst all others to be found later in the literature, see e.g., Simpson and Wintsch, 2007). In this reaction, K-feldspar is replaced by plagioclase and quartz under the influence of Ca and Na-bearing solutions, according to the reaction: $1.25\ KAlSi_3O_8 + 0.75\ Na^+ + 0.25\ Ca^{2+} + 2\ H_2O \rightarrow Na_{0.75}Ca_{0.25}Al_{1.25}Si_{2.75}O_8 + 1.25\ K^+ + SiO_2$. This reaction results in the enrichment of the fluid in silica and K^+, which may cause the precipitation, at close distance, of quartz blebs permeating the rock (Fig. 3b), and/or newly formed K-feldspar microveins (Fig. 3d). It is clear that such alkali mobility and the formation of myrmekite and K-feldspar can only be achieved by brine fluids that can easily move along grain boundaries (Newton and Manning, 2000). Remnants of these brine fluids are commonly found in the immediate vicinity of these metasomatic features, either in the quartz blebs of the myrmekite (e.g., Fig. 5b in Touret, 2009), or in quartz crystals adjacent to the microveins (Fig. 3d).

In addition to metasomatic features, fluid inclusions comprising CO_2 and brines also indicate that a fluid phase was present at and after peak granulite metamorphism. Inclusions formed during prograde metamorphism are exceedingly rare compared to inclusions that are

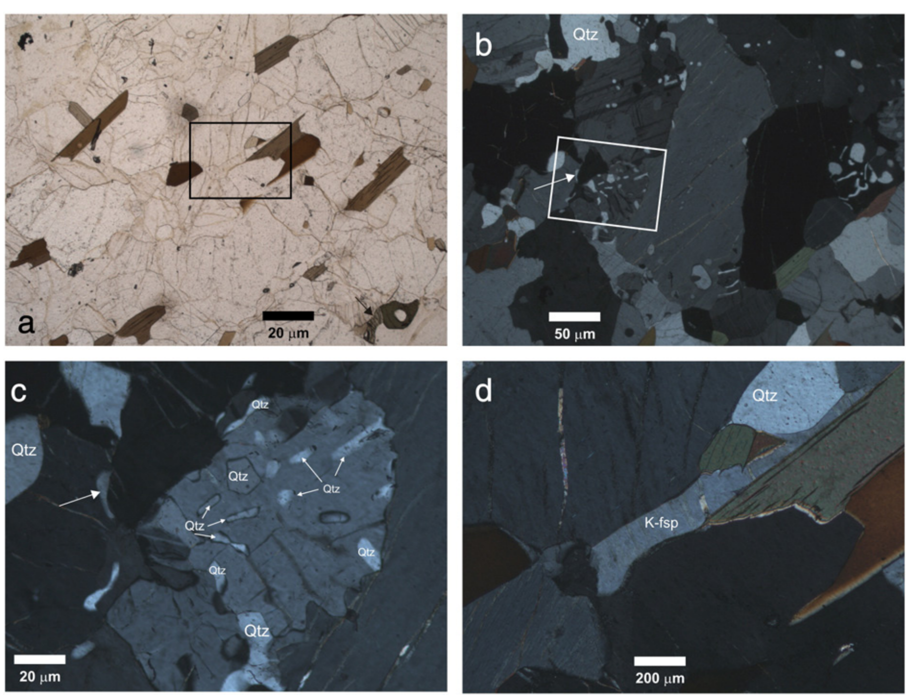

Fig. 3. Metasomatic features caused by percolating brines at grain boundaries (for more details, see Touret and Nijland, in press) in incipient charnockites from Kurunegala in Sri Lanka (Perchuk et al., 2000). (a) Overview of the charnockite comprising quartz, feldspar (mesoperthite), biotite and orthopyroxene (arrow). Black rectangle: field of view of shown in (d). (b) Myrmekite and K-feldspar microvein (arrow) around large mesoperthite crystals. White dots in the upper part of the photograph are quartz blebs with identical optical orientation that are formed from the myrmekite reaction (Touret and Nijland, in press). White rectangle: field of view shown in (c). (c) Detail of the myrmekite. Arrow: K-feldspar microvein along the boundary of two mesoperthite crystals. (d) Detail of a K-feldspar microvein. These veins develop along the boundaries of different minerals (mesoperthite, biotite, quartz), always with well-defined boundaries indicating that they were formed from a percolating fluid. Traces of these fluids are found in the quartz, appearing as small black specks. All photographs are taken with crossed polars except for (a).

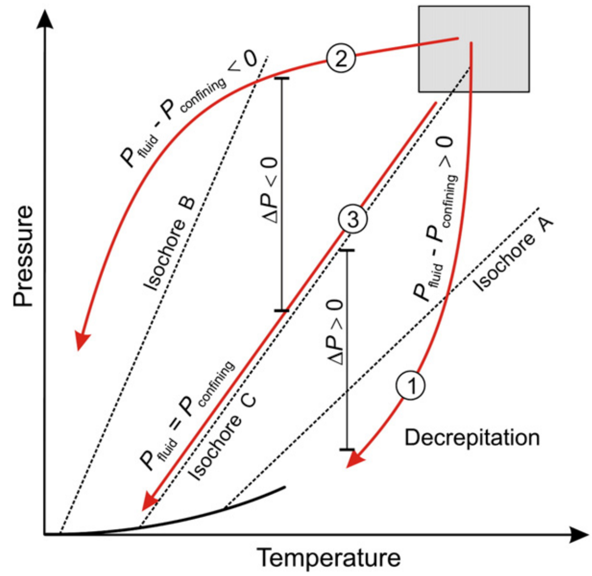

Fig. 4. Behaviour of fluid inclusions during the retrograde *P–T* evolution. *P–T* path 1: near isothermal decompression results in an overpressure of the fluid phase and may lead to inclusion decrepitation. Inclusions that are formed as a result of the decrepitation will have a density corresponding to that of isochore A. *P–T* path 2: near isobaric cooling results in an underpressure of the fluid phase and may lead to inclusion implosion. Inclusions that are formed as a result of the decrepitation will have a density corresponding to that of isochore B. *P–T* path 3: near isochoric decompression-cooling, neither decrepitation nor implosion will occur. This can explain the preservation of high-density CO_2 inclusions in granulites, which have followed a near-isochoric retrograde *P–T* path (e.g., Touret and Hansteen, 1988).

formed during peak or especially retrograde metamorphic conditions (Touret, 2001; Rigby and Droop, 2011). Except for very late fluids, in most cases low-salinity aqueous meteoric solutions, retrograde inclusions in granulites are comparable in composition with peak metamorphic fluids, but they differ grossly in density. This density difference depends to a large extent on the shape of retrograde *P–T* path, i.e. the fluid density in later inclusions decreases for isothermal decompression, whereas it increases in the case of isobaric cooling (Touret, 2001) (Fig. 4). This re-equilibration of fluid densities occurs by a series of ruptures (often called decrepitation in the fluid inclusion literature, see Fig. 4) followed by immediate re-trapping of the liberated fluid. New generations of fluid inclusions are thus formed, according to a phenomenon that fluid inclusion researchers refer to as “transposition”.

The great number of successive generations of CO_2-bearing fluid inclusions found in many granulites, as well as the great number of well preserved peak metamorphic inclusions, which have followed a “pseudo-isochoric” retrograde *P–T* path (path 2 in Fig. 4), is a first indication that a relatively large quantity of low H_2O-activity fluids was present at peak metamorphic conditions. Except for remnants preserved in inclusions, these fluids did not survive the end of the metamorphic episode. The abundance of secondary CO_2-bearing trails (e.g., Touret, 2001; Van den Berg and Huizenga, 2001) (Fig. 2d) indicates that CO_2 migration occurred through successive episodes of microfracturing during the retrograde evolution. Trails of aqueous brine inclusions, on the other hand, are far less common. This is due to the fact that brines move mainly along grain-boundaries because of their low wetting angle at the mineral interface (Gibert et al., 1998). Evidence for their presence is thus from the above mentioned metasomatic features (K-feldspar microveins, myrmekites) rather than from remnants preserved in inclusions.

The final argument supporting the idea of large fluid quantities at peak granulite conditions is the impressive traces that they, especially CO_2, have left when they were expelled from the rock system along large-scale shear zones during the last stages of retrogradation (Newton and Manning, 2002). Examples of these occurrences include the Late Proterozoic Attur Valley of Tamil Nadu, India (Wickham et al., 1994) and Southern Norway (Dahlgren et al., 1993) where the country rocks have been replaced by carbonates with a uniform $\delta^{13}C$ mantle signature (Newton and Manning, 2002). Another line of evidence indicating the movement of granulite (mantle) fluids along shear zones is the association of Archean lode gold deposits with mantle-derived CO_2 (Cameron, 1988).

As the fate of granulite fluids during retrogradation is of prime importance for the scope of this paper, a few examples of Southern Norway will be discussed in some more detail. Fig. 5 illustrates two cases of hydrothermal carbonates in the Bamble Shear Belt, one (Fig. 5a) is a breccia-type that occurs as irregular patches within partly amphibolitised meta-gabbros (Kamerfoss, outcrop described in Dahlgren et al., 1993), the other (Fig. 5b) occurs in a core of a straight metasomatic vein cutting massive, garnet-bearing amphibolite. Both amphibolites grade progressively in sub-ophitic metagabbro (syn-metamorphic intrusions, formerly described as “hyperites”, see below). Details of Fig. 5b, an outcrop recently discovered and described by Timo Nijland, illustrate the variety of metasomatic processes, which have occurred in this region during the vanishing stages of granulite metamorphism. From the amphibolite towards the vein, over a distance of a few decimetres, high-grade minerals (plagioclase, hornblende, garnet) are first replaced by albite, scapolite and phlogopite, then by massive actinolite that is surrounded by albite fading away on both sides of the vein, and finally (in the core of the vein) by massive yellowish carbonates. These carbonates are strongly reminiscent in colour and have a similar appearance to those found in Kamerfoss.

Many years of field experience in Southern Norway has convinced the senior author that outcrops like those shown in Fig. 5 are far more abundant than commonly realised. It is noteworthy to mention that

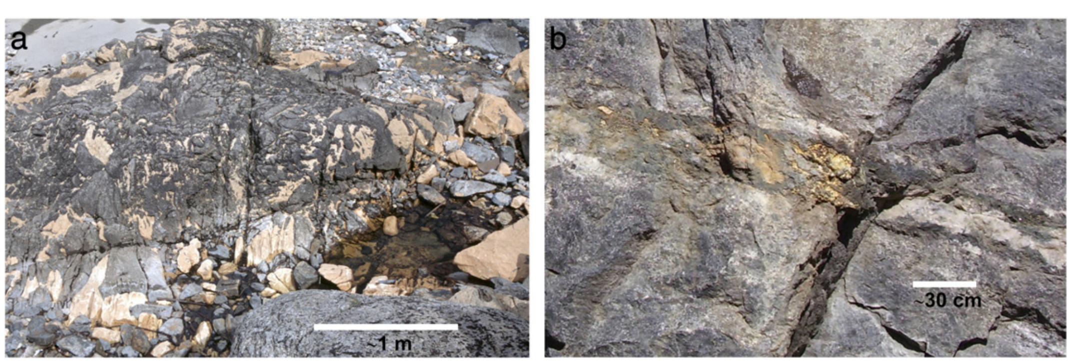

Fig. 5. (a) Hydrothermal dolomite (light brown) in meta-gabbro (dark), emplaced at the end of Sveconorvegian metamorphic event (Kamerfoss, Bamble Province, Norway) (Dahlgren et al., 1993). (b) Metasomatic albite and actinolite-bearing vein in garnet-bearing amphibolite from Southern Norway (Road 42, Froland Verk to Osedalen). White: albite, black (margin of the vein): massive actinolite, light brown: hydrothermal carbonates (like in Kamerfoss). Photo courtesy Timo Nijland.

the two first stages of the metasomatic processes illustrated in Fig. 5b, namely albitisation and scapolitisation, have in some places such a regional extent that they have led to significant ore deposits. Albitisation is accompanied by the destabilisation of ilmenite to produce rutile (Kragerö) and, depending on the nature of the host rock, veins like those represented in Fig. 5b may contain important quantities of apatite (Odegarden), enough to have sustained mining activities in the region until the end of the 19th century. These occurrences, first described in Southern Norway by Bugge (1943) (then referred to albite–carbonate deposits), have recently been studied in great detail by Engvik et al. (2011). They are not restricted to Southern Norway but occur in virtually all exposed deep-crustal terranes worldwide, with major metallogenic consequences including large scale albitite-hosted uranium deposits in Central Sweden and Brazil (Cuney and Kyser, 2009), and the majority of lode gold deposits worldwide (Newton and Manning, 2002). All these retrograde metasomatic features show that (1) deep crustal fluids were able to reach upper crustal levels at the end of the metamorphic episode and (2) the amount of granulite fluids must have indeed been very large.

The great amount of experimental work done, in particular by R.C. Newton and C. Manning at UCLA (e.g., Newton and Manning, 2010), and L.L. Perchuk and co-workers at the Institute of Experimental Petrology (Moscow–Chernogolovka) (e.g., Perchuk et al., 2000; Safonov et al., 2009) indicates that the geochemical role of both granulite fluids, namely CO_2 and brines, is strikingly different. Most mineral species have a very low solubility in CO_2, implying its limited role as a transport medium (Newton and Manning, 2010). Brines, on the other hand, can act as potent solvents at lower crustal and mantle conditions (Newton and Manning, 2000, 2002, 2010). Their geochemical influence on mineral and rock compositions in the lower crust and the upper mantle is, therefore, extremely important.

In conclusion and in line with previous publications (e.g., Touret and Huizenga, 2011; Touret and Nijland, in press), we will accept the model of fluid-assisted granulite metamorphism and discuss its bearing on continent formation and evolution. We emphasise that the fluid-assisted model is by no means contradictory, but rather complementary to the fluid-absent model. As shown by the study of fluid inclusions in migmatites (Touret and Dietvorst, 1983) dehydration melting is an efficient, but not the only possible mechanism to produce low-H_2O activity fluids. Alternatives include the reaction of H_2O with former organic matter (graphite) (e.g., Stevens, 1997), preferential dissolution of H_2O into granitic melts (Fyfe, 1973), persistence throughout the whole metamorphic evolution of sedimentary pore fluids, or introduction at peak-conditions of external (mantle-derived) fluids. Field evidence and isotopic data support the idea that most CO_2 found in inclusions originates by the last mentioned mechanism (mantle derivation) (Touret, 2009) whereas, at least in Southern Norway, brines are associated with evaporites (Touret, 1979; Engvik et al., 2011).

3. *P–T* conditions of granulite metamorphism

3.1. Amphibolite/granulite transition

Many studies on the *P–T* conditions of granulite terranes have resulted in the identification of numerous subfacies in *P–T* space (Fig. 6) (e.g., Brown, 2007a). Typical for this type of diagram is that the transition between amphibolite and granulite facies is always represented by a narrow temperature range (e.g., Spear, 1993). This implies that at a given pressure, the amphibolite/granulite transition is solely temperature dependent. This is not always true; in unique areas where mineral isograds and isotherms have been precisely identified in the field (e.g., Southern Norway) it is clear that isograds and isotherms are not parallel. This discrepancy can be explained by the role that CO_2 can play in decreasing the H_2O activity, i.e. causing the partial pressure of H_2O (P_{H_2O}) to become less than the lithostatic pressure ($P_{lithostatic}$). The amphibolite/granulite transition should rather be indicated by a temperature range (blue area in Fig. 6) in which both amphibolites and granulites can exist due to the fact that $P_{H_2O}=P_{lithostatic}$ for amphibolites and $P_{H_2O}<P_{lithostatic}$ for granulites. In the case of Southern Norway, traces of brine activity are seen in amphibolite-facies rocks through the presence of myrmekites (Touret, 1985). This implies that brines are not related to granulite metamorphism, i.e. the deviation of P_{H_2O} from $P_{lithostatic}$ at the amphibolite/granulite boundary is caused primarily by the dilution of H_2O with CO_2.

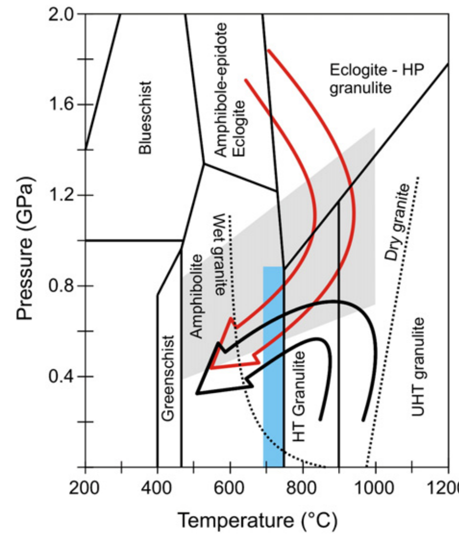

Fig. 6. *P–T* diagram showing the HP and (U)HT fields for granulites (Brown, 2007a). Blue area: *P–T* conditions where amphibolite and granulite facies overlap. Clockwise and anticlockwise *P–T* paths are indicated for HP (red arrow) and (U)HT granulites (black arrow). Note that HP granulites may reach the UHT granulite field (e.g., O'Brien and Rötzler, 2003). HP and (U)HT granulites follow an approximately parallel retrograde *P–T* path. Grey area: *P–T* range for CO_2 isochores with a density between 1.1 and 1.2 g/cm^3 (calculated using the equation of state by Bakker, 1999).

3.2. High-pressure and high-temperature granulites

Granulites are ideal rocks for performing *P–T* estimates: they comprise mineral phases with well-known thermodynamic properties (e.g., feldspars, garnet, and pyroxenes) including well established Fe–Mg distribution coefficients for numerous mineral pairs at granulite facies temperatures (e.g., Spear, 1993). As a result, peak metamorphic conditions of granulites are probably the best known and most accurate of all metamorphic rocks (Spear, 1993). The granulite facies *P–T* field of interest (0.5–1.3 GPa, 700°–>1000 °C, respectively), can be divided into three sub-facies (e.g., Brown, 2007a) (Fig. 6), namely: (1) HP granulites, transitional to eclogites, (2) HT granulites, grading to (3) ultrahigh-temperature (UHT) granulites when the temperature exceeds 900° (e.g., Kelsey, 2008).

Of particular importance is the fact that *P–T* paths are strikingly different for HP and (U)HT series: clockwise and anticlockwise, respectively (Harley, 1989; Bohlen, 1991; Spear, 1993) (Fig. 6). It must be noted that the concept of clockwise and anticlockwise *P–T* paths is only valid near the peak metamorphic conditions. It does not take into account the complexity, depending on the geodynamic setting, that may occur at the beginning of the *P–T* trajectory. The subdivision of granulites into HP granulites with a clockwise *P–T* path and (U)HT granulites with an anticlockwise *P–T* path, eliminates the need for an intermediate granulite type (~7 kbar, ~700–800 °C, respectively), once regarded as typical for lower crustal granulites (e.g., Bohlen, 1987).

Most HP granulites have experienced large, in most cases poorly known, pressure variations. As a consequence, peak metamorphic conditions are commonly defined by the maximum reached temperature. Quite often, their clockwise *P–T* paths show a systematic temperature increase during decompression, allowing some to reach the UHT field (e.g., Limpopo Complex, Belyanin et al., 2010; Tsunogae and Van Reenen, 2011; Van Reenen et al., 2011; or Neoproterozoic Southern Granulite terrane in Southern India, Santosh et al., 2010b). This seems however to be relatively rare: for many HP granulites with eclogitic affinities (e.g., GLA-group, French Massif Central, Fig. 1), the final HT episode is either very weak, or absent. Conversely, anticlockwise *P–T* paths, characterised by a simultaneous increase of both *P* and *T* near peak conditions, reach frequently the UHT field caused by magmatic underplating (Ellis, 1987; Bohlen and Mezger, 1989), allowing metamorphic temperatures to reach the magmatic range.

Post-peak *P–T* paths are quite similar for both HP and (U)HT granulites (Fig. 6), characterised by decompressional cooling at temperatures between ~800° and ~400 °C. This temperature of ~400 °C is roughly the temperature at the base of stable continent crust in a lithospheric plate (i.e., 50–70 km depth, see fig. 2.19 in Stüwe, 2007), marking the end of the metamorphic episode. In this respect, it is interesting to note that this decompressional cooling segment is approximately parallel to high-density CO_2 isochores (Fig. 4). The internal pressure in CO_2 inclusions with a density ≥1 g/cm^3 remains approximately equal to external rock pressure along this part of the *P–T* path (Figs. 4, 6). This is the fundamental reason why a large amount of CO_2 inclusions are well preserved in some granulites whereas brines, with much steeper isochores in *P–T* space, will be constantly transposed and re-equilibrated resulting in collapsed inclusions, which are also found in many granulites (Touret, 2001).

4. Formation of a continent: from compression to extension

The present-day processes at ocean–continent plate boundaries illustrate how continents may grow through lateral accretion of volcanic arc complexes (e.g., Isozaki et al., 2010). HP metamorphism is widespread in the subducted slab, although HT rocks may also be found locally at the base of the continent, under the influence of the fluids released during subduction. This duality of HP/HT series has led Miyashiro (1961) to propose the concept of paired metamorphic belts, using the example of the Sambagawa/Abukuma orogenic belts in Japan (see also Brown, 2010). Later studies have shown that, like in the Variscan orogenic belt, a significant time interval of up to 100 Ma can exist between both series (e.g., O'Brien and Carswell, 1993; Ledru et al., 2001).

The clockwise *P–T* path of HP granulites requires crustal thickening, a condition well realised at the onset of subduction. On the other hand, the anticlockwise *P–T* path of HT and especially UHT granulites are best achieved by crustal extension, during which heat supplied from the mantle, together with a progressive increase of the pressure, explains the anticlockwise loop of the *P–T* path. The additional heat can be supplied by mantle-derived intrusions emplaced during metamorphism (Ellis, 1987; Bohlen and Mezger, 1989), as is observed in many HT granulite terranes. For instance, in Southern Norway, syn-metamorphic (amphibolites and granulite facies) gabbro intrusions (referred to as "hyperites" in the regional literature, Bugge, 1943) cover 20 to 30% of the exposed surface area. A number of arguments, i.e. the mantle $\delta^{13}C$ signature of CO_2 trapped in fluid inclusions (Hoefs and Touret, 1974) and the large abundance of CO_2 fluid inclusions in these rocks, imply that the gabbro intrusions are the main source for CO_2. The source for brine fluids is less obvious in Southern Norway; the relation between brine fluids and meta-evaporites (Touret, 1979; Engvik et al., 2011) implies that the brines are inherited sedimentary pore fluids in which syn-metamorphic intrusives have mainly stimulated the circulation of these fluids. However, the fact that brine remnants are also found in mantle rocks and minerals, including kimberlites (Kamenetsky et al., 2004) and diamonds (Izraeli et al., 2001) indicates that the mantle is also a potential source for brines (Newton and Manning, 2002, 2010).

Fig. 7 illustrates the simplified model which we consider to provide the best explanation for the duality of HP/HT paired belts. Subduction starts with an HP metamorphic regime (Fig. 7a), i.e. rocks in the descending lithospheric slab are progressively transformed into HP granulites and/or eclogites. Slab detachment (e.g., Davies and Von Blanckenburg, 1995) and the assimilation of the detached fragment into the mantle changes the stress regime from compression to an extension (e.g., Zeck, 1996) and an increased heat flow (e.g., Ledru et al., 2001; Santosh et al., in press) caused by the upwelling of the astenosphere, resulting in (U)HT metamorphism (Fig. 7b). It is likely that the progressive assimilation of the detached fragments in the mantle is the cause of mantle metasomatism as they are the prime source of fluids in the mantle environment (e.g., Touret, 2009). Finally, lower crustal CO_2 ascends to higher crustal levels and the

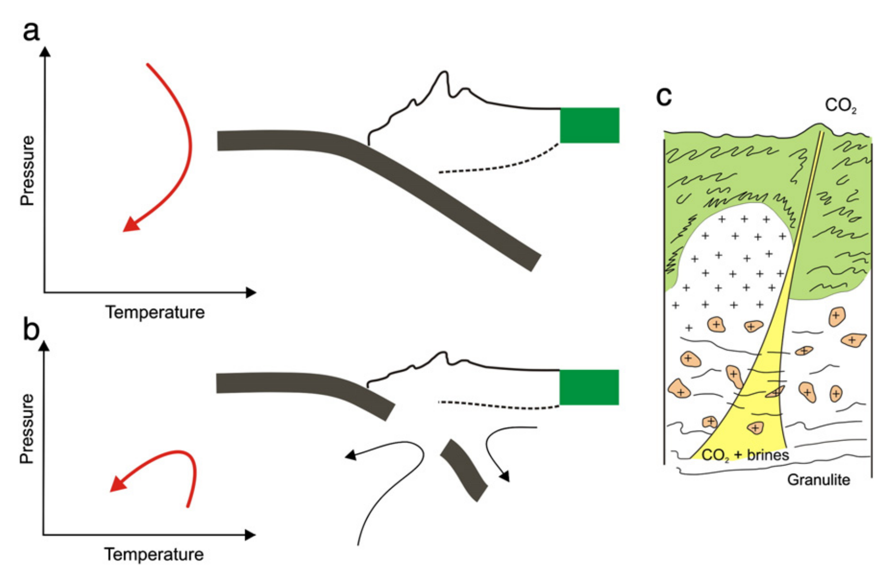

Fig. 7. Formation of HP and (U)HT granulites in a subduction–accretion–collision setting. (a) Formation of HP granulites during subduction, characterised by a clockwise *P–T* path. (b) (U)HT granulites, characterised by an anticlockwise *P–T* path, are formed at the base of the crust as a result of slab detachment and upwelling of the astenosphere (e.g., Santosh et al., in press). (c) Release of CO_2 from the lower crust to higher crustal levels and the atmosphere through megashear zones.

atmosphere along megashear zones (Fig. 7c) during continental breakup.

Despite its simple character, the present model explains an important fact, namely that fluid inclusions in HT and especially in UHT granulites are far more abundant compared to HP granulites. This is in particular illustrated by the occurrence of abundant, well preserved CO_2 inclusions in (U)HT granulite terranes such as in Southern India and Sri-Lanka (Santosh and Tsunogae, 2003). In addition, these rocks show the best examples of retrograde fluid-related metasomatism along large-scale shear zones, in particular carbonation (Newton and Manning, 2002) as has been discussed in Section **2**. On the other hand, shear zones along which HP granulites have been exhumed show little evidence of fluid-related retrograde metasomatism. Some of the HP rocks (e.g., Kullerud et al., 2001) do show evidence (i.e., presence of chloride enriched biotite and amphibole) of the formation of a high-salinity fluid from an initially low-salinity fluid caused by hydration reactions, referred to as desiccation mechanism (Markl and Bucher, 1998). It is clear that brines may be formed by different mechanisms (e.g., Yardley and Graham, 2002) that are difficult to identify because of the lack of appropriate chemical tracers. Considering their importance at the continent/mantle interface, deciphering the brine origin is an obvious challenge for future research.

In conclusion, we will accept in this paper the idea that large quantities of mantle-derived fluids were emplaced in the continental crust during extension, after subduction-related crustal thickening. Magma accumulation at the base of the crust, also responsible for the (U)HT metamorphic regime, leads to vertical crustal growth (e.g., Fountain, 1989) that complements the more lateral crustal growth by subduction-related magmatism.

5. Paired belts through time: granulites and supercontinents

Having discussed in some detail the different types of granulite metamorphism, in particular from the fluid point of view, we now investigate how the concept of paired belts (HP and (U)HT regimes) evolved throughout the Earth's history. It has been known for a long time that granulites, once thought to be exclusively Precambrian, could be of any age (Fig. 8) (Brown, 2007a). The oldest Archean crustal remnants known on Earth (Isua, Greenland, 3.8 Ga) are metamorphosed at the amphibolite/granulite transition zone (e.g., Rollinson, 2002). HT metamorphism at Isua does not show evidence of crustal thickening. The peak pressure did not exceed 0.5 GPa and pre-metamorphic magmatic textures (e.g., pillow lava) are completely preserved (Appel et al., 1998). The interaction between crust and mantle during this metamorphism is marked by the widespread occurrence of ultrabasic rocks (Nutman, 1986). These mantle rocks, now occurring as serpentinites, are partly transformed into carbonates, either occurring as veins or as patches (Nutman, 1986). The carbonate occurrences are so abundant that they were initially mistaken for sedimentary marbles (Nutman, 1986). But it was later found (Rose et al., 1996) that these carbonates had a $\delta^{13}C$ mantle signature and were formed near peak metamorphic conditions at 500–600 °C, i.e. they are comparable to the carbonate or quartz–carbonate zones found in (U)HT granulite terranes as discussed in Section 2.

Granulites are common in many other Archean terranes (Bleeker, 2003), but unfortunately these occurrences are geologically complicated (largely due to structural–metamorphic overprinting) making a sound geological interpretation difficult. Most Archean granulites are igneous (charnockite–enderbite suites) in character. *P–T* conditions (e.g., Jequié Complex, Bahia, Brazil, Xavier et al., 1989) correspond distinctly to HT granulite metamorphism, with typical pressures of 0.5–0.7 GPa at temperatures around 800 °C. Even when *P–T* estimates are higher (e.g., Scourie, North Scotland, 875–975 °C at 0.85–1.15 Gpa, Johnson and White, 2011), there is still no evidence that the rocks have experienced higher pressures during the prograde path (Johnson and White, 2011). According to Johnson and White (2011), crustal growth was dominated by magmatic accretion. This situation can be generalised for the whole Archean period, with the additional comment that the abundance and characteristics of these HT granulites require the presence of a carbonated mantle.

The first evidence of present-day subduction is marked by the occurrence of Proterozoic eclogites (Brown, 2006), e.g., Lofoten (Markl and Bucher, 1997) and Madagascar (Nicollet, 1989; Möller et al., 1995). They indicate the beginning of a plate tectonic system, which closely resembles that of modern time in a probably hotter environment (e.g., Brown, 2009). It had been known for a long time that some orogens, e.g., the Grenville Province in Canada and Southern Scandinavia, did contain a great abundance of granulite occurrences, all of approximately the same age: e.g., ~1 Ga for Grenville, ~1.7 Ga for Svecofennian rocks in Southern Sweden and Finland. Granulite ages did not seem to be evenly distributed in time and any regularity in their age distribution was not apparent. However, at the University of Maryland, Mike Brown made a great step forward by compiling *P–T*-age data of 140 metamorphic belts,

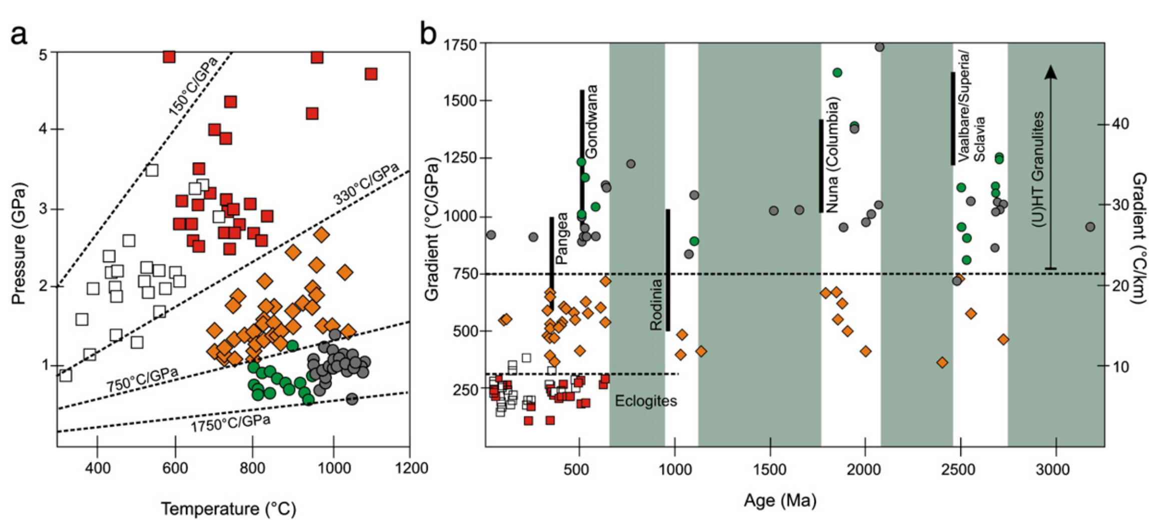

Fig. 8. (a) *P–T* values for different metamorphic rocks in relation to the geotherm (figure slightly modified after Brown, 2007b). Green circles: common granulite belts; grey circles: UHT granulites; orange diamonds: medium pressure eclogites and high-pressure granulites; open squares: low-temperature, high-pressure rocks; red squares: ultrahigh-pressure rocks. (b) Variation of the thermal gradient as a function of the age of peak metamorphism. Periods of supercraton amalgamation (Vaalbara, Superia and Sclavia) and supercontinent amalgamation (Nuna, Columbia) are indicated (after Brown, 2007b).

retaining only cases not affected by polymetamorphism (Brown, 2007a). His results are shown in Fig. 8, illustrating that (U)HT granulites are relatively more abundant compared to HP types during the Proterozoic, whereas for late-Precambrian and post Cambrian times the opposite is true (Fig. 8b). The compilation also shows that periods during which granulite metamorphism is active, lasting between 200 and 500 Ma, are separated by longer periods of stability (Fig. 8b). Each active period ends with the formation of a supercontinent, which is dislocated during the subsequent period of stability.

6. Proterozoic supercontinents: the role of (U)HT granulites in the final stages of supercontinent amalgamation

Fig. 8b shows at least four supercontinents that have been identified between Cambrian and the beginning of Proterozoic, three of them at the end of an "active" metamorphic period: Rodinia (Grenville) at ~1 Ga, Nuna (Columbia) at ~1.7 Ga (Svecofennian in Scandinavia) and the mysterious Vaalbara/Superia/Sclavia (a number of other names can be found in the literature) at ~2.5 Ga. The case of Gondwana is less clear, occurring in a period in which, with the apparition of true eclogites, marks the transition between hotter subduction to present-day colder subduction regimes. It may also be noted that Gondwana is only half of a global supercontinent (the other half being Laurasia), and that it grades almost continuously to the last supercontinent (Pangea), still in a state of continuous evolution. In M. Brown's compilation (Brown, 2007a), UHT granulites are relatively evenly distributed within the "active" periods, relatively more abundant however close to the limit boundaries, either at the beginning or near the end of the amalgamation. This might be due to the fact that metamorphic rocks in the middle of each period might be subjected to polymetamorphism, hence not considered in the compilation. It must also be realised that such a global picture might conceal details of an obviously complicated process. We will concentrate in this paper on Rodinia and Gondwana in order to illustrate the role of UHT granulites during progressive amalgamation of continental fragments and, notably, the fact that they tend to be more developed during the final stage of amalgamation. For Rodinia, details can be quite complicated, such as in Southern Norway (Bingen et al., 2008), the place where CO_2 inclusions in granulites have been observed for the first time. The Sveconorwegian orogenic belt resulted from collision between Fennoscandia and another major plate, possibly Amazonia, at the end of Mesoproterozoic. The first metamorphic event occurred at ~1.1 Ga (Arendal phase), involving closure of an oceanic basin, accretion of a volcanic arc and HT granulite metamorphism at 1140–1125 Ma. Thrusting of the already metamorphosed southern part of the belt (Bamble) onto the Northern Terrane (Telemark) led to underthrusting in the western segment (Iddefjord) and resulted in HP metamorphism at 1050 Ma, followed by uplift. A second phase of high-grade metamorphism followed, with alternating HT and local HP metamorphic episodes, culminating (930 Ma) in the western part of the belt (Rogaland) by the simultaneous emplacement of a major anorthosite–mangerite–charnockite complex and low pressure UHT granulite metamorphism ($P=\sim 0.4$ GPa, $T=>1000$ °C, Jansen et al., 1985), illustrated by the occurrence of osumilite, sapphirine, and quartz (Hermans et al., 1976). This metamorphic event marks the end of the metamorphic evolution. Simultaneously, the eastern part of the belt (Herefoss granite), at a distance of less than 100 km, was already exhumed to a depth of no more than 5 km (Andersen, 1997).

Possibly because of its younger age, the example of Gondwana is even more instructive. Here, amalgamation started at ~750 Ma ago and culminated at ~500 Ma, resulting in an intricate network of elongated granulite belts, best seen on the African continent (Fig. 9). These belts contain some of the most classical granulite occurrences ever described, notably in Southern India, Madagascar and Sri-Lanka. The African and South Indian granulites contain a great number of UHT occurrences with most of them having an age of ~500 Ma, except for Proterozoic fragments embedded in Pan African rocks such as Ouzzal (Hoggar, Algeria). Southern India shows a similar picture as the African continent; Pan-African granulites (Kerala, Sri-Lanka) grade into older

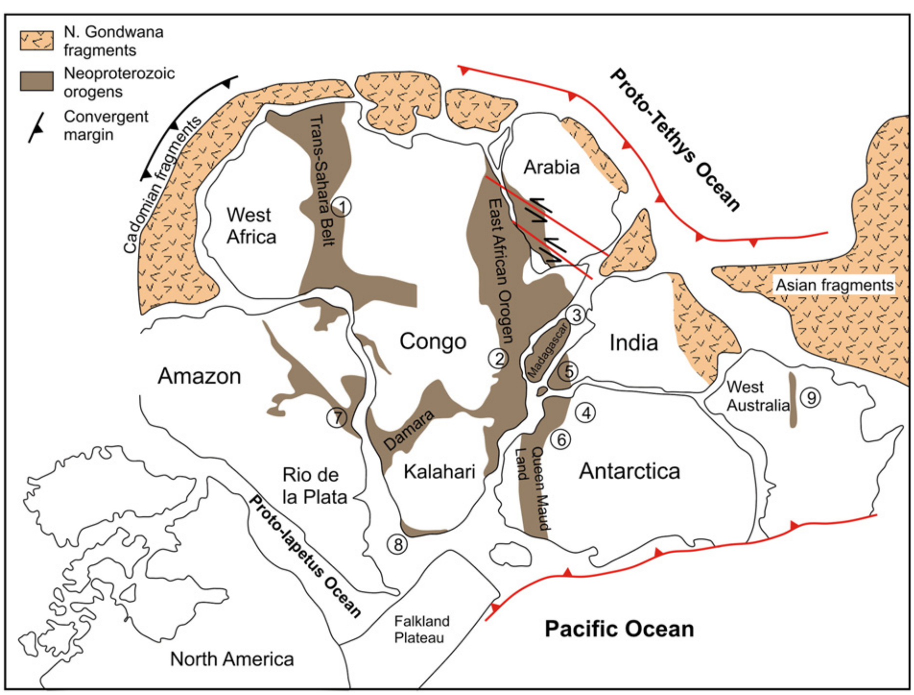

Fig. 9. Pan-African UHT granulite terranes using data supplied by Kelsey (2008) (Gondwana reconstruction after Kröner and Stern, 2004). 1: In-Ouzzal, Hogar, Algeria (e.g., Kienast and Ouzegane, 1987); 2: Furua, Tanzania (Coolen et al., 1982); 3: Madagascar (Paquette et al., 2004); 4: Highland Complex, Sri Lanka (Osanai et al., 2006); 5: Southern India (Tsunogae and Santosh, 2006); 6: Napier Complex, Antarctica (Ellis, 1980); 7: Bahia region, Brazil (Ackermand et al., 1987); 8: Namaqualand, South Africa (Waters, 1986); 9: Warumpi Province, Australia (Scrimgeour et al., 2005).

(Neoarchean and Paleoproterozoic) granulites without any visible boundary in the field.

Fluid inclusions in Pan-African (Gondwana) and Proterozoic (Rodinia) granulites are remarkably similar, both in composition and in density. Moreover, these rocks, in particular Pan-African granulites, contain excellent examples of well preserved primary CO_2 inclusions (Santosh and Yoshida, 1992; Santosh and Tsunogae, 2003; Santosh et al., 2005). This preservation is most likely due to the overlap of the retrograde *P–T* paths and the isochores related to high-density CO_2 inclusions with little fluid-induced re-equilibration. This may also explain the preservation of Proterozoic fragments within Pan-African high-grade metamorphic terranes and the remarkable abundance of gem-quality minerals in Pan-African granulites (Dissanayake and Chandrajith, 1999), including Itrongay orthoclase (Madagascar), Tanzanite (Kenya), sapphire, rubies, and many other gemstones in Madagascar, Sri-Lanka, and Southern India.

In conclusion, we consider it noteworthy that distinct episodes of UHT granulite metamorphism coincides with the final amalgamation of supercontinents (e.g., Santosh et al., 2009a, b), whilst preceding almost immediately their subsequent dislocation. This important observation, for the first time noticed for Gondwana (e.g., Tsunogae and Santosh, 2010), can be extended to all post-Archean supercontinents. It is likely that the fact that amalgamation immediately leads to dislocation might not be fortuitous: fluids, in particular CO_2, stored in the lower crust contribute to exceeding the rupture limit of the lower crust during extension (Miller et al., 2004).

7. What happened at the end of Rodinia and Gondwana?

The fate of the two major Neoproterozoic supercontinents, Rodinia and Gondwana, respectively, provides further compelling evidence that deep fluids, in particular CO_2, could have reached the atmosphere (e.g., Melezhik et al., 2005), with important climatic consequences (Santosh and Omori, 2008b). During the time interval between breaking-up of Rodinia and Gondwana amalgamation, several episodes of glaciations have occurred, almost covering the entire planet (Snowball Earth, e.g., Hoffman, 1999). The most important glaciations, i.e. Sturtian (~700 Ma) and Marinoan (~635 Ma) (e.g., Goddéris et al., 2007) are marked by widespread glacial deposits covered by 3–30 m thick carbonate rocks in continuous layers (cap carbonates). Petrographic features of these cap carbonates, most of them dolomite-rich (dolostone) are different from standard carbonate deposits by their massive character, lack of macroscopic fossils, thick sea floor cements and, above all, a light $\delta^{13}C$ signature ($\delta^{13}C<0‰$) (e.g., Kaufman et al., 1997). Numerous explanations have been proposed to explain these remarkable occurrences. The cap carbonate appears to be comparable to the hydrothermal dolomite breccias in gabbroic intrusions, found in Southern Norway and described by Dahlgren et al. (1993) (Fig. 5a), or in carbonate–amphibole veins, occurring in a similar environment (Fig. 5b). All have a similar mineralogical and petrographic character, comparable $\delta^{13}C$ signature, and possible evidence of microbial activity. The comparison needs further detailed investigations, but the resemblances are significant enough to lead us to suggest that cap carbonates are similar to the retrograde features identified in (U)HT granulite terranes as described here. Following the line of evidence discussed in this paper, we propose that deep crustal granulite fluids expelled along megashear zones (Fig. 7c) are responsible for the formation of these carbonates and associated minerals (quartz and/or amphibole in the shear zones), and also for the increase in atmospheric CO_2 concentration, leading to a sudden end of the snowball glaciations. Climatic effects and carbonation phenomena are less obvious at the end of Gondwana/Pangea although relatively large glaciation episodes are known during the Paleozoic (e.g., Ordovician). It can, however, be observed that a significant increase of CO_2 concentrations in the atmosphere occurred during the stable period before the disruption of Gondawana (atmospheric CO_2 up to 18 times greater than at present values) and, to a lesser extent, Pangea (atmospheric CO_2 five times greater than present values) (Berner and Kothavala, 2001). Furthermore, the $\delta^{13}C$ is inversely related to the concentration at the end of the Gondwana period, suggesting a mantle CO_2 source (Ghosh et al., 2001). It is also during these periods that major biological changes occurred at the Earth's surface, i.e. an increase in both diversity and quantity. We, therefore, consider that the release of large volumes of lower crustal fluids into the hydro- and atmosphere during the final stages of supercontinents assembly played a significant role in a long evolution, leading to the present-day world.

8. Conclusion

In conclusion, it seems now well established, in line with earlier suggestions (e.g., Santosh and Omori, 2008a,b) that orogenic episodes, leading to (U)HT metamorphism, have been instrumental for the amalgamation of supercontinents, at least since the beginning of Proterozoic. These episodes have led to the accumulation in the lower crust of large quantities of low H_2O-activity fluids, particularly high-density pure CO_2 and high-salinity brines. The supercontinents did not last for long; amalgamation immediately preceded rifting and dislocation, in such a way that the coincidence is probably not fortuitous. Fluids accumulated in the lower crust may have induced a global weakness of the continental plate. The extensional framework of supercontinent dislocation allows these fluids to leave the lower crust. Reactive brines were consumed by metasomatic mineral reactions whereas CO_2 reached the atmosphere, with drastic climatic consequences. The scale of this phenomenon raises the question of the ultimate CO_2 origin. Mantle metasomatism induced by subducted slabs, as advocated here, is indeed a possibility, especially for UHT granulite metamorphism following the HP regime. But subducted slabs, especially oceanic, are dominantly hydrous and the CO_2 involved in pre-amalgamation UHT metamorphism may well require another source. In this respect, challenging views have been proposed recently by the Japanese group (Santosh et al., 2010a), namely plumes originating at the lower mantle–core boundary, fed by truly juvenile fluids that were dissolved in the Earth's core at the time of its differentiation 4.5 Ga ago. This would have the important consequence that CO_2 is not only constantly recycled, but also periodically added from an external source. Isotopic tracers, notably rare gases such as He give some support to this hypothesis. But, to our knowledge, clear evidence is not yet available to make this challenging view an established reality. This is an obvious challenge for future research!

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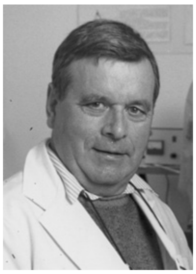

Jacques L Touret was educated as an engineer (ingénieur-géologue) at Nancy (France) and started a university career under the guidance of professor Tom Barth (Oslo). Being convinced (from field evidence) on the significant role of fluids in high-grade rocks, he developed, together with Bernard Poty and Alain Weisbrod (Nancy) the theory and practice of fluid inclusion studies in rocks. He was appointed in 1980 as a full professor at the Free University Amsterdam in the Netherlands, where he maintained this line of research until his retirement in 2001. At present, he is in Paris as a guest scientist at the Musée de Minéralogie, (MINES-ParisTech). He is member of the Royal Dutch Academy of Science and Letters, Academia Europe and the Royal Norwegian Academy of Sciences (foreign member). He received an honorary doctorate at the Université de Liège (Belgium).

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