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Research paper

From granulite fluids to quartz-carbonate megashear zones: The gold rush

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A B S T R A C T

At peak granulite-facies metamorphic conditions, lower continental crust is arguably fluxed by large amounts of two key low water activity fluids: (i) high-density CO_2 and/or (ii) concentrated saline solutions. These fluids are either internally-derived, generated by mineral reactions or dehydration melting or, notably for CO_2, externally-derived, issued from the underlying mantle. Postmetamorphic evolution results in complete disappearance of these fluids, except for minute remnants preserved in minerals as fluid inclusions. Two major processes are involved: (i) at peak conditions, granitoid magmas form, migrate upward, and crystallize as shallow intrusions in the upper crust (mineralized porphyry types or reduced intrusions); (ii) during the rapid decompression which almost systematically follows a period of post-peak isobaric cooling, especially for ultrahigh-temperature granulites (anticlockwise *P*-*T* paths), quartz-carbonate megashear zones are formed by repeated periods of seismic activity. Seismic activity may continue until all free fluids have disappeared, resulting in the ultramylonites and pseudotachylites that are found in many granulite domes. A great majority of vein-type Au deposits worldwide occur in the above-mentioned settings or nearby. We suggest that the Au has been scavenged by the granulite fluids, then redistributed and concentrated during the formation of veins and related phenomena.

1. Introduction

After years of discussion, it has been gradually accepted that low water activity fluids could be present in the lower continental crust during peak granulite-facies metamorphic conditions, in sufficiently large enough amounts to account for the various dehydration reactions occurring at this metamorphic grade (e.g., Newton, 1986; Harlov, 2012). When granulites are uplifted towards the Earth's surface, these fluids leave the rock systems, except for minute remnants preserved in rock-forming minerals as fluid inclusions. Although very high temperatures can be reached during granulite-facies metamorphism, about 1000 °C or higher in the case of ultrahigh-temperature (or UHT) granulites, minerals such as zircon and coesite are not completely re-equilibrated (e.g., Möller et al., 2003; Ruiz-Cruz and de Galdeano, 2012). This permits an accurate reconstruction of the pressure-temperature-time (*P*-*T*-*t*) trajectory of the rocks en route to the Earth's surface. It is known that *P*-*T* estimates are particularly complicated when minerals such as mica or amphibole are involved (Spear, 1993), as is the case for most metamorphic rocks that have not experienced dehydration. In this respect, the scarcity of hydroxyl-bearing mineral phases, as well as the extreme range of *P* and *T* conditions, make granulites almost ideal rocks for mineral thermobarometry, as evidenced by the continuous flow of publications (e.g., Harley, 1989; Kelsey, 2008). These works have confirmed the hypothesis, first proposed by Beloussov (1960), that granulites constitute the lower continental crust and are equilibrated when granitoid magma intrudes at upper crustal levels (i.e., granulite-granite connexion: Clemens, 1990). The average Au content of granulites worldwide is low, ca. 0.2–1.5 ppb (e.g., Sighinolfi and Santos, 1976; Cameron, 1993), but many vein-type Au deposits are located in or in close proximity to granulite complexes. Examples include: (i) Griffin's Find, Yilgarn,

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Western Australia (<0.1 Moz Au; Phillips and Powell, 2009; Tomkins and Grundy, 2009); (ii) Challenger, South Australia (2 Moz Au; Tomkins and Mavrogenes, 2002; Phillips and Powell, 2009); and (iii) Renco, southern Zimbabwe (1 Moz Au; Kisters et al., 1998; Phillips and Powell, 2009). The idea that Au mineralization occurs in granulite terranes is not new, it enjoyed even some popularity in the early 90's (e.g., Kyser and Kerrich, 1990; Barnicoat et al., 1991). But it was then not widely accepted, mainly because of the supposedly "dry" (fluid-absent) character of granulite-facies metamorphism (p. 206 in Kyser and Kerrich, 1990). The situation is now different, and the potential role of fluids during and after granulite-facies metamorphism much better understood (e.g., Harlov, 2012). The object of the present paper is to discuss in some detail the way by which granulite-facies metamorphism in general, and granulite fluids in particular, have contributed to the formation of these deposits.

2. High-pressure granulites versus high-temperature granulites

Granulites are rocks metamorphosed above minimum granite melting temperature, that is, approximately between 700 and 1000 °C. It has been known from simple diagnostic minerals (e.g., aluminium silicate and cordierite) that the pressure was slightly more variable than the temperature: occurring between two extremes, the "classical" intermediate granulites cluster around pressures of about 7 kbar (i.e., depth of about 20 km). High-pressure (HP) granulites, transitional to eclogites cluster around 10–12 kbar. Lastly high-temperature (HT) granulites, sometimes equilibrate at pressures as low as 5 kbar (e.g., Newton et al., 1980). Harley (1989) was the first to show that there is a striking difference in the *P-T* trajectory between HP-granulites and HT-granulites, i.e., clockwise for the former (like eclogites) and anticlockwise for the latter, with a correlative increase in both temperature and pressure at peak metamorphic conditions (Fig. 1). This can only be accounted for by the accumulation of mantle-derived intrusions at the crust-mantle interface. The heat is provided for by the intrusion, and the pressure increase is provided for by a vertical thickening of the crust during the metamorphic episode (Touret, 2009). Such a model is corroborated by the fact that the granulite-facies lower crust becomes increasingly igneous at depths, as shown by the marked opposition between granulite complexes occurring at the Earth's surface (mainly supracrustal origin) and the almost exclusively igneous origin of granulite xenoliths in lavas from recent volcanoes (Bohlen and Mezger, 1989). Many UHT-granulites are low-pressure types "par excellence", but it must also be noted that the UHT field can also be reached by HP-granulites through a significant temperature increase during decompression clockwise *P-T* path, e.g., the Limpopo Belt (Tsunogae and van Reenen, 2011) or Dabie Shan, China (Tong et al., 2011). Both types may occur in a single orogen, conforming to the concept of "paired belts" as proposed by Miyashiro (1961) from the example of the Sanbagawa and Ryoke orogens in northeastern Japan. Both orogens are relatively young in age (Cretaceous) and approximately contemporaneous, but this has later proven to be more the exception than the rule. In the Hercynian (or Variscan) orogen of central Europe, for instance, Eo-Hercynian high pressure metamorphism, occurring notably in the Moldanubikum unit, predates by about 100 Myr the widespread Carboniferous low-pressure Hercynian metamorphism, during which the European crust has acquired its present structure. This includes the granitic migmatite and voluminous granites emplaced in the middle crust (now outcropping at the surface in the basement complexes which defines the whole Variscan chain) and the granulite lower crust, known from xenoliths in recent volcanoes. The French Massif Central is a good regional example (Fig. 2), which could be repeated in virtually all other Hercynian massifs such as Central Spain, Brittany, Vosges, and Bohemia (e.g., Matte, 1991). Known locally as the "Complexe leptyno-amphibolique", the Eo-Hercynian HP metamorphic rocks, dated at about 400 Ma, are thrusted across lower-grade basement in nappes

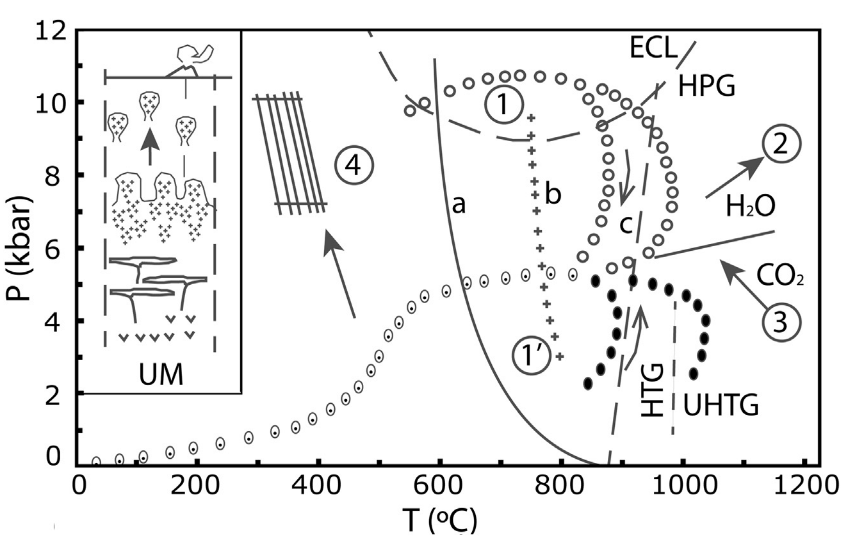

Figure 1. Fluid control during clockwise high-pressure (HP, open circles) and anticlockwise high-temperature (HT, solid circles) *P-T* paths and retrogression (circles with dots) (adapted from Fig. 6, Touret and Huizenga, 2012). HP-paths: ECL (eclogite), grading into high-pressure granulite (HPG). Rocks contain a limited amount of internally-generated fluids, either inherited from the surface or progressively released during dehydration/decarbonation reactions ①. HT-paths: high- or ultrahigh-temperature granulite (HTG/UHTG), also involving internally-generated fluids during the prograde part of the path ①'. For all paths, crustal melting occurs via anatexis, for HP-paths through decompression melting along the curve b, anywhere between a (wet granite melting) and c (dry granite melting), depending on the local H_2O activity. Free water is then dissolved in the melt, transported to upper levels in ascending magmas (crosses in sketch to the right), then released during magma crystallization ②. For HT/UHT paths, the anticlockwise trajectory, corresponding to a simultaneous increase of pressure and temperature, is caused by the stacking of mantle-derived intrusions at the base of the crust (see inset for sketch, UM = Upper Mantle). These intrusions introduce into the rock system large quantities of externally-generated, mantle-derived CO_2 ③, possibly also other types of low-H_2O activity fluids (brines). These fluids remain in the lower crust during isobaric cooling at the end of the metamorphic episode, eliminated along megashear zones during a rapid decompression (regional uplift) which occurs typically at lower temperatures ④.

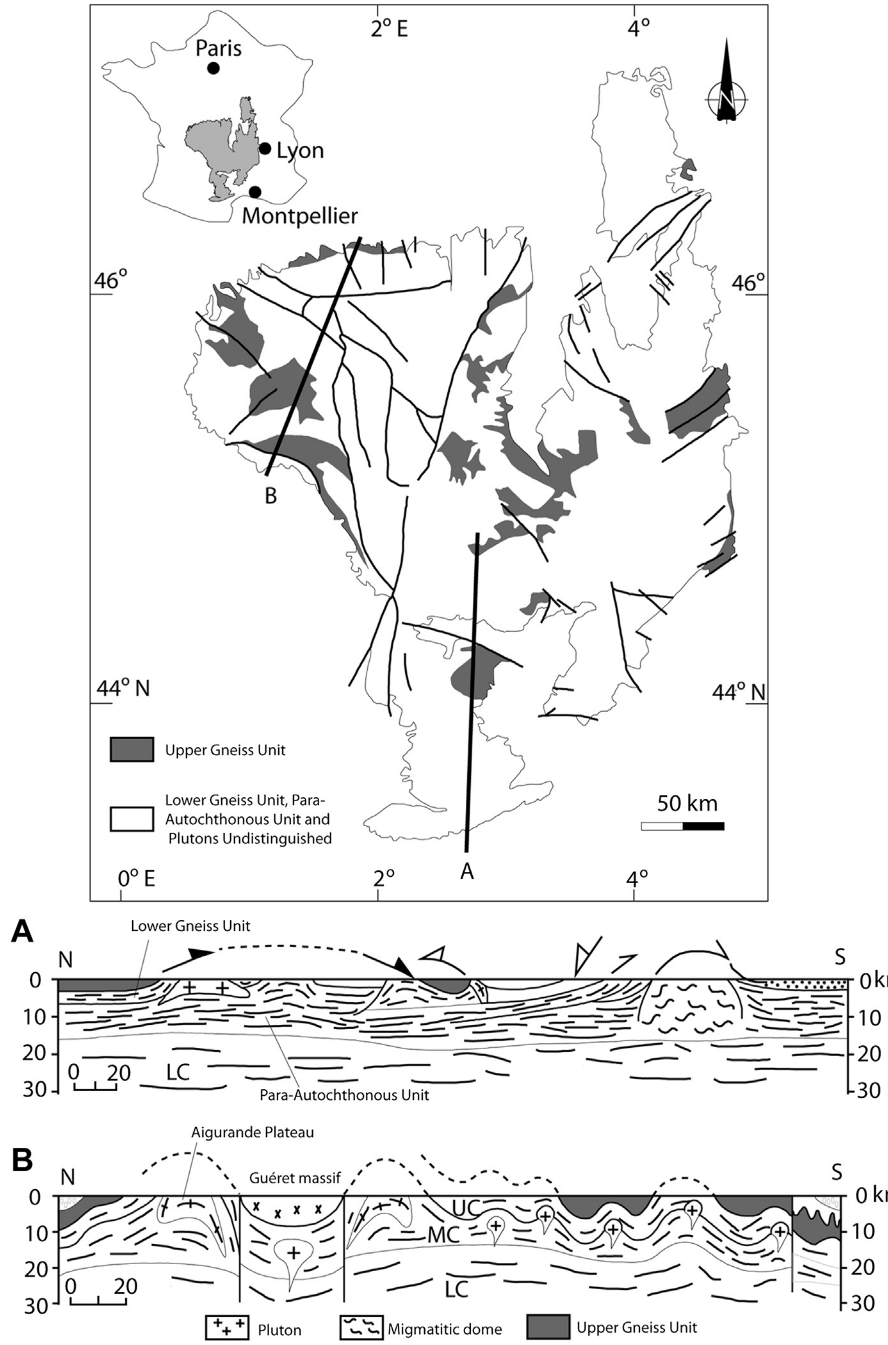

Figure 2. Simplified map and cross sections in the French Massif Central after Faure et al. (2005, 2010). Early HP-granulites were thrust over HT-granite/granulite continental crust, contributing to the formation of the French Massif Central (Variscan orogen). Abbreviations: UC, upper crust; MC, middle crust; LC, lower crust (unknown Proterozoic basement); HP, high-pressure; HT, high-temperature.

extending over 150 km. In this complex, eclogites are associated with HP-granulites, indicating a direct relation with the subduction processes. The tangential large-scale deformation occurred shortly after metamorphism (380–350 Ma: Guérangé-Lozes and Burg, 1990), suggesting that the compressional setting continued for a few 10's of Myr after metamorphism. But, conversely, the space needed in the lower crust for the accumulation of mantle-derived magmas, together with the absence of later thrusting at depth, requires an extensional regime during the later ~300 Ma, HT metamorphic episode. The same juxtaposition of relatively early HP and late HT metamorphic belts is found in orogens of all ages and in many locations, however, with considerable differences in the timing of the HP and HT episodes (e.g., Brown, 2009). This led Touret and Huizenga (2012) to propose a model regarding the formation of the continental crust. That is continents may grow not only laterally above subduction zones, as commonly accepted, but also vertically, through the accumulation of mantle-derived material at the base of the crust. Both episodes of crustal growth are related to, but distinct in time from, the crustal accretion that postdates the lateral growth by an unspecified number of Myr's. On a model of collision between oceanic and continental plates, justified by the occurrence of eclogites in the HP rocks, a reasonable, even if fully hypothetical, explanation could be the detachment of a segment of the descending, oceanic slab, as illustrated in

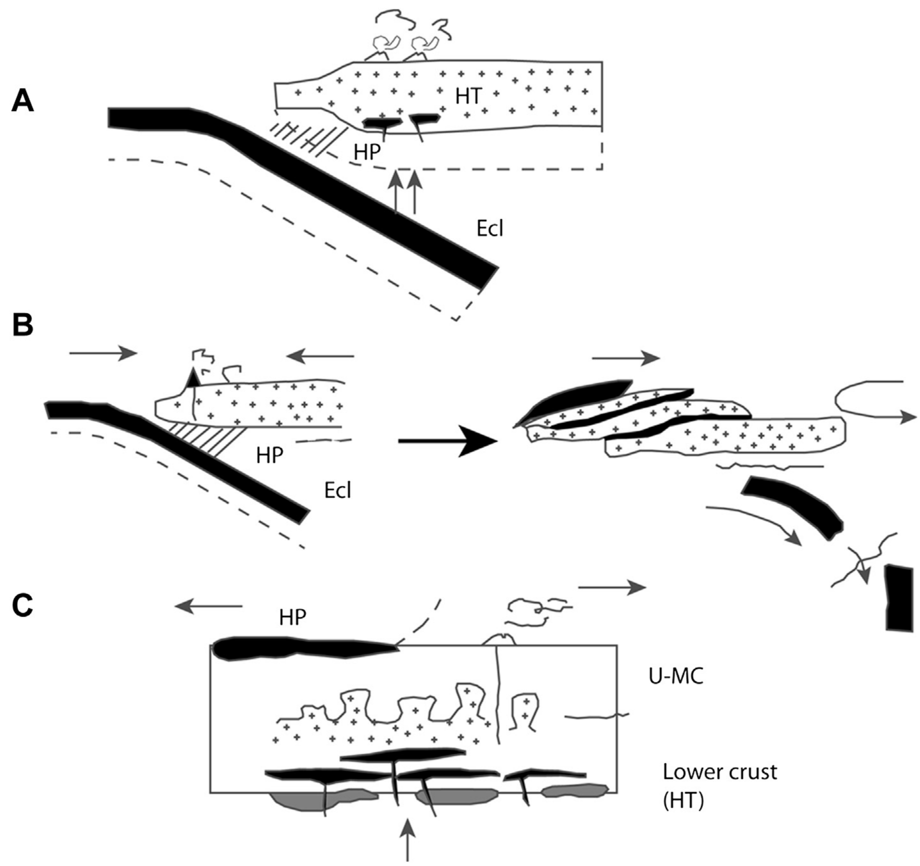

Figure 3. Two competing models for paired HP and HT metamorphic belts, revisited. (A) The first model is after Miyashiro (1961). A single subduction event causes both HP and HT regimes, the last one through the fluids (or melts) issued from the descending slab. (B) and (C) An alternative model, explaining the time difference between both regimes (about 100 Myr in the case of the Variscan orogen). (B) Subduction (ocean/continent) causes lateral accretion of the continent (magmatism), above the HP regime at depth. Further compression induces thrusting of the metamorphosed oceanic crust onto the continent, until a transition compression/extension, possibly initiated by slab detachment in the mantle (right). (C) HT-metamorphism in the extended continent is caused by the massive influx of mantle-derived magmas in the lower crust (vertical accretion by magma stacking at the crust/mantle interface). Abbreviations: U-MC, upper-middle crust; Ecl, eclogite; HP, high-pressure; HT, high-temperature.

Fig. 3. Volatiles liberated during assimilation could induce magmatic activity in the underlying mantle, which is responsible for the HT-metamorphic regime at the base of the continent (Touret and Huizenga, 2012).

3. Fluid regimes in granulites

Decades of fluid inclusion studies in granulites from a wide range of ages and origins provide a wealth of information on the nature and, to some extent, the origin of the free fluid phase present during all stages of rock evolution. In some cases, the distinct relationship between the composition of the fluid inclusions and the nature of the protoliths indicates that some fluids may have been inherited from the surface, and they were able to survive the entire metamorphic process. For HT-granulites, a typical example is highly saline inclusions found in former evaporate-bearing metasediments, for instance, from the Proterozoic Bamble Sector of southern Norway (Touret and Nijland, 2013). Diagnostic minerals such as anhydrite are not unusual in high-grade complexes (Nash, 1972; Butchins and Mason, 1973). Metamorphic saline fluids may be formed by the retrogression of Cl-bearing prograde phases such as scapolite, but it has also been argued that former pore sedimentary fluids could be a major source. These are found, almost systematically, in granulite-facies rocks, notably in the amphibolite-granulite transition zone (e.g., Newton et al., 1998; Touret and Nijland, 2013). Despite the fact that most early brine inclusions have been strongly modified and re-equilibrated after the peak of metamorphism ("collapsed inclusions": e.g., Touret, 2009), the widespread occurrence of saline fluids is further indicated by a number of features constantly found in all granulite domains. First of all, one of the typical mineral textures occurs notably at K-feldspar/plagioclase intergranular boundaries in the form of K-feldspar micro-veins or myrmekites (Touret and Nijland, 2013). These are especially well developed in charnockites and granitoid intrusions generated and emplaced in the granulite-facies environments (Touret and Huizenga, 2012). The other indication of brine occurrence in granulites is the high oxidation state that is sometimes found on a regional scale as documented in the Shevaroy Hills Massif, southern India (Harlov et al., 1997), Lofoten-Vesteraalen, northern Norway (Griffin et al., 1978), Labrador, Canada (Currie and Gittins, 1988), the Bamble Sector, southern Norway (Harlov, 1992, 2000), and the Ivrea-Verbano Zone, northern Italy (Harlov and Förster, 2002a, 2002b). This widespread occurrence shows that the metasedimentary source advocated above is only one possibility, and can be eventually supplemented by fluids migrating upwards along grain boundaries from crystallizing basement ultramafic magmas (Harlov, 2012). In this respect, the recent finding by Giuliani et al. (2013) of oxidized sulphate-rich mantle fluids in kimberlite xenoliths opens a distinct possibility for some granulite-facies brines to derive ultimately from the mantle, a promising line of research that awaits further work.

The other granulite-facies fluid that is systematically found in minerals as fluid inclusions from HT-granulites is high density, pure CO_2 (e.g., Touret, 1971). In the same manner as for the saline fluids, the origin can be either internally-derived, i.e., generated by mineral reactions or fluid-melt interactions in the metamorphic environment, or externally-derived, i.e., issued from the underlying mantle. Peak metamorphic CO_2 fluid inclusions can also be strongly modified during uplift, with new generations of fluid inclusions being created when the pressure difference between the fluid in the inclusion and that in the surrounding host rock exceeds the host mineral strength. This fluid transposition mechanism, either explosion (decrepitation) if the inclusion fluid is over-pressured, or implosion (collapse) in the case of under pressure, is well known in the fluid inclusion literature, notably through the morphological features which help to identify each case (e.g., Touret, 2001). CO_2-rich fluids have much higher wetting angles than aqueous saline fluids (Watson and Brenan, 1987; Gibert et al., 1998), thus they are much less able to coat grain edges and migrate for long distances along intergrain boundaries. CO_2 migration can happen, however, on a limited scale, if the whole mineral is re-equilibrated along its margin. For instance, in coronitic gabbros worldwide that have been emplaced in granulite-facies environments, abundant Fe-Ti oxide needles cloud the magmatic plagioclase, whereas the metamorphic, recrystallized plagioclase is very clear and completely inclusion-free. Presumably, oxides have been expelled from the feldspar lattice during recrystallization, by a mechanism that remains to be explained. Primary CO_2-inclusions occur in the recrystallized margin, indicating that CO_2 can move along the edges of the crystal if the mineral host is wholly recrystallized. But this case remains rather exceptional. In general, CO_2 inclusions occur along successive networks of healed micro-fractures (known in the fluid inclusion literature as "secondary trails"), of decreasing density in the case of rapid decompression, or increasing density if the temperature decreases progressively at relatively constant depth (isobaric cooling *P-T* path: Touret, 2001). A marked exception is the case when the postmetamorphic *P-T* trajectory has roughly followed the fluid isochore (constant density line in the *P-T* space) on a *P-T* interval large enough to reach conditions for which the inclusion may reach the surface without further damage (roughly $P < 2$ kbar, $T < 300$ °C, corresponding to a depth of about 5–6 km) (pseudo-isochoric *P-T* path). In this case, preserved inclusions that formed at peak metamorphic conditions can be unambiguously identified, both from their location (primary inclusions in metamorphic minerals, e.g., garnet, pyroxene or plagioclase) and because the fluid isochore matches the metamorphic *P-T* conditions. Both granulite-facies fluid types, i.e., saline brines and dense CO_2, occur in different domains, always clearly separated, indicating that they have remained immiscible even under the highest *P-T* metamorphic conditions, and that they are probably derived from different sources.

Fluid inclusions found in HP-granulites are not fundamentally different from those occurring in HT-granulites. However, they are smaller, as a rule less abundant, and more heavily transposed. A more striking difference exists when the HP-granulites have been through the eclogite field in the well known high-pressure to ultrahigh-pressure (HP/UHP) complexes at Dabie-Sulu, eastern China. Besides the "common" granulite fluids (high-salinity brines and CO_2), N_2 and CH_4-rich fluids have also been identified, identical to fluids found in eclogites (e.g., Fu et al., 2001, 2003a,b; Xiao et al., 2001; Fig. 4). Homogenization temperatures for CO_2 inclusions in HP/UHP-eclogites, clinopyroxenites, and the granulite-facies equivalents are different, reflecting the temperature difference during the granulite-facies metamorphic event (Fu et al., 2001, 2003a,b; Xiao et al., 2001). A further difference is that, in HP-

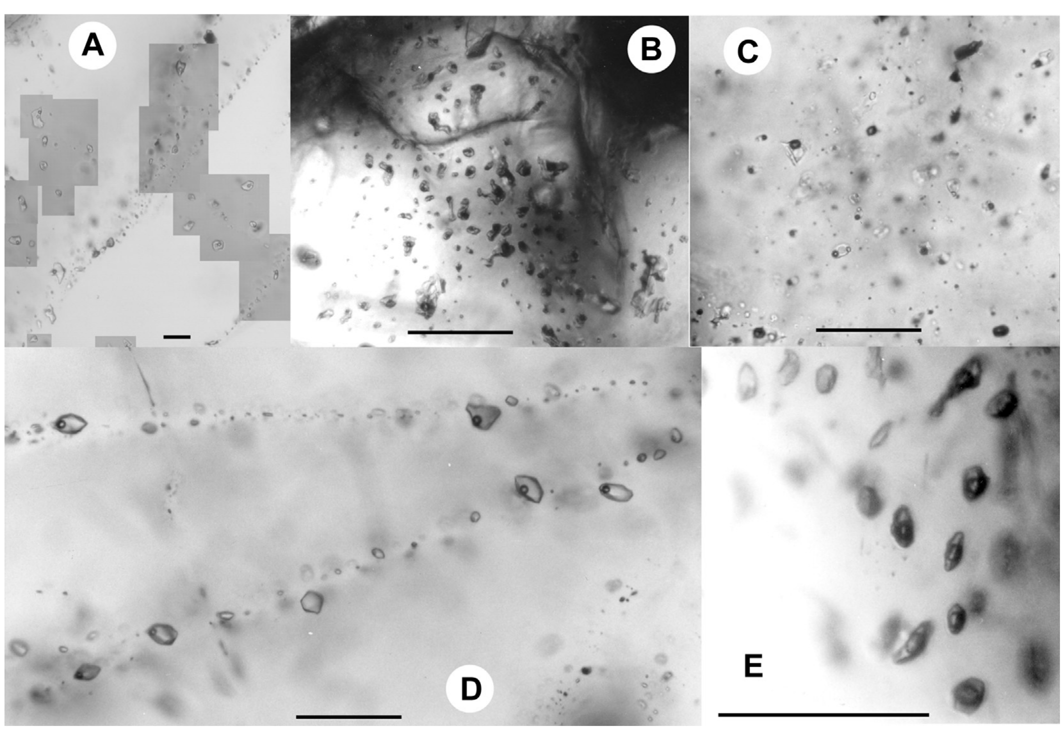

Figure 4. Microphotographs of fluid inclusions. These include halite-bearing Ca-rich fluids (high-salinity brine), Na-dominated aqueous fluids, and low-salinity aqueous fluids, from the Dabie-Sulu Orogenic Belt including the Jiaobei terrane, eastern China. (A) scattered halite-bearing (Ca-rich) brine inclusions and trail-bound low-salinity aqueous fluid inclusions from a quartz vein, Dabie; (B) brine inclusions in garnet porphyroblast from an UHP-eclogite, Dabie; (C) CH_4-bearing, ferropyrosmalite-bearing (multi-solid), hypersaline brine inclusions in quartz from an UHP-eclogite, Sulu; (D) low-salinity aqueous inclusions in quartz from an UHP-eclogite, Sulu; and (E) mixed CO_2 − low-salinity aqueous inclusions in quartz from an UHP-eclogite, Sulu. Scale bars: 50 μm. For examples of fluid inclusions in the Jiaodong Au deposits, see Fan et al. (2003) and Hu et al. (2013).

granulites, brine inclusions are dominant, whereas CO_2 examples are rare or absent. As noted at Dabie-Sulu, CO_2 is very commonly mixed with N_2, a typical gas found in eclogites (liberated from NH_4^+ ions during feldspar/mica breakdown), and/or CH_4. Reduced C-species in high-grade rocks commonly occur by reaction of former organic matter with aqueous fluids released during dehydration melting, suggesting a dominant local origin for this CH_4. The situation changes drastically when, in HP-granulite complexes, the temperature increases markedly during decompression. This happens rather frequently in nature, to such an extent that HP-granulites can reach the UHT field, e.g., the Limpopo Belt, South Africa (Tsunogae and van Reenen, 2011). In this case, CO_2 inclusions are extremely abundant, similar to the more common anticlockwise-path UHT-granulites. In HT-granulites, which constitute the majority of granulites occurring in regionally exposed complexes, notably in the core of Precambrian cratons, the situation is opposite, i.e., brine inclusions are rare or absent. Except in the characteristic above-mentioned lithologies (meta-evaporates), CO_2 inclusions are abundant or very abundant, especially in or near the mantle-derived meta-intrusions that are present in virtually all granulite complexes and now occur as a variety of intermediate to basic meta-igneous lithologies (i.e., enderbites/gabbros or pyroxene granulites; e.g., Fonarev et al., 1998). These features, along with noble gas or carbon isotope data (e.g., Dunai and Touret, 1993), lend support to the idea that most CO_2 derives ultimately from the mantle, brought into the lower crust by (or together with) synmetamorphic intrusions, which have provided the heat responsible for the HT character of the granulite-facies metamorphism.

4. Gold mineralization in high-grade metamorphic complexes

If the lower crust was loaded with fluids at the time of the metamorphic climax, it is also clear that these fluids have left the rock system somewhere on the way towards the Earth's surface (e.g., Phillips and Powell, 2010). In all types of granulites, the preservation of lower crustal assemblages in rocks presently exposed at the Earth's surface requires rapid exhumation when thermal pulses, caused by mantle-derived intrusions, occur roughly at temperatures below 700 °C. This rapid exhumation is necessary, not only to avoid re-equilibration of peak metamorphic mineral assemblages, but also to protect the rocks from the action of the ubiquitous aqueous fluids from the brittle upper crust. A recently discovered new support for rapid exhumation is the presence in granulites of supercooled melt inclusions, with devitrification features resembling those found in volcanic lavas (Hiroi et al., 2014). Even if this result requires confirmation, it was found to be widely present in a series of initial studies (e.g., Limpopo, Greenville, Sri Lanka, Antarctica, and Bohemia). The impressive range of metasomatic transformations observed in the amphibolite/granulite transition zones of the Bamble Sector, southern Norway, including scapolitization, albitization, and feldspar replacement in graphic pegmatites (Touret and Nijland, 2013), occur during this last part of the retrograde path, at temperatures equal to or below about 400 °C. Brine inclusions are systematically found to occur during these transformations. The first observations had already been done at the end of the 19th century (Judd, 1889). It shows that brines remain active until about 400 °C, probably corresponding to the rapid uplift identified on the *P-T* path. Detailed studies have mainly been done in southern Norway (e.g., Bingen et al., 2008). However, similar observations in many other complexes (e.g., Harley, 1989; Kelsey, 2008) suggest that they are representative for HT-granulites in general. The fate of CO_2 is different. At temperature below about 500 °C, the remaining CO_2 fluids are either trapped in inclusions or may react with aqueous saline solutions to form carbonates, which are indeed found in many granulites as minute aggregates, possibly corresponding to former fluid inclusions (Touret, 1985).

Field observations and petrological studies (e.g., Touret, 1985; Touret and Huizenga, 2011) indicate that the fluids leave the granulite-facies environment during repeated retrograde events, most of them finally ending in the formation of quartz veins of different generations and sizes. Many of these veins are metal-bearing, notably Au, and the resulting vein-type Au deposits account for a significant share of the total global Au production (e.g., Weatherley and Henley, 2013). As the temperature of equilibration of the veins is typically within the range of 300–400 °C, these deposits would be classified as being mesozonal in the classification of Goldfarb et al. (2005). The formation of a vein is a complicated phenomenon. In most cases, it is induced by some seismicity, resulting in a strong and sudden pressure decrease at the opening of the veins. This is followed by the progressive filling by a mineral sequence, which itself is determined by the temperature and pressure decrease, and the chemistry of the input fluids. A variety of processes are involved. These include element migration in the host rock (causing alteration), drastic phase separation, and Au solubility in the fluid. Moreover, the formation of a vein is rarely a single event. Many minerals in veins are conspicuously zoned, indicating repeated episodes of opening, with the possible influx of fluids of different compositions. However, we believe that, despite this complexity, it is possible to see some major trends, which enable us to relate the granulite fluids to mesozonal Au deposits. The arguments are as follows:

(a) The fact that many Precambrian high-grade metamorphic complexes worldwide host world class vein-type Au deposits;
(b) Besides veins associated with granitoid intrusions, a number of deposits occur in or near "quartz-carbonate megashear zones", which are associated with many high-grade metamorphic complexes worldwide (Newton and Manning, 2002). There are examples where the mineral assemblages in these shear zones require a distant high-temperature magmatic source (e.g., Beakhouse, 2007), but HT-granulite metamorphism would be within the same temperature range;
(c) Inclusions in Au-bearing quartz veins are extremely complicated, resulting from a sequence of processes. Low-grade metamorphic veins (epithermal to mesothermal) contain mainly low-salinity aqueous fluids, either meteoric waters or fluids resulting from dehydration mineral reactions (Nesbitt, 1988). But the earliest fluids in "deep" veins show distinct analogies with granulite fluids (e.g., Kolb and Meyer, 2002);
(d) The limited number of available data indicates that some granulites are depleted in Au, suggesting that the granulite fluids have scavenged Au at this level (Cameron, 1993).

4.1. *Some examples of vein-type Au deposits in high-grade metamorphic complexes*

On the regional scale, high-grade metamorphic complexes contain a number of Au deposits, many of them of world-class importance. These include the Dharwar craton in southern India, parts of the Yilgarn craton in Western Australia, the Kaapvaal craton in South Africa, and the Jiaobei terrane in the North China Block. The Jiaodong Peninsula, eastern China, which includes both Triassic UHP-eclogites (metamorphosed to granulite-facies) in the Sulu terrane and notably Archaean high-grade metamorphic complexes in the adjacent Jiaobei terrane, which is described here in more detail (Fig. 5).

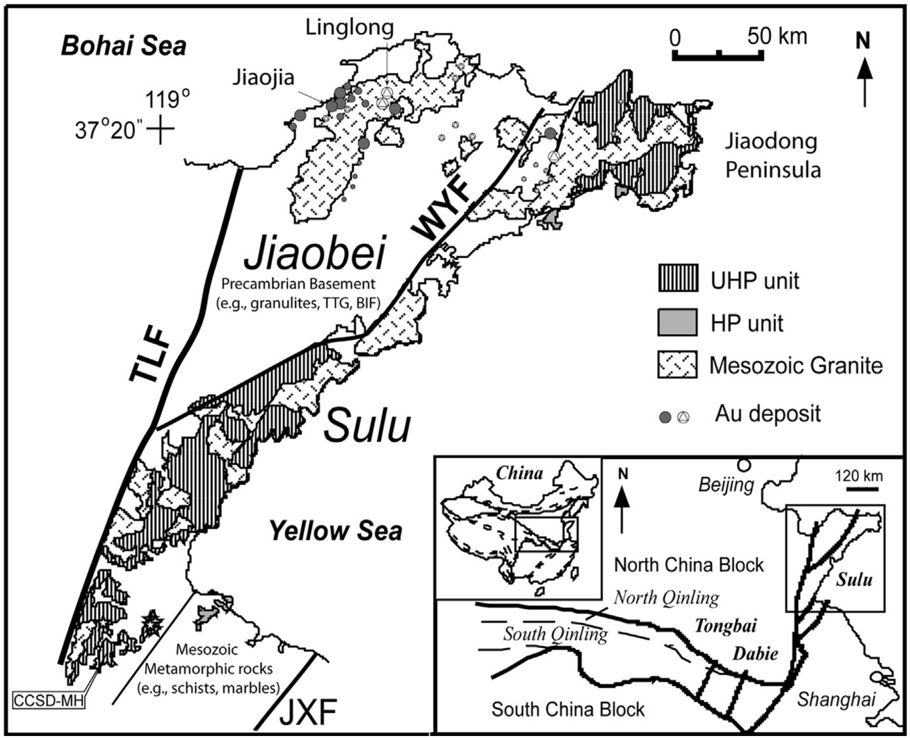

Figure 5. Simplified geological map of the Shandong Peninsula, eastern China. Locations of Au deposits, Linglong-type (triangle in circle) and Jiaojia-type (solid circle), were after Fan et al. (2003). Differently sized symbols of Au deposits mean different Au resources: big symbol means > 50 t Au, small symbol means < 50 t Au. CCSD-MH, the Chinese Continental Scientific Drilling main hole. Abbreviations: TLF, Tan-Lu Fault; JXF, Jiashan-Xiangshui Fault; WYF, Wulian–Yantai Fault; TTG, tonalite-trondhjemite-granodiorite; BIF, banded iron formation.

The Sulu terrane, together with the Dabie terrane, marks the eastern part of a Triassic suture zone between the North China and South China blocks. The Sulu terrane is a low to high-grade metamorphic complex composed of an HP unit in the south, a UHP unit in the centre, and a high-temperature unit in the north (e.g., Zhang et al., 2009 and references therein). The UHP unit is made of abundant gneisses, marble, jadeite/kyanite quartzite, minor eclogite lenses, and garnet-bearing ultramafic complexes. Inclusions of coesite or quartz pseudomorphs after coesite in garnet, omphacite, or metamorphic zircons may occur all over the unit in both the eclogites and the gneisses, indicating that the eclogites as well as their host rocks, notably garnet clinopyroxenites, went through regional UHP metamorphism. UHP-eclogites commonly contain abundant coesite and hydrous phases (e.g., talc, zoisite/epidote, nyböite, and phengite). Similar rock types occur in the HP unit, and are metamorphosed at epidote-amphibolite-facies and blueschist-facies conditions. These include quartz-mica schist, chloritoid-kyanite-mica-quartz schist, marble, rare blueschist, and minor eclogite. HP/UHP eclogites and garnet clinopyroxenite have been interpreted, either as metamorphosed sedimentary or volcanic rocks, which occur as interlayers or pods associated with supracrustal rocks, including quartzofeldspathic gneiss/schist, marble, and garnet-bearing jadeite or kyanite quartzite or metamorphosed mafic or ultramafic intrusions. Metamorphic *P-T* conditions vary from ca. 700 °C and >27 kbar for the UHP rocks to ca. 600 °C and 20 kbar for the Dabie HP-eclogites, and ca. 800 °C and >27 kbar for the Sulu UHP rocks (e.g., Zhang et al., 2009 and references therein). Some garnet peridotites may record much higher-P than the surrounding coesite-bearing eclogites, reaching 40 kbar or above. Both ultramafic and eclogitic rocks experienced a complex metamorphic history, which include at least three distinct stages of recrystallization: (i) primary formation, (ii) peak UHP metamorphism (630 to 890 °C and >27 kbar), (iii) pressure decrease with corresponding strong temperature increase, resulting in ultra-high temperature (UHT) granulite-facies overprinting (Tong et al., 2011), and (iv) final amphibolite- to even greenschist-facies overprinting. However, the metamorphic evolution may have occurred as a continuum of recrystallization and fluid-rock interaction during the Triassic plate subduction, continental collision, and exhumation. The whole metamorphic evolution from peak UHP metamorphism, through UHT-granulite-facies metamorphism to amphibolite-facies retrogression occurred at 245–200 Ma (Zheng et al., 2003). After which, voluminous, post-orogenic (Cretaceous) granites intruded in the region.

The Jiaobei terrane is located between the Tan-Lu Fault and the Wulian–Yantai Fault (Fig. 5). Part of it actually occurs on the southeastern margin of the Archaean North China Block (3.8–2.5 Ga) that was significantly reworked during both Proterozoic and Phanerozoic orogenies (e.g., Zhang et al., 2003). The Wulian–Yantai Fault is generally regarded as the boundary between the North China and South China blocks. The Precambrian basement in the Jiaobei terrane consists of Archaean tonalite-trondhjemite-granodiorite (TTG) and supracrustal rocks including banded iron formations (BIF), and Palaeoproterozoic amphibolite- and granulite-facies rocks.

Giant Au deposits such as Linglong (4.1 Moz Au; Zhou and Lü, 2000) and Jiaojia (3.7 Moz Au; Zhou and Lü, 2000) occur at different places in Shandong Peninsula (Fig. 5). Most of the large Au deposits in this region are hosted in Mesozoic (i.e., middle Jurassic to mid-Cretaceous or Yanshanian in Chinese literature: e.g., Wang et al., 1998; Zhang et al., 2003) granitoids that have intruded into the eclogites (undergoing granulite-facies metamorphism) and associated gneissic rocks of Mesozoic orogen and the amphibolite- to granulite-facies rocks of the Precambrian basement. Although the massive occurrence of Mesozoic intrusions in the Dabie-Sulu orogen and adjacent areas in eastern China suggest that the associated Au ores of the eastern Sulu region may have originally been part of a single, large Cretaceous circum-Pacific gold province due to the overlap in time (Goldfarb et al., 2007; Goldfarb and Santosh, 2014), it is likely that the intrusion-hosted Au deposits in the eastern Sulu region formed during exhumation of the basement subsequent to the collision between the North China and South China blocks (e.g., Zhou and Lü, 2000; Fan et al., 2003, 2007).

Gold occurs in quartz veins, which are widespread within metamorphic rocks in the entire Dabie-Sulu Orogenic Belt. An important feature, which makes these veins rather different from those occurring in the megashear zones (see below), is the scarcity or absence of carbonates. The time of formation of the quartz veins in eclogites and gneisses is not easy to assess precisely. In eclogites, vein size ranges in width from sub millimetres to several decimetres. They are almost identical in HP/UHP rocks (Li et al., 2001; Zheng et al., 2003), and many features indicate that they postdate significantly the peak of metamorphism, e.g.,

(a) Both the paragenesis and quartz content of eclogite at the vein margins are identical to eclogite far away from the veins;
(b) There is a distinct boundary between each vein and the host rock;
(c) No UHP index mineral has been identified in the vein minerals;
(d) Vein minerals are free of deformation.

The giant Linglong-type (lode) deposits at Linglong, Denggezhuang (1.7 Moz Au; Zhou and Lü, 2000) and Jinqingding (>0.5 Moz Au; Zhou and Lü, 2000) occur in faults cutting Mesozoic granitoid intrusions. Quartz veins occur as lenticular discrete bodies bounded by the fault gauge. Wallrock alteration includes silicification, sericitization, sulphidation, and potassic alteration (Fan et al., 2003, 2007). Other Au deposits at Sanshandao and Yinggezhuang (i.e., Jiaojia-type) are characterized by fault-hosted disseminated and stockwork-style mineralization (Fan et al., 2003; Hu et al., 2013).

4.2. Quartz-carbonate megashear zones

The Sulu example shows that, during exhumation, massive granitic intrusions were emplaced some 100 Myr after peak metamorphic conditions, namely during the period of rapid decompression following isobaric cooling. This example is not unique. In the Bamble Sector of southern Norway, for instance, peak metamorphism occurred at ca. 1 Ga, which is the same approximate time as the Grenville orogeny. The rocks were then at a depth of about 20–30 km. Post-orogenic granites (Grimstad, Herefoss) were emplaced at ca. 960 Ma, the rocks being then at a depth of less than 3 to 5 km. It is reasonable to believe that rapid decompression has promoted crustal melting at depth. The granitoid magmas drained the granulite fluids at depth and rose in the crust to end up as shallow intrusives (e.g., Petford et al., 2000). A subset of these intrusions took the form of reduced intrusion-related Au systems (RIRGS) (e.g., Hart, 2007) or mineralized porphyry types. There is, however, another way to rapidly uplift lower-crustal high-grade complexes towards the surface. In contrast to low-grade metamorphic terranes, several medium- and high-grade areas display pervasive and vein-controlled carbonate metasomatism of country rock along regional scale shear zones, which Newton and Manning (2002) call “carbonated megashear zones”. Examples include the post-Hercynian South Tien Shan Fault Zone (Baratov et al., 1984); the late Proterozoic Attur Valley, Tamil Nadu, India (Wickham et al., 1994); the late Archaean Chitradurga area, Karnataka, southern India (Chadwick et al., 1989); the mid-Proterozoic Mary Kathleen Fold Belt, Queensland, Australia (Oliver et al., 1993); the mid-Proterozoic Bamble Shear Belt, southern Norway (Dahlgren et al., 1993); and rock unit from Madagascar (Pili et al., 1999). Exposed country rocks in some of these metasomatic zones have been replaced by up to 20 vol.% carbonate over tracts of 100 km by 10 km. The host rocks are metamorphosed to various grades, from upper amphibolite- to granulite-facies. Minor intrusions of alkali-gabbros, syenites, or carbonatites occur in a number of these terranes, e.g., the Attur Valley, Tamil Nadu, India or South Tien Shan, China (Newton and Manning, 2002), which indicate that these megashear zones may be rooted in the mantle, going through the entire lower crust. Carbonate replacement occurred at temperatures between 500 and 700 °C (Newton and Manning, 2002), namely at the onset of decompression following a period of isobaric cooling. In the eastern Mt Isa Block, NW Queensland, Australia (Fig. 6), the carbon isotopic composition of carbonate from these shear zones (calcite and ankerite) is quite uniform with a typical mantle value, i.e., $\delta^{13}C_{PDB}$ is equal to about −7‰ (Marshall et al., 2006 and references therein). Fluid-inclusion analysis of the Queensland occurrence, coupled with the Na-Ca metasomatism of associated carbonate-poor lower-grade rocks, indicate that highly saline fluids were involved during metasomatism (de Jong and Williams, 1995). Hydrothermal carbonates (dolomite, ankerite), closely resembling those found in megashear zones, notably through the mantle-like carbon isotopic signature, have also been found in the Bamble Sector (Dahlgren et al., 1993).

4.3. Fluids in Au-bearing quartz veins

Few types of ore deposits have been more intensively studied than Au-bearing quartz veins. Virtually all types of fluids found in fluid inclusions (e.g., Roedder, 1984) occur there. These include low to high-salinity brines, S compounds, gases like CO_2, CH_4, N_2, and so on. In Archaean greenstone belts (e.g., Barberton, South Africa; Yilgarn Block, Western Australia; Abititi Province, Canada) Au

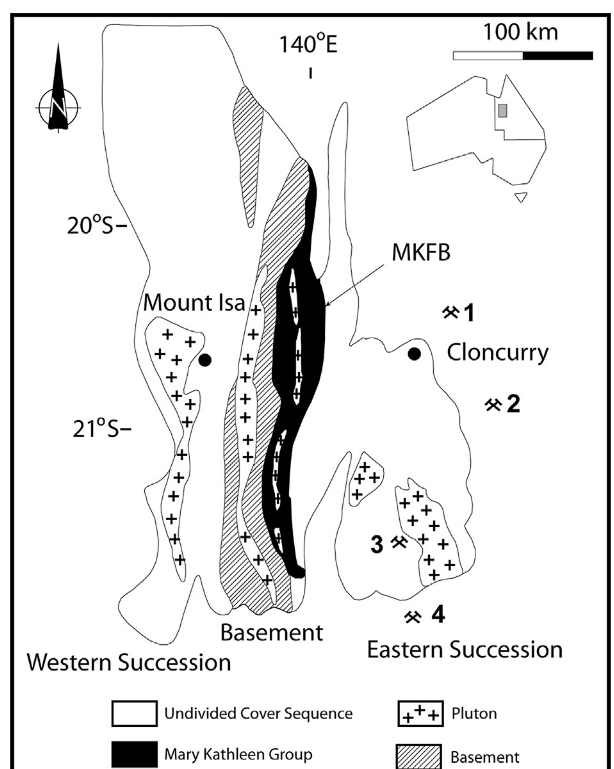

Figure 6. Simplified geological map of the Mt Isa Block, NW Queensland (after Oliver et al., 1993). MKFB, Mary Kathleen Fold Belt. Major Cu-Au deposits include: (1) Ernest Henry Cu-Au; (2) Eloise Cu-Au; (3) Starra Cu-Au; (4) Osborne Cu-Au.

mineralization occurs at about 1–2 kbar and between 250–450 °C, with or after carbonate formation in the vein. Metamorphic S may be released by the breakdown of pyrite and chlorite under amphibolite-facies conditions (Tomkins, 2010). Fluids in inclusions are uniformly low-salinity aqueous, with gas (dominantly CO_2) systematically present but at values not exceeding a few mol.% (e.g., Huizenga, 1995). Loucks and Mavrogenes (1999) contended that this fluid is the phase remaining after Au deposition, but it has little to do with the fluid responsible for Au transportation. Gold dissolves in water by the reaction $Au(c) + H^+(aq) = Au^+(aq) + 0.5H_2(g)$ (Loucks and Mavrogenes, 1999). Here, c = crystalline, aq = aqueous, g = gaseous. In pure water, the concentration of $Au^+(aq)$ is extremely low. The situation changes drastically in the presence of a ligand, ion, or molecule that binds the central metal atom to form a ligand. In the presence of a complexing ligand L, the reaction becomes $Au(c) + H^+(aq) + nL^{z-} = AuLn^{1-nz}(aq) + 0.5H_2(g)$. Sulphur is present in most magmas, notably those generated in the lower crust and upper mantle. Gold solubility in aqueous fluids released from these magmas is controlled by a reaction like $Au(c) + 4H_2S(g) = AuHS(HS)_3^0\,(aq) + 0.5H_2(g)$, with S as a ligand. Note that this reaction, by liberating H_2, will hence have a reducing effect, indicated in sulfide-bearing veins by the common occurrence of graphite (instead of carbonate) and reduced C-species (CH_4) in the gaseous phase of the fluid. Other very effective ligands are CO_2 (carbonyl complexes) or Cl, which explains the drastic increase in metal solubility in highly saline brines (e.g., Newton and Manning, 2010). The role of these two ligands becomes obvious in veins formed at a deeper level than those occurring in epithermal deposits. Carbonates are rare or absent, but fluids are much more CO_2-rich, to the point that its aqueous concentration has been proposed as a prospecting guide for veins occurring in or near high-grade complexes, for instance, in French Guyana, South America (Machairas, 1970). A typical example of inclusions found in these veins, occurring in Rwanda, is shown in Fig. 7. Inclusions show strong signs of transposition through decrepitation, corresponding to the strong pressure decrease at the time of the opening of the vein. The CO_2 content in the inclusion ranges from about 50% to more than 90% by volume. It closely resembles typical CO_2-rich granulite fluids, mixed with a variable amount of water that could conceivably have been issued from dehydration melting at depth.

The other granulite fluid, highly saline brines, is also found in a number of shear zones, however, in very variable amounts. Besides Au, the other metal commonly found in these veins is Cr, as shown by spectacular quartz-fuchsite assemblages. Some are rather close to the metabasites, suggesting rather short distances for Cr transportation. But this is not always the case. The beautiful fuchsite rock that Greenland has chosen as a national symbol ("Greenlandite") (Fig. 8) occurs, for instance, in Isua along km-size shear zones, far away from any outcropping peridotites. The role of ultramafic rocks in regulating volatiles and metal concentration during deep crustal metamorphism is of obvious importance (Rosing and Rose, 1993), but it may occur over long, km-size distances. Many quartz-carbonate veins also contain a small amount of early halite-bearing aqueous inclusions, indicating that immiscible brines were present in the early CO_2-rich fluid. It has been demonstrated that in granulites these brines have a marked oxidizing effect (see above). It can be hypothesized that brines have prevented the reducing effect shown in S-bearing fluids, allowing most of the carbon present in the fluid source to be finally hosted in carbonates. The low-salinity water released at the time of the carbonate formation will dilute most brines still present at this late stage.

4.4. Gold depletion in granulite terranes

The idea that Au can be scavenged in the lower crust by granulite-facies fluids, then redistributed at upper crust levels by melt extraction or transport in shear zones, is further supported by the fact that supposedly immobile elements or minerals can be quite mobile during granulite-facies metamorphism (Touret and Nijland, 2013). The best examples are Zr-bearing minerals such as zircon, strongly affected through dissolution-reprecipitation processes (Harlov and Dunkley, 2010; Giustina et al., 2011). Mobility can be obvious on the scale of the outcrop, leading to the formation of "zirconite" veins, which are a bimineral association of ilmenite and zircon. Other examples are REE, U or Th, redistributed between major or accessory minerals in the amphibolite- to granulite-facies transition zone (e.g., Bingen et al., 1996; Hansen and Harlov, 2007). During the metamorphism of carbonate-bearing rocks, the enrichment can be so significant that it leads to deposits of economic value, being the source of urano-thorianite-rich placers along the southeastern coast of Madagascar (Rakotondrazafy et al., 1996).

A number of arguments then suggest that Au may be added to the long list of supposedly "immobile" elements such as Zr, Ti or Cr which becomes highly mobile under granulite-facies conditions. The granulite-facies rocks, which serve as the Au source, are highly depleted in Au, as noted by Cameron and co-workers both in the

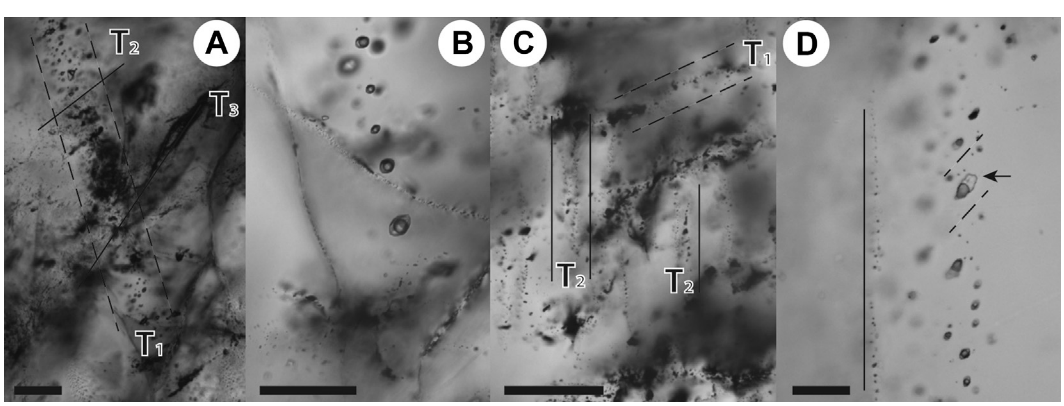

Figure 7. Early inclusions in Au-bearing quartz veins, before massive carbonate deposition, Miyove, Northern Province, Rwanda. (A) Early trail (T_1), transposed along T_2. Later trails (T_3) grade into open fractures. (B) Detail of inclusions along T_2: homogeneous CO_2 (supercritical) in the central bubble, H_2O along the walls of the cavity. Katchgar, Urals. (C) Decrepitation features. Early large inclusions, now empty (dark) are aligned along trail T_1. They are exploded (decrepitated) and now surrounded by a number of smaller inclusions. Transposition occurs through later, suborthogonal T_2 trails (very small aqueous inclusions). (D) Detail of transposed CO_2-rich inclusions (identical to B) and later aqueous trails (dashed line). Carbonates may occur at the intersection (arrow). Scale bars: 100 μm.

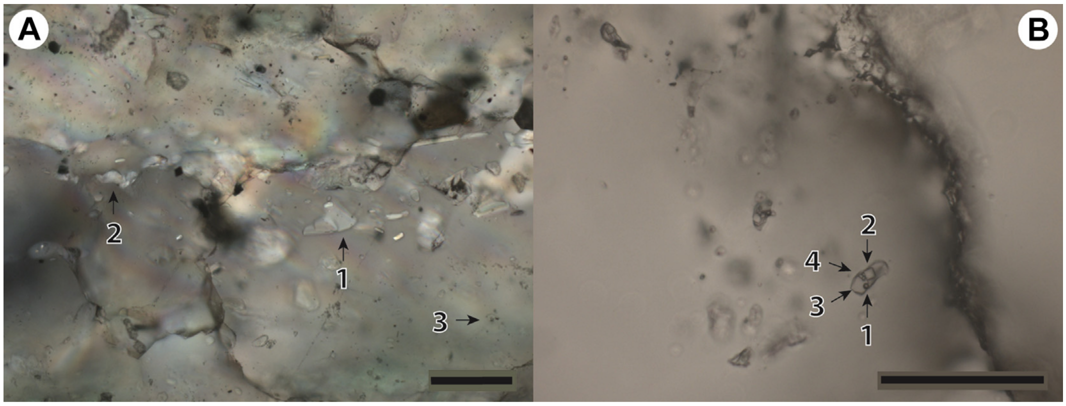

Figure 8. Brine inclusions and late carbonates in an early Archaean quartz-fuchsite shear zone, Isua, Greenland. (A) Polarised light: Many carbonate inclusions (light colours) (1) and (2), often occur also along quartz grain boundaries. Fluid inclusions as small dark points in the quartz (3); (B) detail of multiphase solid brine inclusions: 1 = vapour bubble, 2 = halite cube, 3 = carbonate, and 4 = a solid phase yet to be determined (rutile?). Scale bars: 100 μm.

Bamble Sector (Cameron, 1989) and in the Lewisian Complex, Scotland (Cameron, 1994). In the same region, these authors have identified highly oxidized deep ductile shear zones (Cameron, 1988; Cameron and Hattori, 1994) that we consider as being the roots of the quartz-carbonate megashear zones described above. High oxidation requires the occurrence of brines (Harlov, 2012), actually found in inclusions both in the ductile shear zones and in the host rocks (Touret, 1985). In these cases, it is then logical to believe that brine is the fluid that has scavenged the regional Au from the deep crust, and later transported it to more superficial levels during retrogradation. It is true also that the number of well investigated cases is small, mostly limited to southern Norway, and that it remains to be demonstrated if the model proposed for this region is of general value. Unfortunately, the line of research initiated by Cameron and co-workers has now stopped, as far as present authors know. It would be highly desirable if the southern Norway example could be supplemented by similar studies of other regions.

5. Conclusions

Regional investigations of Au trace element contents in granulite complexes have apparently been discontinued since the work of E.M. Cameron and his group. However, the existing data supports the idea that most Au now occurring in quartz-carbonate veins at the Earth's surface derives ultimately from the lower continental crust, although there are some that argue for the release of crustal Au from Au-concentrating stores such as diagenetic pyrite during the prograde pyrite to pyrrhotite transition (Large et al., 2011). Gold has been scavenged at peak metamorphic conditions by low water activity fluids (CO_2 and highly saline brines) and accumulated in the upper crust during high-grade metamorphism. Mantle-derived fluids (CO_2, and possibly brines; Giuliani et al., 2013) mix with fluids issued from prograde metamorphic reactions or, in the case of brines, inherited from the pre-metamorphic protolith (meta-evaporates). Massive influxes of mantle-derived fluids occur when granulites reach the HT/UHT-granulite field, which in all cases is in a geodynamic setting (compressional or extensional). Both CO_2 and brine fluids remain immiscible even at peak temperature, requiring a very high salinity for the brines. Gold appears to be very mobile in this fluid-rich, oxidizing environment. It is transported and concentrated in the upper crust by two major processes:

(i) Purely magmatic processes. At the onset of retrograde metamorphism, temperatures are still above granite minimum melting temperature, during granitoid melt extraction. These magmas are somewhat enriched in H_2O, Au, and, perhaps F, as a result of dehydration melting. They are able to rise to the upper crust to depths not exceeding 3 to 5 km. Intense hydrothermal activity takes place during magma crystallization. The fluids expelled first closely resemble granulite fluids with the CO_2 being, however, of lower density and somewhat diluted by the water dissolved in the granitic magma (Weisbrod, 1981; Roedder, 1984). In the 1970's, it was thought that magmatic fluids were mixed with large quantities of surface water through convection induced by the heat of the intrusion (Barnes, 1997). More recent studies suggest, however, that with regard to fluids in the melt the magmatic source is the more important (Heinrich et al., 2005), especially for reduced intrusion-related Au systems (RIRGS) (e.g., Baker, 2002; cf. Fu et al., 2014).

(ii) Deep crustal metamorphism. The formation of most granitoid magmas occurs in the lower crust through dehydration melting under granulite conditions, but this concerns only part of the granulite complex. Large segments are preserved, especially in HT-granulite settings, which are identified by mineralogy or pre-metamorphic composition and structure (mainly sedimentary). These are still full of granulite fluids when the granulite complex experiences rapid decompression, approximately at 500 °C at which point the formerly ductile lower crust becomes brittle. The remaining fluids are drained along megashear zones, which explain the rapid, repeated seismic events. Seismicity may continue when all fluids have left the granulite domain, causing the ultramylonites or pseudotachylites found in many exposed granulite-facies terranes (e.g., West Uusimaa, Finland). Provided that the oxidation conditions generated by the brines persist in the veins with decreasing temperature, the final fate of the granulite fluids will be identical in all types of veins. They will either cause alteration in the host rock such as albitization (Touret and Nijland, 2013) or, for CO_2 with bivalent cations, such as Ca, Fe, Mg, and Mn, be stored in carbonates. Mineral formation occurs mainly during pressure decrease, which results in an opening of the veins, resulting in a solubility decrease for the metals, most notably Au, due to the destruction of complexes. After which it will then require other specific conditions, such as a greater quantity of more active fluids, e.g., brines from meta-evaporates, relatively high concentrations of Au in the protolith, or fluid processes and/or fluid-rock interaction during vein formation, to form Au deposits with economic value.

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