УДК 553.068.7

КРУПНОМАСШТАБНЫЕ ПЕРЕНОСЫ ФЛЮИДА МЕЖДУ МАНТИЕЙ И КОРОЙ ПРИ АМАЛЬГАМАЦИИ И РАСПАДЕ СУПЕРКОНТИНЕНТОВ

Ж.Л.Р. Туре¹, Ж.М. Хайзинга^{2,3}

¹ Institut de Minéralogie, Physique des Matériaux, Cosmochimie, Sorbonne Universités, 4 Place Jussieu, F-75005 Paris, France

² Economic Geology Research Institute (EGRU), College of Science and Engineering, James Cook University, Townsville, Oueensland, OLD 4811, Australia

³ Department of Geology, University of Johannesburg, Auckland Park, 2006, Johannesburg, South Africa

Суперконтиненты являются уникальной особенностью планеты Земля. Краткий обзор суперконтинентов, образовавшихся начиная с архея, показывает, что до позднего докембрия амальгамация суперконтинентов, в частности Гондваны и Родинии, происходила через высокотемпературные подвижные пояса, и все они имели проявления ультравысокотемпературных гранулитов. Во время последнего этапа амальгамации нижняя континентальная кора достигала магматической температуры (от ~900 до более 1000 °C) в течение интервала времени, варьирующего от менее 10 млн лет в молодых короткоживущих орогенах до более 150 млн лет в позднедокембрийских (Гондвана) или неопротерозойских (Родиния) долгоживущих орогенах. Во всем мире ультравысокотемпературные гранулиты содержат одни и те же типы флюидных включений, в частности плотные растворы СО, и сильносоленые водные растворы. Количество флюида на пике метаморфизма определяется количеством сохранившихся флюидных включений (в первую очередь СО2) и вторичными эффектами, вызванными вынесенными из нижней коры флюидами, в том числе фельдшпатизацией, альбитизацией или скаполитизацией и образованием либо окисленных (кварц-карбонат), либо восстановленных (графитовые жилы) зон мегасдвига. Хотя некоторые флюиды могут образовываться локально в ходе минеральных реакций или из унаследованных седиментационных вод, углеродные изотопные характеристики и петрографические данные указывают на то, что большинство флюидов как СО2, так и сильносоленые растворы являются производными карбонатитовых расплавов, образовавшихся при частичном плавлении метасоматизированной мантии. Метаморфизм ультравысоких температур имеет определяющее значение для амальгамации суперконтинентов, но флюид, связанный с метаморфическими процессами, вызывает их дестабилизацию и разрушение вскоре после амальгамации.

Суперконтиненты, ультравысокотемпературные гранулиты, флюидные включения, ${\rm CO_2}$, соленые растворы.

LARGE-SCALE FLUID TRANSFER BETWEEN MANTLE AND CRUST DURING SUPERCONTINENT AMALGAMATION AND DISRUPTION

J.L.R. Touret and J.M. Huizenga

Supercontinents are a unique feature of the planet Earth. A brief review of supercontinents formed since the Archean shows that before the Eocambrian, supercontinents, notably Gondwana and Rodinia, amalgamated through high-temperature mobile belts, all of them containing ultrahigh-temperature granulite occurrences. During the final stage of the amalgamation, the lower continental crust was brought to magmatic temperature (from ~900 to more than 1000 °C) during a variable time span, from less than 10 Ma in the recent short-lived orogens to more than 150 Ma in the Eocambrian (Gondwana) or Neoproterozoic (Rodinia) long-lived orogens. Ultrahigh-temperature granulites worldwide contain the same types of fluid inclusions, namely, dense CO₂ and highly saline aqueous brines. The fluid amount in the peak metamorphic conditions is indicated by the amount of preserved fluid inclusions (especially CO₂) and by the secondary effects caused by the fluids when they left the lower crust, including regional feldspathization, albitization or scapolitization, and formation of megashear zones, either oxidized (quartz–carbonate) or reduced (graphite veins). While some fluids may be locally derived either from mineral reactions or from inherited sediment waters, carbon isotope signature and petrographical arguments suggest that most fluids, both CO₂ and high-salinity brines, are derived from carbonatite melts resulting from partial melting of metasomatized mantle. Ultrahigh-temperature metamorphism is critical for supercontinent amalgamation, but the associated fluid causes instability and disruption shortly after amalgamation.

Supercontinents, ultrahigh-temperature granulite, fluid inclusions, CO₂, brines

© Ж.Л.Р. Туре, Ж.М. Хайзинга[™], 2020

[™]e-mail: jan.huizenga@jcu.edu.au DOI: 10.15372/GiG2019128

INTRODUCTION

The Earth is the only planet in the Solar System to have continents, generated by the destruction of subducted oceanic crust since the early days of the Earth's formation. Throughout their history, continents remained mobile on the Earth's surface, periodically assembled in supercontinents, which after some time are fragmented into small pieces, en route towards a new supercontinent. No piece of continental crust has survived the late heavy meteorite bombardment, ca. 4 Ga, but the oldest gneiss found on Earth, generated very shortly after this event (Acasta gneiss, Canada, up to 4.28 Ga (Stern and Bleeker, 1998)) shows that continents already existed at this time. The oldest supercontinent Vaalbara is assumed to be about 3 to 4 Ga old (Cheney, 1996). The most recent supercontinent Pangea (about 0.3 Ga) is now in its final stage of disruption before the formation of a hypothetic new supercontinent in the future. The first overview of supercontinent formation was given by John Rogers in 1996: Ur, formed at ca. 3 Ga and accreted to most of East Antarctica in the middle Proterozoic to form East Gondwana; Arctica, a ca. 2.5–2 Ga continent that contained Archean terranes of the

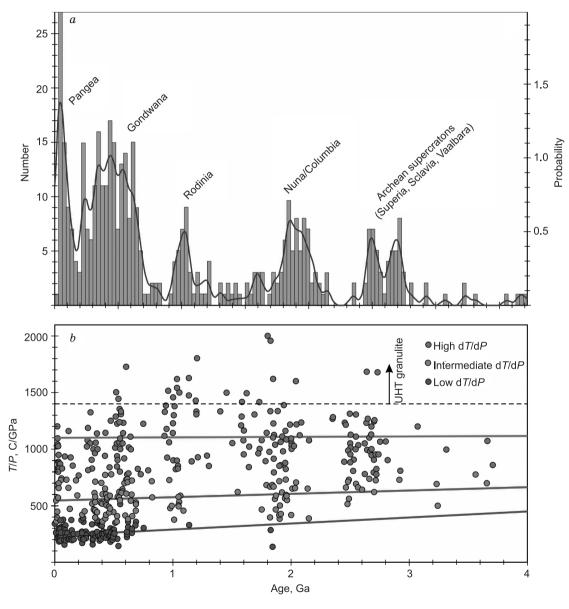


Fig. 1. a, Supercontinents throughout the Earth's history illustrated by metamorphic ages (histograms) recorded in 456 localities modified from (Brown and Johnson, 2018); b, metamorphic temperature gradients (dT/dP) in these localities, subdivided into high, intermediate and low dT/dP groups modified from (Brown and Johnson, 2018).

Colored solid lines: average values (low, intermediate and high dT/dP gradients, respectively); black dashed: separation between HT- and UHT granulites.

Canadian and Siberian shields and Greenland, and Atlantica, formed at ca. 2 Ga of cratons that now occur in west Africa and eastern South America. Arctica grew at ca. 1.5 Ga by accretion of most of East Antarctica and Baltica to form the continent of Nena (or Nuna). Collision of Nena, Ur, and Atlantica, plus minor plates, formed the supercontinent Rodinia at ca. 1 Ga. Rifting of Rodinia between 1 and 0.5 Ga formed three continents: East Gondwana, Atlantica (which became the nucleus for West Gondwana), and Laurasia (which contained North America, Greenland, Baltica, and Siberia). Gondwana formed at ca. 0.5 Ga by amalgamation of its eastern and western parts. Various plates accreted to Laurasia during the Paleozoic, and collision of Gondwana with Laurasia created Pangea at ca. 0.3 Ga. Although other names and assembly models have later been proposed, there is a general consensus on the limited periods of supercontinent occurrences (Fig. 1a). These include the mysterious Eoarchean (ca. >2.5 Ga) supercratons (Superia, Sclavia, Vaalbara), the better defined Nuna/Columbia (ca. 2 Ga), Rodinia (ca. 1.1 Ga), and finally the complicated Phanerozoic period, starting with the two semi-supercontinents (one in each hemisphere) of Laurasia and Gondwana, and ending with Pangea, presently in the process of disruption. Supercontinents older than Proterozoic, from Rodinia onwards, have been completely eroded. showing only their lower crustal roots. Disruption typically occurs by rifting along mobile belts, characterized by complicated relations between amalgamation and disruption. Laurasia and Gondwana started to break off immediately after the amalgamation that resulted in Pangea. Older supercontinents, on the other hand, had longer periods of existence, ca. 0.2 Ga for Rodinia and ca. 0.4 Ga for Nuna/Columbia.

SUPERCONTINENTS SEALED BY ULTRAHIGH TEMPERATURE METAMORPHIC EPISODES

All supercontinents are mainly made of metamorphic rocks, with no significant changes in high to intermediate thermal gradients since the Neoarchean (ca. 2.7 Ga) (Fig. 1b). Low thermal gradients are only abundant in recent orogens, but eclogites as old as 1.87 Ga have been reported to occur in the Kola Peninsula, Russia (Imayama et al., 2018). The regression line of low to intermediate dT/dP gradients shows indeed some lowering after ca 0.8 Ga, but an even greater lowering also occurs with Archean rocks. Hence, a secular change in the thermal state of the crust cannot be the prime cause of the abundance of high-pressure rocks in recent orogens.

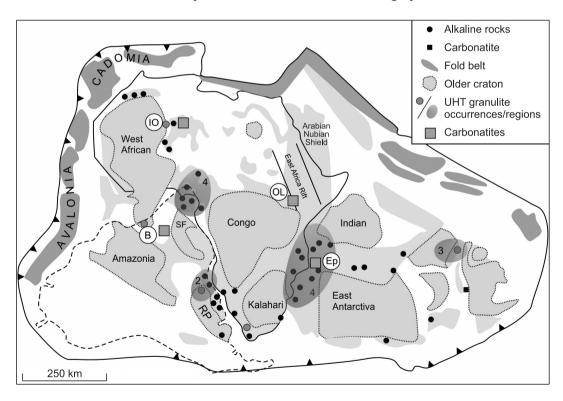


Fig. 2. Reconstruction of the Gondwana supercontinent at ca. 530–500 Ma modified from (Veevers, 2007) (yellow: older cratons, light-grey: Pan African mobile belts) except for Australia (uncertain age) and early Proterozoic spots along the margin of the West African craton (B, Bakhuis, Suriname; IO, In Ouzzal, Sahara, Algeria,), UHT occurrences (after Kelsey and Hand, 2015) occur in Pan-African mobile belts connecting African cratons: Congo, India, East Antarctica (Madagascar, Sri-Lanka and Southern India, Antarctica), Central Africa and South America, Amazonia and West African craton. Ep, Eppawala, Sri-Lanka; OL, Oldyono Lengai, Tanzania).

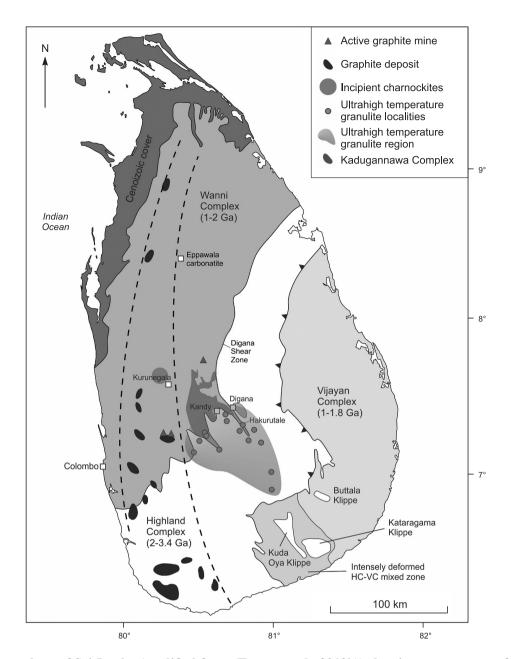


Fig. 3. The geology of Sri-Lanka (modified from (Touret et al., 2019b)) showing occurrences of graphite, key localities for incipient charnockites (Kurunegala), UHT granulites including Hakurutale (Bolder-Schrijver et al., 2000), the Digana Shear Zone (Binu-Lal et al., 2003), and the Eppawala carbonatite (Manthilake et al., 2008).

It is more likely related to the level of erosion: after having been brought to great depth in collisional orogens, high-pressure rocks reached the greatest altitudes during post-collisional extension. Temperature regimes have not changed significantly since the end of Archean for the majority of lower-crustal rocks, which are assumed to be granulites (Rudnick and Fountain, 1995). Normal, high-temperature granulites constitute the greatest part of all cratons and correspond to high dT/dP metamorphic gradients, from ca. 800 °C/GPa, to more than 1200 °C GPa. Extreme gradients, taken somewhat arbitrarily at dT/dP > 1400 °C/GPa (Fig. 1b), characterize ultrahigh temperature (UHT) granulites, in which the metamorphic temperature exceeds 900 to 1000 °C. This temperature range covers the magmatic temperature range, at least for felsic rocks (charnockites). In these rocks, the difference between metamorphic and magmatic is non-existent; a magma that is emplaced at this temperature will show a metamorphic overprint after crystallization (Touret and Huizenga, 2012). The initial discovery of UHT granulites in the Napier Complex, Antarctica, is relatively recent (Ellis, 1980). Currently,

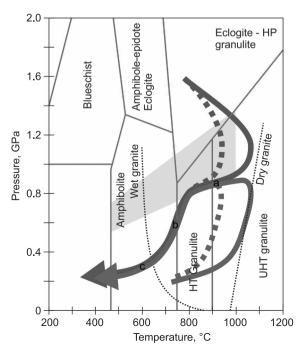
these spectacular rocks have only been found in the three most recent supercontinents (Columbia, Rodinia, and Gondwana), with a majority of documented occurrences in Rodinia and Gondwana (Kelsey and Hand, 2015).

For both Columbia and Rodinia, fragments of the supercontinents are so scattered that no clear relationships can be seen between UHT occurrences and supercontinent amalgamation. However, UHT occurrences in Southern Norway, either near the Rogaland anorthosite complex (Drüppel et al., 2015) or in the Bamble sector (Kihle and Bucher-Nurminen, 1992) are dated at ca. 1 Ga, which is close to the final stage of Rodinia amalgamation. The situation is more apparent for Gondwana. Figure 2 shows that most UHT occurrences in this supercontinent, including the type localities in Central Sri-Lanka and Antarctica, are located in the Pan-African mobile belt which did connect the Archean cratons during the final stage of Gondwana's assembly. They occur near intra-continental boundaries, the best places for finding traces of mantle plumes (Burov et al., 2007). The time of formation of this special case of extreme metamorphism is best established by single crystal dating of accessory minerals, notably zircon and monazite, e.g. SIMS or LA-ICP-MS (Kelsey and Hand, 2015). In Central Gondwana, UHT metamorphism starts in the Eocambrian (Pan-African orogeny, 0.6-0.5 Ga). Exceptions are two isolated spots on the edge of the West African craton, Bakhuis, Suriname (Kroonenberg et al., 2016), or In Ouzzal, Sahara, Algeria, (Bendaoud et al., 2004) (Fig. 2). Both are much older (early Proterozoic), possibly inherited from Columbia (Santosh et al., 2006). Note, however, that for these occurrences the regional geologic context is less known than in Gondwana, either because of political problems (Sahara) or because of limited exposure (Suriname). Despite these limitations, it does not change the main conclusion that UHT occurrences seal the final stage of the supercontinent amalgamation (Santosh et al., 2011).

Figure 3 illustrates in more detail the mode of occurrence of UHT granulites in Central Sri-Lanka. They are found in a rather wide area within the high-grade Highland Complex, flanked on both sides by the lower grade Wanni Complex, to the West, and the Vijayan Complex to the East. The age of the protoliths is early Proterozoic to Archean for the Highland Complex and somewhat younger (1 to 1.8 Ga) for the two lower grade complexes. Like other UHT occurrences elsewhere, they form small spots, a few hundred of meters to a couple of kilometers across, dispersed within a rather well defined zone. As typical mineral assemblages (sapphirine, osumilite) require high Mg-Al lithologies (e.g., Kelsey, 2008) it is highly probable that the actual number of UHT occurrences is higher than reported, but not enough to make continuous, kilometer-size outcrops. The isolated UHT domains typically grade into normal high-temperature granulites, equilibrated at peak temperature of about 850 °C.

P-T-T CONDITIONS OF UHT GRANULITE METAMORPHISM

With their exotic mineralogy and spectacular reaction textures, UHT granulites are among the best rocktypes to perform geothermobarometry. Early temperature determinations (in the 2000's) were based on element distribution (mainly Fe-Mg) in coexisting mineral pairs. More recent studies use complicated pseudo-sections for artificially defined chemical systems such as Fe-Mg-Al-Si (FMAS), later extended to include H₂O (H) and



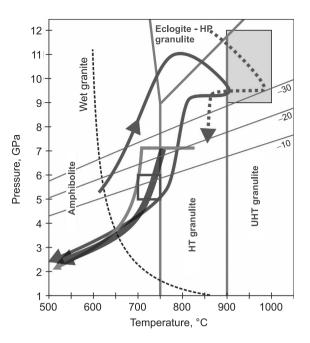
potassium (K), and more recently Fe and Ti oxides (KF-MASHTO) (details discussed in Kelsey and Hand (2015)). Pseudo-sections are calculated using various algorithms (Thermocalc, Perple_X, Theriac Domino), making use of an internally consistent set of thermodynamic data. Results are illustrated by beautiful, highly complicated and spectacular phase diagrams (see e.g. Fig. 6 in Kelsey and Hand (2015)). Pseudo-sections seem to be a must for any modern paper in metamorphic petrology, but we contend that it is not easy to interpret them in terms of deriving actual *P-T* paths for at least two reasons: (1) the artificial system will never reproduce completely the real composition; (2) the fluid phase is assumed to be either absent, or grossly sim-

Fig. 4. Model P-T paths (clockwise and anticlockwise) for HT- and UHP/UHT granulites (dashed line: HT granulite, solid line: UHP/UHT granulite).

In all cases, the post-metamorphic evolution is the same: initial isobaric cooling (a) followed by rapid decompression (b), and finally slow uplift towards the surface (c). Metamorphic facies are after (Brown, 2007).

Fig. 5. Pressure-temperature paths constructed for (1) the Highland Complex (red arrow) (Dharmapria et al., 2017); (2) UHT granulites from the Highland Complex (red dashed arrow, Dharmapria et al., 2015); (3) granulites from the Wanni Complex (orange arrow, Raase and Schenk, 1994; Touret et al., 2019b). The yellow box indicates peak P-T conditions for the UHT granulites in Highland Complex determined by Osanai et al. (2016). The purple box indicates the P-T conditions of charnockitization (5–6 kbar, 700–750 °C, Perchuk et al., 2000) at Kurunegala. The dark grey arrow indicates the P-T conditions of graphite vein formation (Touret et al., 2019b). Metamorphic facies are after Brown (2007).

plified to be pure H₂O whereas fluid inclusion data (or simple common sense) indicate the presence of concentrated NaCl-bearing aqueous solutions (see below). Our view is that much work is still needed before that pseudo-



section sections can fully represent the reality. Pseudo-sections are, however, helpful to reduce earlier estimates of peak temperatures by as much as 150 °C in the case of In Ouzzal (Doukkari et al., 2018). However, it is our opinion that, in general the general shape of UHT granulite P-T path is best approached by conventional thermo-barometry. The prograde path remains a challenge (Fig. 4): either anticlockwise, characterized by a simultaneous increase in pressure and temperature to reach peak conditions, or clockwise when UHT granulites are issued from high-pressure, subduction related rocks. In all cases, peak metamorphism corresponds to a sharp temperature increase (100 to 200 °C) at constant temperature (Fig. 5), suggesting an additional heat supply at depth. The retrograde metamorphic evolution is marked by an initial episode of isobaric cooling followed by decompression. According to S. Harley (2008): "Many UHT terranes have evolved from peak P-T conditions of 8–11 kbar and 900–1030 °C to lower pressure conditions of 8 to 6 kbar whilst still at temperature in the range of 950 to 800 °C". The rapid decompression is marked by partial destabilization of some minerals, notably garnet, to give typical symplectic decompression assemblage of orthopyroxene and plagioclase. This stage lasts until a temperature of ca. 600-500 °C, for a pressure drop of 2 to 3 kbar. It implies that rocks are at a temperature of ca. 500 °C at less than 10 km depth. The surface is attained by slow cooling during progressive erosion of the above lying exposed crustal levels. Figure 5 illustrates this type of P-T path, documented for the UHT granulites of Central Sri-Lanka (Touret et al., 2019b), which can be taken as typical example for UHT granulites worldwide.

In contrast to the relatively consistent P-T paths, the duration of UHT metamorphic episodes is rather variable. Based on single-zircon dating, Harley (2016) finds less than 15 Ma for the recent granulites from Hokkaido (Japan) or Seram (Indonesia) to more than 100 Ma in the long-lived and slowly-cooled Napier complex (Antarctica), the most classic example of a UHT terrane. Comparable values are indicated by Kelsey and Hand (2015): ca. 15 million years for Southern Madagascar and Southern India to more than 150 million years for the Eastern Ghats Province in India. Values in other regions vary regularly between these two extremes. These observations have led Kelsey and Hand (2015) to distinguish short (< 5 to 40 Ma) and long (> 40 Ma) timescale categories for UHT metamorphism. The reasons of this discrepancy are not clearly understood. Some authors believe that the cause for extreme UHT conditions are mainly crustal, caused by elevated heat crustal production and slow erosion rates (e.g. Clark et al., 2011, 2015). In India, the duration decreases progressively between the two regional metamorphic episodes, Archean or early Proterozoic in the north (ca. 2.4 Ga), Eocambrian (Pan-African) in the south. UHT metamorphism lasts for more than 150 Ma in the north (Eastern Ghats Province, which includes in its southern part the famous outcrops of Shevaroy or Nilgiri), 20 Ma in the Central Indian Tectonic Zone, which marks the limit between the two domains, and less than 10 Ma in the Pan-African rocks of Kerala and Sri-Lanka. It is hard to see any drastic changes in the lithology or erosion rates between these different domains, which in the field appear to be continuous. It is difficult to understand how crustal processes, e.g. rates of erosion or amount of heat-producing minerals could explain these differences in the duration of UHT-metamorphism. A more logical explanation for this is the presence of a mantle plume head, i.e. magma intrusions at the base of the crust or at the crust-mantle interface during the whole period of UHT metamorphism. This plume head has a different duration in short-versus long-lived orogens, but in all cases, the disruption of the supercontinent starts very shortly after the end of the plume activity.

FLUIDS IN ULTRAHIGH TEMPERATURE GRANULITES

Virtually all granulites worldwide contain the same types of fluid inclusions, namely pure CO₂ of variable density and highly saline aqueous solutions (brines) (e.g. Touret and Huizenga, 2011; Harlov, 2012). Ultrahigh temperature granulites are no exception, but they differ from normal high-temperature granulites in several ways.

Abundance and density of CO₂ fluids. Compared to other granulites, CO₂ inclusions in UHT granulites show three specific characteristics:

(1) Number and size of inclusions. Most rock-forming minerals (first of all orthopyroxene, plagioclase and garnet) contain many large (about 30–50 microns in size) and well preserved primary CO₂ inclusions, most of them with a nice negative-crystal shape. Typical UHT minerals (sapphirine, osumilite) are best found in Mg-Al-rich, silica-deficient rocks. Hence quartz is relatively rare, but when occurring it contains often very few or no inclusions at all, in sharp contrast to what is observed in most other rock types. This is probably due to the easy recrystallization of quartz at high temperature, which tends to wipe out all inclusions. Early publications on UHT granulites from former Gondwana have all insisted on the number of well-preserved, high-density and primary CO₂ inclusions: Napier Complex, Antarctica (Tsunogae et al., 2002), Shevaroy Hills, Southern India (Santosh and Tsunogae, 2003), Eastern Ghats Belt, India (Santosh et al., 2004), Sri-Lanka (Bolder-Schrijver et al., 2000). The same can be said for older UHT occurrences on the margin of the African craton (Fig. 3): In Ouzzal, Sahara, Algeria (Cuney et al., 2007) and the Bakhuis, Suriname (this study). This list is not limited. In fact, we do not know any example of UHT granulites, which do not contain this type of primary, high-density

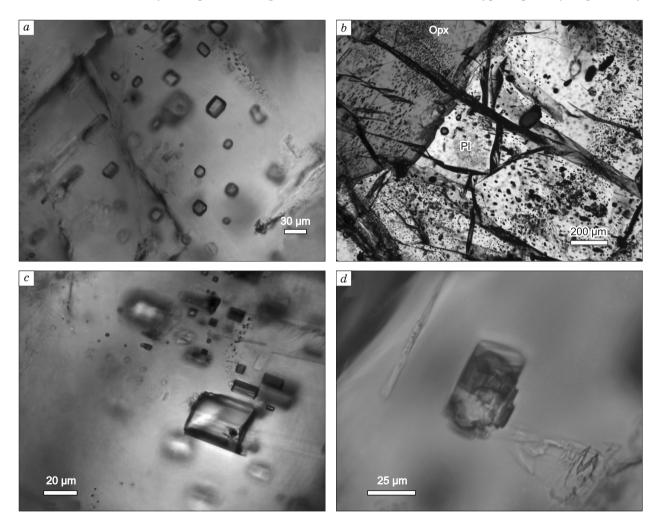
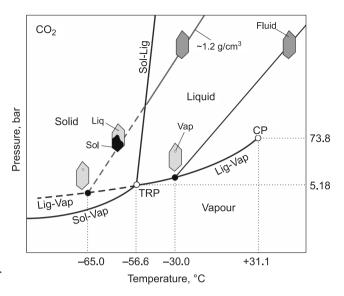


Fig. 6. CO_2 fluid inclusions in UHT sapphirine-bearing granulite from Harukutale, Central Sri-Lanka (Bolder-Schrijver et al., 2000). a, Group of primary CO_2 inclusions in plagioclase; b, illustration of the large fluid inclusion abundance (black dotes) in plagioclase (Pl) and orthopyroxene (Opx); c, detail of primary, negative crystal shaped CO_2 fluid inclusions in plagioclase; d, dark CO_2 fluid inclusion containing a crystal of Mg-carbonate (white).

Fig. 7. CO_2 pressure-temperature diagram showing isochores for a dense (black solid line, Th is ca. -30 °C) and a super dense (dark-grey solid and dashed line, metastable Th is ca. -65 °C) CO_2 fluid inclusions. Th: temperature of homogenization into the liquid phase; CP, Critical point for pure CO_2 (+31 °C); TRP, triple point of pure CO_2 (56.6 °C).

pure CO₂ fluid inclusions. One example is given in Fig. 6, UHT granulites in sapphirine-bearing granulites from Udadigana, Harakutale, Central Sri-Lanka (Bolder-Schrijver et al., 2000). A photo can only show inclusions close to the surface of the section, but these are equally dispersed within the whole mass of the rock. A very conservative estimate of their number would be several 100's per mm³, cor-



responding to 5–10 vol.%. As most inclusions contain CO_2 of a density approaching (or exceeding) 1 g/cm³ (see below), it indicates that the quantity of CO_2 still contained in the rock is of the order of 1 to 3 wt.%! The abundance of CO_2 at peak metamorphic conditions is further indicated by the high CO_2 content in channel minerals, first of all cordierite: 1.05 wt.% in the Napier Complex (S. Harley in Santosh et al. (2004)), up to ca. 3 wt.% in a metapelite from Bakhuis, Suriname, the highest level known from nature (De Roever et al., 2016).

(2) Dense and super dense CO₂ fluid: The CO₂ density varies from sample to sample, but it is generally high to very high. In garnets from Salem (Shevaroy Hills, Southern India) UHT granulites, Santosh et al. (2004) found CO₂ fluid inclusions which homogenize within the narrow temperature range of -52.3±2.4 °C, corresponding to densities of 1.17–1.16 g/cm³. These were at this time considered to be by far the highest density CO₂ yet reported from continental crust. However, even greater densities have been found in real super dense CO₂, namely a fluid which homogenize below CO₂ triple point (-56.6 °C), along the metastable extension of the liquid-vapor curve (Fig. 7). These remarkable fluids (Van den Kerkhof and Thiéry, 2001) are not restricted to UHT granulites. They can occur through late volume decrease of the inclusion cavity during post-metamorphic isobaric cooling (see discussion in Touret, 2001). However, in the case of UHT granulites, the primary character of the inclusions, the lack of any sign of post-trapping perturbation and the fact that fluid isochores

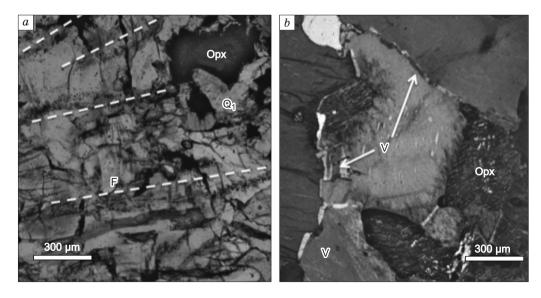


Fig. 8. Contrasting mechanisms of fluid migration in incipient charnockites from South India and Sri-Lanka. a, Kottavatam, Kerala (Touret et al., 2019a). Orthopyroxene (Opx) is growing in a cavity (geode) mantled by idiomorphic quartz crystals with a Tessiner habitus (Q_1). Post-metamorphic fluids are trapped as secondary fluid inclusions along healed microfractures (F) b, Kurunegala, Sri-Lanka (Perchuk et al., 2000). V = K-feldspar microveins along intergrain boundaries between quartz and perthite phenocrysts.

match P–T conditions of mineral equilibration indicate with a good degree of confidence that these dense and super dense inclusions have really preserved fluids, which were present in the rock system at peak metamorphic conditions.

Minimum values measured so far (-65 °C) is in fluid inclusions in quartz in Bakhuis, Suriname, UHT granulites. It indicates a CO₂ density of ca. 1.2 g/cm³ corresponding to a pressure of more than 10 kbar for a temperature of 1000 °C. The very good preservation of these primary inclusions, despite the very high P-T metamorphic conditions, is best explained by the fact that, during the early stage of the retrogression, the pseudo-isobaric P-T path is roughly parallel to the high-density CO₂ isochores (Fig. 7). Starting above 1000 °C, this P-T trajectory typically continues to about 500 °C, at which the internal CO₂ pressure in the inclusion is of the order of ca. 5 kbar. This is followed by decompression from ca. 5 kbar to less than ca. 2 kbar occurs in less than 100 °C, resulting in the large decompression textures around garnet, found in many UHT granulites. The reason why CO₂ inclusions have better resisted to this decompression than many rock-forming minerals is not well understood. In normal, quartz-bearing granulites, such a decompression leads to prominent sets of secondary inclusion trails, progressively grading into open microfractures. An example is given in Fig. 8a, an incipient charnockite from Kurunegala, Sri-Lanka (Fig. 3) (Perchuk et al., 2000). In this rock, some orthopyroxene crystals grow inside a circular cavity mantled by idiomorphic quartz crystals. Invariably one crystal, much longer than the others, displays a typical "Tessiner" habitus (Rykart, 1995), indicative of a CO₂-rich fluid environment (Mullis, 1996). Many remnants of CO₂ inclusions are found in this quartz, showing that, at peak metamorphic conditions, free CO₂ fluids did exist in the rock, able to maintain open cavities at a crustal depth of at least 60 to 70 km. During retrogression, these fluids leave the rock system through a set of parallel healed fractures (trails of secondary inclusions, Fig. 8a), progressively grading into open microfractures. The presence of these microfractures may be numerous, giving sometimes the impression of a secondary cleavage (unpublished observations of the senior author in the Bamble province of Southern Norway). Minerals like orthopyroxene or feldspars are less sensitive than quartz to secondary recrystallization, possibly helped by the equilibrium, the negative-crystal shape of the inclusions, and the non-reactive character of the CO₂ fluid.

(3) Carbonate inclusions in dense CO₂ fluids: As briefly mentioned earlier, many CO₂ inclusions contain solid carbonate, either a single, more of less cubic-shaped crystal, or an aggregate of several idiomorphic microcrystal. SEM images of opened inclusions indicate a Mg-rich composition of the carbonate (dolomite or magnesite) (Bolder-Schrijver et al., 2000). The volume ratio between carbonate and CO₂ and the composition of the carbonate are constant in all inclusions and not related to the inclusion shape or nature of the mineral host. In contrast to the carbonate globules mentioned earlier, these carbonates cannot be issued from a reaction between CO₂ and brines fluids (which would have consumed all CO₂) or from a reaction between the CO₂ fluids and the mineral host (which would have produced carbonates of different size and composition, depending on the Mg-Ca composition of the mineral host). Homogenization temperature of the CO₂ in carbonate-bearing inclusions is about 20 °C higher than in carbonate-free inclusions. Consequently, the CO₂ isochore of the carbonate-bearing inclusions passes significantly below the P-T box of peak metamorphic conditions, but it approaches those P-T conditions if the molar volume of the solid carbonate is reintegrated into the total molar volume of the fluid-solid system (see discussion in Bolder-Schrijver et al., 2000). For these reasons, we believe that the incoming CO₂ in UHT granulites at Harakutale at peak metamorphic conditions was not pure CO₂, but a mixture of CO₂ and carbonate. We have hypothesized that the source could be a carbonate melt, partly destabilized when entering the lower crust (Touret and Huizenga, 2012b).

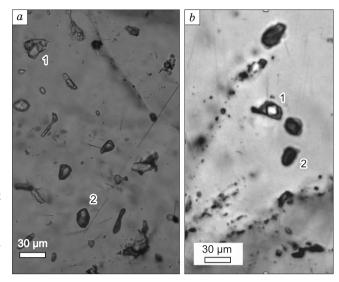
Brine fluids. Besides CO₂, UHT granulites contain also traces of high salinity aqueous fluids (brines), like all other granulites (Touret and Huizenga, 2011). These fluid inclusions are less abundant and smaller than CO₂ inclusions, and, therefore, much more difficult to be studied. A liquid phase is commonly absent, inclusions are made of a small, irregular cavity squeezed around one or several solids, the larger being invariably isotropic halite (referred to as collapsed inclusions, (Touret, 2001)). Isochores for brine fluid inclusions are invariably steeper than those for CO₂, i.e. the fluid pressure in brine inclusions is less than the lithostatic pressure during the early stage if the retrogression, unable to remain unchanged at these high temperature. But the fact that they were already NaCl saturated implies a very high salinity, at least 50 wt.%. Luckily, some fluid inclusions have escaped this transposition, one example being the Bakhuis UHT granulites (Surinam, South America), in which isolated brine inclusions occur within groups of more abundant CO₂ inclusion (Fig. 9a). With a few exceptions, e.g. in In Ouzzal UHT granulites (Cuney et al., 2007), immiscible brines and high-density CO₂ fluids occur in virtually all UHT granulites studied so far. Figure 9b shows another example in an igneous charnockite occurring in Southern Norway. This rock belongs to a granulite "island", north of the regional amphibolite-granulite isograd (Nijland et al., 1998). The high metamorphic temperature of this rock (>900 °C) is indicated by feldspar mesoperthites. Similar rocks are found in most UHT granulite occurrences, indicating that the very high temperature reached is caused by magmatic intrusions during the peak of metamorphism.

Brine inclusions are less common than CO₂ inclusions, especially in UHT granulites. However, their role in rock transformation is significant due to their mobility and capacity of element transport. They are instrumen-

Fig. 9. Examples of primary brine (1) and dense to high-dense CO_2 (2) fluid inclusions demonstrating immiscibility in UHT granulites at peak metamorphic conditions: a, Bakhuis, Suriname; b, Igneous charnockite, Bjornasvatn, Southern Norway. In the lower part of the section, trails of smaller secondary inclusions correspond to the post-metamorphic escape of the primary fluids.

tal for the formation of the so-called "incipient charnockites", namely the local (cm to m scale) transformation of gneiss into igneous-looking charnockites (Touret et al., 2019a), as will be discussed in more details in the following section.

In conclusion, fluid inclusion studies demonstrate without ambiguity that free fluids did exist in



the rock system at peak metamorphic conditions, in the form of dominant dense to super dense CO₂, accompanied by immiscible brines. Then the next obvious questions arise: how much fluids were present and where do they come from?

HOW MUCH FLUID: INDIRECT EVIDENCE

Well-preserved fluid inclusions only provide a minimum estimate of the total fluid quantities present in the rock system at the time of the inclusion formation. As discussed above, this minimum is far to be negligible, especially for CO₂. On the other hand, another indication is given by the extraordinary variety and extent of secondary effects caused by metamorphic fluids at the onset of regression. These effects have been discussed elsewhere (Touret and Nijland, 2013; Newton et al., 2019; Touret et al., 2019a) and only a brief summary will be given here.

Fluids are introduced in the lower crust at peak UHT-conditions in the form of immiscible droplets of high-density CO_2 and high-salinity brines, as found e.g. in Bakhuis, Suriname (Fig. 9a). However, their reactivity and migration capacity are grossly different, highly reactive brines being much more mobile along intergrain boundaries. Besides the often quoted difference in wetting angle (Watson and Brennan, 1987; Harlov, 2012), a major cause for the difference in migration capacity is to be found in the systematic difference in fluid isochore slope between carbonic and aqueous systems (see discussion in (Touret, 2001)). Flat high-density CO_2 isochores are roughly parallel to the isobaric cooling P-T path, which occurs systematically after UHT-metamorphism. Internal fluid pressure in the fluid inclusion remains similar to the lithostatic pressure until the end of isobaric trajectory. Only during the subsequent sudden decompression, e.g. in Sri-Lanka at a temperature of about 700 °C (Touret et al., 2019b), CO_2 will migrate through a network of healed microfractures. Alternatively, aqueous fluid inclusions have a fluid pressure, which is less than the lithostatic pressure at the onset of regression, resulting in successive transpositions, allowing the fluid to reach the grain boundary of the host mineral.

The result is that both fluid systems behave independently, not interacting with each other until the final stage of regression at ca. 300–400 °C, at which brine fluids can react with CO₂ to give late carbonates (seen systematically in all granulites as micro-inclusions, notably in feldspars). Free CO₂ remains exclusively within the granulite domain. Its geochemical influence is minor, restricted to the stabilization of the characteristic granulite anhydrous mineral assemblage (first of all, orthopyroxene). Alternatively, saline aqueous fluids circulate pervasively in and outside of the HT and UHT granulite areas with important geochemical consequences, especially within the igneous looking charnockites (Touret and Huizenga, 2012). In the granulite domain, brine streaming along intergrain boundaries results mainly in the formation of K-feldspar microveins along quartz-plagioclase or plagioclase-plagioclase grain boundaries (e.g., Harlov and Wirth, 2000). When they reach the outer limit of the granulite areas, they are able to cause the transformation of various lithologies (metapelites, amphibolites, and rocks belonging to the typical Archean trondhjemite, tonalite, granodiorite (TTG) series) into coarse-grained, igneous looking charnockites. These incipient charnockites were first identified in the Archean Darwar craton of India (Pichamutu, 1960) (Fig. 3a) and later in the Neoproterozoic Province of South India (Kerala) and Sri Lanka. They occur at close distance (typically a few 100 m to km) of the regional granulite/amphibolite facies boundary in most HT/UHT granulite terranes. The fact that some incipient charnockites oc-

cur along a network of brittle fractures led most authors to postulate that they were caused by fluid streaming (Friend, 1985), first believed to be CO₂ when it was thought that CO₂ was the only active granulite fluid (e.g. Santosh et al., 1990). However, a recent re-investigation of fluid inclusions in incipient charnockites from type localities in Southern India (Touret et al., 2019a) confirms without ambiguity that brines are the fluids responsible for incipient charnockitization; CO₂, when present, is a consequence and not the cause. Besides brittle fractures, incipient charnockites may also occur as rounded patches, with textures showing evidence of melting (Perchuk et al., 2000, Touret and Huizenga, 2012a) (Fig. 3b). A detailed investigation of the conditions imposed by the brines (P, T, H₂O activity) shows indeed that gneiss melting can occur under the influence of brine streaming, suggesting this mode of formation for a number of massive charnockites (Newton et al., 2019). Brines may focus along large shear-zones to invade the adjacent amphibolite domain, as seen in India north of the so-called Fermor line, the northern limit of Archean granulites. The occurrence in this domain of elongated granite bodies (including the famous Closepet granite) suggests a new way to look at the relations between charnockite and granite in the low to middle crust (Newton et al., 2019). The Closepet granite was formed at a lower pressure than the charnockite (4 to 8 kbar (Moyen et al., 2003)). At a relatively low pressure (ca. 4 kbar), CO₂ fluids are expelled out of the crystallizing magma in the form of veins (Battacharya et al., 2014). Biotite remains stable and the final rock is a granite. The relationships between charnockite and granite appears then to be strictly fluid-dependent: high-salinity aqueous brines induce the metasomatic-melting effects, while CO₂, if pervasively present, stabilizes the charnockite diagnostic minerals including orthopyroxene. The systematic occurrence of saline inclusions in potassic granites and syenites, unrelated to boiling except for very shallow intrusions (Cu-porphyries (Roedder, 1984)) (e.g., Konnerup-Madsen, 1984), suggests that this process of fluid streaming through the crust might be important for the formation of granites in general.

In all granulite terranes, high-temperature transformations are continued at decreasing temperature by intense alkali metasomatic effects, which under various names (albitization, scapolitization, K-feldspathization) can cover region of subcontinental size, e.g. in the southern part of the Scandinavian shield (Sweden, Hoeve, 1978; or Norway, Engvik et al., 2014). This type of metasomatism, which has important metallogenic consequences (Fe-Ti ores in Norway, U in Sweden and elsewhere), is far more developed in the regional amphibolite-grade gneisses than in the granulite core. The source of brines is probably multiple, but in Southern Norway there is compelling evidence (Touret and Nijland, 2013; Engvik et al., 2014) that brine fluids were released during prograde metamorphism of pelitic sediments, including evaporites. These fluids were present almost everywhere at the onset of peak granulite metamorphism, closely associated with metasediments. Besides halides, sulfates (notably anhydrite) are common daughter minerals in inclusions, explaining the highly oxidized character of many granulitic areas (Harlov et al., 1997). It is not certain, however, that the example of Southern Norway is valid everywhere. The carbonatite connection for the origin of CO₂, discussed below, offers another possibility, which needs to be further documented.

EFFECTS INDUCED BY CO₂ FLUIDS: QUARTZ-CARBONATE AND GRAPHITE MEGASHEAR ZONES

The widespread occurrence of late- to post-metamorphic Na-metasomatism indicates that most metamorphic Na-bearing aqueous fluids left the rock at the end of the metamorphic episode. However, a significant quantity of brine fluid may remain trapped along the intergrain boundaries, as indicated by high-conductivity layers at the base of most continental areas (Manning, 2018). The conservation of CO₂ is more problematic, except for the fluid remnants preserved in mineral inclusions. Many granulite terranes are either intersected or surrounded by prominent crustal-scale shear zones, sometimes up to ca. 100 km long with a width of ca. 1 km showing evidence of large scale, fluid-induced metasomatism. Some, containing mainly pure quartz, have been termed carbonated shear zones, displaying pervasive and vein-controlled carbonate metasomatism (Newton and Manning, 2002). Examples include the late Proterozoic Valley of Tamil Nadu, India (Wickham et al., 1994), the Late Archean Chitradurga area of Karnataka, India (Chadwick et al., 1989) and the mid-Proterozoic Bamble Shear belt of Southern Norway (Dahlgren at al., 1993). On the contrary, other shear zones are highly reduced, with the best example being the vein-type graphite of Sri-Lanka, the world largest resources of highly crystalline graphite (Fig. 3). Graphite veins are also surrounded by potassic feldspar alteration zones of variable size (Touret et al., 2019b). Fluid inclusion studies and P-T-oxygen fugacity interpretation of the conditions of formations of the different veins (see discussion in (Touret et al., 2019b)) demonstrate the involvement of both granulite fluids in the two types of shear zones redox settings: brines in the oxidized shear zones, and CO₂ in a reduced setting. In the quartz-carbonate megashear zones, the high oxygen fugacity is imposed by the sulfate-bearing brines (Newton and Manning, 2004), whereas the reduced character of the graphite veins is caused by abundant sulfide minerals in the host-rocks (Katz, 1987). During the reduction of CO₂, the residual fluid becomes enriched in H₂O (e.g., Huizenga, 2011; Touret et al., 2019b), which is responsible for the alteration zones found on both sides of the graphite veins.

Megashear zones are the most spectacular effects induced by the release of lower crustal CO₂ fluids at the end of the UHT-metamorphic episode. They are the best illustration of the fundamental difference in the mechanisms of migration of brines and CO₂ fluids within the crust (Fig. 8). Successive episodes of brittle deformation must have caused micro seismicity, grading into more significant seismicity as indicated by the frequent occurrence of pseudotachylites in granulites (Clarke and Norman, 1993). Propagating through the entire crust, this mechanism is a very effective way to transfer deep CO₂ towards the surface, as shown by the persistent CO₂ emissions following major earthquakes (Girault et al., 2018). This effect is, however, limited in geological time. Alternatively, the common occurrence of high-conductivity layers at the base of continents indicates that a significant quantity of brine fluids, probably stored along mineral intergrain boundaries, might remain within the lower crust at the end of the metamorphic episode (Manning, 2018).

FLUID ORIGIN: THE CARBONATITE CONNECTION

The dry character of granulite terranes has led to the widely accepted concept of fluid-absent (or vaporabsent) rocks, in which any free fluid, tacitly or explicitly assumed to be water, cannot exist (Thompson, 1983). Syn-metamorphic fluid is either dissolved in melts, or bound in mineral structures. This is indeed the case for pure water: at HT- or UHT metamorphic temperatures, any free water will immediately provoke partial melting and be dissolved in the melt. This mechanism is quite obvious in all high-grade rocks, but, contrary to common belief, melts do not leave the system systematically. Many granulites are migmatites, in which both initial lithology and products of partial melting still coexist at the outcrop scale. Melts are only expelled towards more superficial levels if produced in sufficient quantity or in zones of special deformation. As the rate of melt production depends on the availability of H₂O (relatively little in the dry granulite terranes), granite melts are far more abundant in amphibolite-grade migmatites than in granulites. However, in these, partial melting is a mechanism able to generate both CO₂, when the protolith was carbon-bearing (metapelites), and brines, by consuming progressively the water released by crystallizing melts to build up hydrous minerals (desiccation effect (Markl and Bucher, 1998)). Both processes have been documented, but they are not likely to produce large fluid quantities. This is evident for brines, which, according to the desiccation mechanism, can only be minor remnants strictly confined to former granitic melts. For CO₂, burning of former organic matter, notably in metapelites, can provide important CO₂ quantities. This is indeed observed in the Bamble province of Southern Norway, which contain a number of graphite-bearing paragneisses, but these contain also sulfides, leading to immediate reduction of high-temperature CO₂ to form graphite during retrogression (Huizenga and Touret, 2012). Another possible source of carbon would be sedimentary carbonates, occurring in many granulite areas in the form of marbles or, in Bamble, in garnet-clinopyroxene bearing skarns. In Southern Norway, these skarns contain the major iron-ore deposits (Arendal region), but, interestingly, only brine inclusions have been found in these rocks; CO₂ is very rare or completely absent (Touret, 1985). The fact that sedimentary carbon can only make a minor part of the high temperature CO₂ found in HT and especially UHT granulite is further indicated by the isotopic signature of the inclusion fluid. Inclusions in felsic granulites from the Bamble area contain isotopically light carbon (δ ¹³C = 24.2 to 14.1‰), interpreted as a mixture between light organic and a variable proportion of heavier carbon, either a minor quantity of sedimentary carbonate or some mantle-derived CO₂ (Pineau et al., 1981). The mantle signature is more clear for synmetamorphic carbonates found in partly amphibolized basic intrusions (hyperites), emplaced at peak metamorphic conditions ($\delta^{13}C = 8.2\pm1\%$). These data refer to HT granulites, but UHT granulites show much more clearly a dominant, if not exclusive mantle signature, either from noble gases (mainly He) (Dunai and Touret, 1993) or from the carbon isotope composition of graphite in Sri-Lanka (Touret et al., 2019b).

Due to the lack of a suitable geochemical tracer, the origin of brines remains uncertain (e.g., Manning, 2018). Brines issued from metasediments, including former evaporites are widespread in Bamble area in and out of the regional granulite area (Touret, 1985). Derived from former sedimentary formations waters, these brines are probably responsible for regional albitization-scapolitization or for the formation of granulite islands, but we have no evidence that they could participate to an immiscible brine-CO₂ system at UHT peak conditions. Alternatively, the association of CO₂ and carbonates, both with a mantle signature, indicates a clear relation with carbonatite melts. Such an origin fits also well with the rather limited size of the UHT granulite occurrences: carbonatite melts can be transported by magmas emplaced at the base of the crust, providing both the heat and fluids of UHT granulite metamorphism. This carbonatite connection is further supported by the many intrusions of carbonatites and associated rocks (nepheline-syenite and other types of alkaline intrusions) found in the vicinity of in former UHT areas. Figure 3 illustrates the great number of alkaline intrusions in Gondwana UHT occurrences. Typical carbonatites are less abundant, but some are found in all regions mentioned in this paper, e.g. Eppawala, Wanni Complex, Sri-Lanka (Weerakoon et al., 2001; Manthilake et al., 2008), Tamil Nadu, India (Natarajana et al., 1994), In Ouzzal, Algeria (Ouzegane et al., 1988), and Bakhuis, Suriname (Kroonenberg et al., 2016). These intrusions are of different age: Paleoproterozoic in Tamil Nadu or along the

margin of the West African craton (In Ouzzal and Bakhuis), Eocambrian (Pan-African) in Central Gondwana. They post-date the metamorphic episodes by a variable time span: tens of millions of years in most cases, up to several hundred million years when they are related to large-scale rifting in the consolidated cratons: e.g., the Fen carbonatite in Southern Norway (Permian Oslo graben) and active carbonatite volcanism in the East Africa Rift.

The carbonatite connection offers another possibility for the origin of brines in the UHT granulites. Salts are common constituents of carbonatites (e.g., Safonov et al., 2010) which contains inclusions strikingly similar to those found in UHT granulites (Guzmics et al., 2011). Rapidly grown fibrous diamonds contain highly saline fluid inclusions. These brine fluids were trapped at a depth of at least 200 km and derived from silicic and carbonatitic deep mantle melts (Weiss et al., 2015). Fluids issued from subducted, salt-bearing altered oceanic crust (Sharp and Barnes, 2004) cause mantle metasomatism, which is the source of carbonatite melts (Meen, 1987). Remnants of chloride-carbonate melts have also been found in diamonds from metasomatized mantle eclogites (Zedgenizov et al., 2018). Spectacular associations of carbonate and halite have been observed in Udachnaya-East kimberlite (Kamenetsky et al., 2012; Abersteiner et al., 2018) (Figs. 13-8, p. 821 in (Frezzotti and Ferrando, 2018)). Chlorides are found in many kimberlite worldwide but the Udachnaya kimberlite is unique by its extremely high chlorine concentration (up to 6 wt.%) and the presence of chloride-bearing segregations. It is, however, not clear in this case if the salt is magmatic or resulted from evaporite contamination during kimberlite eruption (Grishina et al., 2014a,b; 2018). However, besides this discussed example, there is ample evidence that, in general, carbonatites can provide a substantial amount of the salts found in UHT fluids, with significant geochemical consequences. Carbonatite melts are formed by partial melting of a mantle metasomatized by fluids issued from former subduction. That these fluids may carry salts is not surprising. More surprising, though, is the fact that the mantle metasomatic source may have remained at the same place for millions of years, despite the complicated history of the overlying crust.

ULTRAHIGH TEMPERATURE AND SUPERCONTINENT AMALGAMATION: A SYNTHESIS

Petrological evidence and fluid remnants contain in mineral inclusions indicate that the lower continental crust was invaded by large quantities of mantle-derived CO₂ fluids during episodes of UHT metamorphism. This CO₂ might be accompanied by high-salinity aqueous brines, or mix with internally derived fluids to form an immiscible CO₂-brine system. UHT occurrences were relatively limited in space, related to the proximity of synmetamorphic magmatic intrusions and responsible for the very high temperature attained during peak UHT metamorphism. UHT occurrences were instrumental for sealing the final stage of supercontinent amalgamation. However, the large fluid amount stored in the lower crust might have caused instability, preparing a disruption which has started immediately after amalgamation (Touret and Huizenga, 2012). Post-metamorphic evolution was determined by the physical-chemical properties of the migrating fluids: pervasive granulitization and alkali metasomatism for the mobile brines, brittle fracturing for the less mobile CO₂. This type of evolution is best documented for the Gondwana supercontinent, but there are indications that it was also valid for older supercontinents including Rodinia and Columbia.

Figure 10 illustrates tentatively the details of this four-stage evolution, based on the Gondwana UHT occurrences (South India and Sri-Lanka). During final amalgamation of a supercontinent (Fig. 10a), two cratonic blocks are separated by a mobile belt, which seals the amalgamation. Ancient altered oceanic crust, subducted near the asthenosphere/lithosphere boundary, liberates fluids and melts which metasomatize the overlying mantle. Remnants of these fluids can be found in inclusions in diamonds, e.g. saline fluids issued from melted altered oceanic crust and/or sediments. During this process, various types of restite material can be formed, notably garnet-bearing eclogite and lherzolite (Simakov, 2003).

The amalgamation starts with an episode of UHT metamorphism in the lower crust of the mobile melt, triggered by a plume head rising from the deep mantle. It induces melting of the metasomatized mantle and the formation of carbonatite melts, immiscible in silicic melts and carried upwards in these melts, of average basaltic composition.

The extreme temperature of UHT metamorphism at the base of the crust (>1000 °C) is sufficient to maintain temperatures above minimum granite crustal melting in most of the continental crust, up to a depth of few kilometers (Fig. 10b). It will generate a variety of granitoid magmas, possibly mixed with differentiated melts issued from the mantle. The example of Southern India (Karnataka, Madras, and massive charnockite and Closepet granite) shows that final mineralogy at magma crystallization depends on local H₂O activity: charnockite (orthopyroxene-bearing) for low H₂O activity, granite (biotite-bearing) for high H₂O activity. The difference in H₂O activity delimitates two crustal domains: a granulite facies lower crust and amphibolite facies middle crust. The transition between charnockite and granite is progressive, incipient charnockites are situated at the granulite/amphibolite facies boundary.

The H₂O activity is determined by fluids are found in mineral inclusions. In lower crustal granulites, fluids are made of dense CO₂ and high salinity brines, buffering the H₂O activity to ca. 0.2–0.3 at the end of

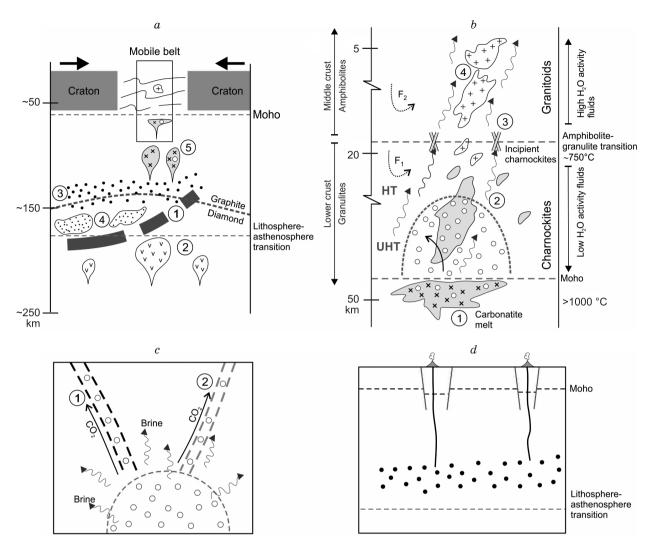


Fig. 10. Synthesis of UHT granulite and supercontinent amalgamation (see text for a detailed explanation).

a, Supercontinent final amalgamation stage. 1, Subducted former altered oceanic crust; 2, Rising deep-mantle plumes; 3, Metasomatized mantle; 4, Restite from melted subducted crust (garnet-bearing eclogite and lherzolite); 5, Carbonatite melts and basaltic melts carrying immiscible carbonatite melts; b, detail of the insert (mobile belt) in a. 1, mantle intrusions carrying immiscible carbonatite melts. The dashed red line indicates the accumulation of dense/hyper dense CO₂ (open dots) in UHT granulite domains; 2, lower crustal charnockite (massive). 3, incipient charnockite; 4, middle crustal granites (for example Closepet granite, South India) including coarse-grained monzogranite and fine-grained subvolcanic granite. F1 and F2 (fine-dashed arrows): Locally derived fluids in the lower- and middle crust, respectively. Thick arrow: CO₂, thin contorted arrow: brine; c, Elimination of CO₂ in lower crust UHT granulites during decompression and cooling. 1, graphite veins; 2, quartz-carbonate mega-shear zones; d, carbonatite eruptions in the stabilized craton triggered by extension.

charnockite magma crystallization (F_1 , Fig. 10b). The H_2O activity is much higher (close to 1) in the amphibolite facies middle crust, due to a much greater amount of H_2O (F_2 , Fig. 10b). A great amount of these fluids is locally derived by a variety of processes: progressive dehydration or decarbonation, preferential dissolution of H_2O in granite melts, oxidation of former organic matter (CO_2) or, for brines, inherited sediment waters of dissolution of evaporites. These fluids are found in all granulites and granites, but, for UHT granulites, a special characteristic is the great quantity of additional CO_2 (and possibly brines) liberated by the breakdown of carbonatite melts issued from the underlying metasomatized mantle.

Mobile brines can circulate freely in the lower crust, either in the UHT granulite or neighboring normal HT granulite. They can reach the granulite/amphibolite boundary, being responsible for the formation of incipient charnockites. Less mobile CO₂ remains in place as long as the temperature and lithostatic pressure temperature are high enough to maintain a fluid density that is in equilibrium with the surrounding rock.

When the pressure and temperature decreases at the end of the UHT metamorphic episode, CO₂ has to leave the lower crust (Fig. 10c). The CO₂ release is mediated through a system of extension fractures, ranging

from crystal-size microfractures to crustal-scale zones, probably related to major earthquakes. Depending on the redox state, CO₂ is either reduced (graphite veins) or oxidized in the form of quartz-carbonate veins (Touret et al., 2019b). Mobile brines can invade pervasively large areas, causing various forms of alkali metasomatism (albitization, scapolitization). The oxygen fugacity is either internally or externally controlled, i.e. through sulfate-bearing brines (oxidized), or sulfide minerals (reduced), respectively.

Noteworthy is the fact that the layer of metasomatized mantle may persist for a very long time at the base of the lithosphere under the stabilized supercontinent (Touret and Huizenga, 2012b; Touret et al., 2016). It reacts to surface rifting by emitting various types of alkaline magmas and carbonatites (Fig. 10d). Examples of these intrusions are best seen on the periphery of the Gondwana supercontinent (Fig. 2), but they do occur in virtually all UHT occurrences worldwide.

ACKNOWLEDGEMENTS

This work is dedicated to professors V.S. Sobolev and Yu. A. Dolgov. The senior author had the privilege of meeting them in Novosibirsk more than fifty years ago.

REFERENCES

Abersteiner, A., Kamenetsky, V.S., Golovin, A.V., Goemann, K., 2018. Was crustal contamination involved in the formation of the serpentine-free Udachnaya-East kimberlite? New insights into parental melts, liquidus assemblage and effects of alteration. J. Petrol. 59, 1467–1482.

Bhattacharya, S., Panigrahi, M.K., Jayananda, M., 2014. Mineral thermobarometry and fluid inclusion studies on the Closepet granite, Eastern Dharwar Craton, south India: Implications to emplacement and evolution of late-stage fluids. J. Asian Earth Sci. 91, 1–18.

Bendaoud, A., Djemai, S., Ouzegane, K., Kienast, J.R. 2004. Le métamorphisme de trés haute température des granulites ferrifères et basiques de la region d'Amesmessa (Sud In'Ouzzal, Hoggar Occidental, Algérie). 20th Colloquium of African Geology, Orléans, France, Abstracts, p. 73.

Binu-Lal, S.S., Kehekpannala, K.V.W., Satish-Kumar, M., Wada, H., 2003. Multistage graphite precipitation through protracted fluid flow in sheared metagranitoid, Digana, Sri-Lanka: evidence from stable isotopes. Chem. Geol. 197, 253–270.

Bolder-Schrijver, L.J.A., Kriegsmann, L.M., Touret, J.L.R., 2000. Primary carbonate/CO₂ inclusions in sapphirine-bearing granulites from central Sri-Lanka. J. Metamorph. Geol. 18, 259–269.

Brown, M., 2006. Duality of thermal regimes is the distinctive characteristic of plate tectonics since the Neoarchean. Geology 34, 961–964.

Brown, M., Johnson, T., 2018. Secular changes in metamorphism and the onset of plate tectonics. Am. Mineral. 103, 181–196.

Burov, E., Guillou-Frottier, L., D'Acremont, E., Le Pourhiet, L., Cloething, S., 2007. The plume head–lithosphere interactions near intra-continental plate boundaries. Tectonophysics 434, 15–38.

Chadwick, B., Ramakrishnan, M., Vasudev, V.N., Viswanatha, M.N., 1989. Facies distributions and structure of a Dharwar volcanosedimentary basin: evidence or Late Archean transpression in southern India. J. Geol. Soc. London. 146, 825–834.

Cheney, E.S., 1996. Sequence stratigraphy and plate tectonic significance of the Transvaal succession of southern Africa and its equivalent in Western Australia. Precambrian Res. 79, 3–24.

Clark, C., Fitzsimons, I.C.W., Healy, D., Harley, C.L., 2011. How does the continental crust get really hot? Elements 7, 235–240.

Clark, C., Healy, D., Johnson, T., Collins, A.S., Taylor, R.J., Santosh, M., Timms, N.E., 2015. Hot orogens and supercontinent amalgamation: a Gondwana example from southern India. Gondwana Res. 28, 1310–1328.

Clarke, G.L., Norman, A.R., 1993. Generation of pseudotachylite under granulite facies conditions, and its preservation during cooling. J. Metamorph. Geol. 11, 319–335.

Cuney, M., Coulibaly, Y., Boiron, M.C., 2007. High-density early CO₂ fluids in the ultrahigh-temperature granulites of Ihouhaouene (In Ouzzal, Algeria), Lithos 96, 402–414.

Dahlgren, S., Bogoch, R., Magaritz, M., Michard, A., 1993. Hydrothermal dolomite marbles associated with charnockitic magmatism in the Proterozoic Bamble Shear Belt, south Norway. Contrib. Mineral. Petrol. 113 (3), 394–409.

De Roever, E.W.F., Hooijschuur, J., Harley, S., Huizenga, J.M., Nanne, J., Van Ryt, M., 2016. Widespread CO₂-rich cordierite in the UHT Bakhuis granulite belt, Suriname, in: 35th International Geological Congress, Capetown, South Africa. Abstract 1843.

- **Dharmapriya, P.L., Malaviarachchi, S.P.K., Santosh, M., Li Tang, Sajeev, K.,** 2015. Late-Neoproterozoic ultrahigh-temperature metamorphism in the Highland Complex, Sri-Lanka. Precambrian Res. 271, 311–333.
- **Doukkari, S., Diener, J., Ouzegane, K., Kienast, J.R.,** 2018. Re-evaluation of *P*–*T* estimates and reaction textures in the In Ouzzal UHT terrane, northwestern Hoggar, Algeria, through mineral equilibrium modelling and calculated chemical potentials, in: Granulites and Granulites 2018. Abstracts, p. 38.
- **Drüppel, K., Elsässer, L., Söhncke, B., Gerdes, A.,** 2013. Sveconorwegian mid-crustal ultrahigh-temperature metamorphism in Rogaland, Norway: U–Pb LA-ICP-MS geochronology and pseudosections of sapphirine granulites and associated paragneisses. J. Petrol. 54, 305–350.
- **Dunai, T.J., Touret, J.L.R.,** 1993. A noble-gas study of a granulite sample from the Nilgiri Hills, Southern India: implications for granulite formation. Earth Planet. Sci. Lett. 119, 271–281.
- Ellis, D.J., 1980. Osumilite-sapphirine-quartz granulites from Enderby Land, Antarctica: *PT* conditions of metamorphism, implications for garnet-cordierite equilibria and the evolution of the deep crust. Contrib. Mineral. Petrol. 74 (2), 201–210.
- Engvik, A.K., Ihlen, P. M., Austrheim, H., 2014. Characterisation of Na-metasomatism in the Sveconor-wegian Bamble Sector of South Norway. Geosci. Front. 5, 656–672.
- **Friend, C.R.L.,** 1985. Evidence for fluid pathways through Archaean crust and the generation of the Closepet Granite, South India. Precambrian Res. 27, 239–250.
- Frezzotti, M.L., Ferrando, S., 2018. The role of halogens in the lithospheric mantle, in: Harlov, D.E., Aranovich, L. (Eds.), Role of Halogens in Terrestrial and Extraterrestrial Geochemical Processes. Springer, pp. 805–845.
- Girault, F., Adhikari, L.B., France-Lanord, C., Agrinier, P., Koirala, B.P., Bhattarai, M., Mahat, S.S., Groppo, C., Rolfo, F., Bollinger, L., Perrier, F., 2018. Persistent CO₂ emissions and hydrothermal unrest following the 2015 earthquake in Nepal. Nat. Commun. 9, 2956.
- Grishina, S.N., Maksimovitch, Ya.E., Goryainov, S.V., Karmanov, N.S., Smirnov, S.Z., 2018. Chloride-containing segregations in the Udachnaya–Vostochnaya kimberlite pipe, in: The Problems of Magmatic and Metamorphic Petrology, Geodynamics and Genesis of Diamonds: Abstracts of International Conference Dedicated to the 110th Anniversary of Acad. V.S. Sobolev. Novosibirsk, p. 112.
- Grishina, S.N., Polozov, A.G., Smirnov, S.Z., Mazurov, M.P., Goryainov, S.V., 2014a. Inclusions in chloride xenoliths from the Udachnaya-East Kimberlite. Geochem. Int. 52, 595–603.
- Grishina, S.N., Polozov, A.G., Mazurov, M.P., Goryainov, S.V., 2014b. Genesis of chloride-carbonate segregations of the Udachnaya East Pipe. Dokl. Earth. Sci. 458, 1129–1131.
- Guzmics, T., Mitchell, R., Szabi, C., Berkesi, M., Milke, R., Abarth, R., 2011. Carbonatite melt inclusions in coexisting magnetite, apatite and monticellite in Kerimasi calciocarbonatite, Tanzania: melt inclusions and petrogenesis. Contrib. Mineral. Petrol. 161 (2), 177–196.
- **Harley, S.L.,** 2008. Refining the *P-T* record of UHT crustal metamorphism. J. Metamorph. Geol. 26, 125–154.
- **Harley, S.L.,** 2016. A matter of time: The importance of the duration of UHT metamorphism. J. Mineral. Petrol. Sci. 111, 50–72.
- **Harlov**, **D.E.**, 2012. The potential role of fluids during regional granulite facies dehydration in the lower crust. Geosci. Front. 3, 813–827.
- **Harlov, D.E., Newton, R.C., Hansen, E.C., Janardhan, A.S.,** 1997. Oxide and sulphide minerals in highly oxidized, Rb-depleted Archean granulites of the Shevaroy Hills Massif, south India: Oxidation states and the role of metamorphic fluids. J. Metamorph. Geol. 15, 701–717.
- **Harlov, D., Förster, H.J.,** 2002. High-grade fluid metasomatism on both a local and a regional scale: the Seward Peninsula, Alaska and the Val Strona di Omegna, Ivrea Verbano Zone, Norhern Italy. Part I. J. Petrol. 43, 769–799.
- **Harlov, D.E., Wirth, R.,** 2000. K-feldspar-quartz and K-feldspar-plagioclase phase boundary interaction in garnet-orthopyroxene gneisses from the Val Strona di Omegna, Ivrea-Verbano Zone, northern Italy. Contrib. Mineral. Petrol.140, 148–162.
- **Hoeve, J.,** 1978. Composition and volume changes accompanying soda metasomatic alterations, Västervik area, SE Sweden. Geol. Rundsch. 67, 920–942.
- **Huizenga, J.M.,** 2011. Thermodynamic modelling of a cooling C–O–H fluid–graphite system: implications for hydrothermal graphite precipitation. Mineral. Deposita 46, 23–33.
 - Huizenga, J.M., Touret, J.L.R., 2012. Granulites, CO₂ and graphite. Gondwana Res. 22, 789–809.
- Imayama, T., Oh, Ch.W., Baltybaet, Sh.K., Park, Ch.S., Yi, K., Jung, H., 2017. Paleoproterozoic high-pressure metamorphic history of the Salma eclogite on the Kola Peninsula, Russia. Lithosphere 9, 855–873.

- **Kamenetsky, V.S., Kamenetsky, M.B., Golovin, A.V., Sharygin, V.V., Maas, R.,** 2012. Ultrafresh salty kimberlite from the Udachnaya-East pipe (Yakutia, Russia): a petrological oddity or fortuitous discovery? Lithos 152, 173–186.
- **Katz**, **M.B.**, 1987. Graphite deposits of Sri Lanka: a consequence of granulite facies metamorphism. Mineral. Deposita 22, 18–25.
 - Kelsey, D.E., 2008. On ultrahigh-temperature crustal metamorphism. Gondwana Res. 13, 1–29.
- **Kelsey, D.E., Hand, M.,** 2015. On ultrahigh temperature crustal metamorphism: Phase equilibria, trace element thermometry, bulk composition, heat sources, timescales and tectonic settings. Geosci. Front. 6, 311–356.
- **Kihle, J., Bucher-Nurminen, K.,** 1992. Orthopyroxene-sillimanite-sapphirine granulites from the Bamble granulite terrane, southern Norway. J. Metamorph. Geol. 10, 671–693.
- **Konnerup-Madsen, J.,** 1984. Composition of fluid inclusions in granites and quartz syenites from the Gardar continental rift province (South Greenland). Bull. Mineral. 107, 327–340.
- **Kroonenberg, S.B., de Roever, E.W.F., Fraga, L.M., Reis, N.J.,** 2016. Paleoproterozoic evolution of the Guiana Shield in Suriname: a revised model. Neth. J. Geosci. 95, 491–522.
 - Manning, C., 2018. Fluids of the lower crust: deep is different. Ann. Rev. Earth Planet. Sci. 46, 67–97.
- **Manthilake**, **M.A.G.M.**, **Sawada**, **Y.**, **Sakai**, **S.**, 2008. Genesis and evolution of Eppawala carbonatites, Sri Lanka. J. Asian Earth Sci. 32, 66–75.
- **Markl, G., Bucher, K.,** 1998. Composition of fluids in the lower crust inferred from metamorphic salt in lower crustal rocks. Nature 391, 781–784.
- **Meen**, **J.K.**, 1987. Mantle metasomatism and carbonatites: An experimental study of a complex relationship. Geol. Soc. Am. Spec. Paper. 215, 91–102.
- Moyen, J.F., Nedelec, A., Jatyananda, M., Martin, H. 2003. From the roots to the roof of a granite: the Closepet granite of South India. J. Geol. Soc. India. 62 (6), 1–10.
- **Mullis, J.,** 1996. *P-T-t* path of quartz formation in extensional veins of the Central Alps. Schweiz. Mineral. Petrogr. Mitt. 76, 159–164.
- Natarajana, K., Bashkar Rao, B.B., Parthasarathy, B., Kunar, A., Gopalan, K., 1994. 2.0 Ga old pyroxenite-carbonatite complex of Hogenakal, Tamil Nadu, South India. Precambrian Res. 65, 167–181.
- **Newton, R.C., Manning, C.E.,** 2002. Experimental determination of calcite solubility in H₂O-NaCl solutions at deep crust/upper mantle pressures and temperatures: Implications for metasomatic processes in shear zones. Am. Mineral. 87, 1401–1409.
- **Newton, R.C., Manning, C.E.,** 2004. Solubility of anhydrite, CaSO₄, in NaCl–H₂O solutions at high pressures and temperatures: applications to fluid-rock interaction. J. Petrol. 46, 701–716.
- **Newton, R.C., Aranovich, L. Ya, Touret, J.L.R.,** 2019. Streaming of saline fluids through the Archean crust: Another view of charnockite-granite relations in southern India. Lithos 346–347, Article 105157.
- **Nijland, T., Touret, J.L.R., Visser, D.,** 1998. Significance of brines in the genesis of isolated granulite facies islands (Hauglandsvatn, Southern Norway). J. Geol. 106, 575–590.
- Osanai, Y., Sajeev, K., Nakano, N., Kitano, I., Kehelpannala, W.K.V., Kato, R., Adachi, T., Malaviarachchi, S.P.K., 2016. UHT-granulites of the Highland Complex, Sri-Lanka I: Geological and petrological background. J. Mineral. Petrol. Sci. 111, 145–156.
- Ouzegane, K., Fourcade, S., Kienast, J.R., Javoy, M. 1988. New carbonatite complexes in the Archaean In'Ouzzal nucleus (Ahaggar, Algeria): mineralogical and geochemical data. Contrib. Mineral. Petrol. 98 (3), 277–292.
- **Perchuk, L.L., Safonov, O.G., Gerya, T.V., Fu, B., Harlov, D.E.,** 2000. Mobility of components in metasomatic transformation and partial melting of gneisses: an example from Sri Lanka. Contrib. Mineral. Petrol. 140 (2), 212–232.
 - Pichamutu, C.S., 1960. Charnockite in the making. Nature 188, 135–136.
- **Pineau, F., Javoy, M., Behar, F., Touret, J.,** 1981. La géochimie isotopique du faciès granulite du Bamble (Norvège) et l'origine des fluides carbonés dans la croûte profonde. Bull. Minéral. 104, 630–641.
 - **Roedder, E.,** 1984. Fluid Inclusions. Reviews in Mineralogy, Vol. 12, Mineralogical Society of America. **Rogers, J.W.,** 1996. A history of continents in the past three billion years. J. Geol. 104, 91–107.
- **Rudnick, R.L., Fountain, D.M.,** 1995. Nature and composition of the continental crust: a lower crustal perspective. Rev. Geophys. 33, 267–309.
- **Rykart, R.,** 1995. Quarz-Monographie: Die Eigenheiten von Bergkristall, Rauchquartz, Amethyst und Anderen Varietäten. Ott Verlag, Thun.
- **Safonov, O.G., Kamenetsky, V.S., Perchuk, L.L.,** 2010. Links between carbonatite and kimberlite melts in chloride–carbonate–silicate systems: experiments and application to natural assemblages. J. Petrol. 52, 1307–1331.

- **Santosh, M., Tsunogae, T.,** 2003. Extremely high density pure CO₂ fluid inclusions in a garnet granulite from southern India. J. Geol. 111, 1–16.
- **Santosh, M., Kusky, T., Wang, L.,** 2011. Supercontinent cycles, extreme metamorphic processes, and changing fluid regimes. Int. Geol. Rev. 53, 1403–1423.
- Santosh, M., Tsunogae, T., Yoshikura, S.I., 2004. Ultrahigh density carbonic fluids in ultrahigh temperature metamorphism. J. Mineral. Petrol. Sci. 99, 164–179.
- Santosh, M., Harris, N.B.W., Jackson, D.H., Mattey, D.P., 1990. Dehydration and incipient charnockite formation: a phase equilibria and fluid inclusion study from South India. J. Geol. 96, 915–926.
- Santosh, M., Sajeev, K., Li, J.H., 2006. Extreme crustal metamorphism during Columbia supercontinent assembly: evidence from the North China Craton. Gondwana Res. 10, 256–66.
- **Sharp, Z.D., Barnes, J.D.,** 2004. Water-soluble chlorides in massive seafloor serpentinites: a source of chloride in subduction zones. Earth Planet. Sci. Lett. 226, 243–254.
- **Simakov**, S.K., 2003. Physicochemical Conditions of Formation of Diamondiferous Eclogite Parageneses in Upper-Mantle and Crustal Rocks [in Russian]. SVNTs DVO RAN, Magadan.
- Stern, R. A., Bleeker, W., 1998. Age of the world's oldest rocks refined using Canada's SHRIMP: The Acasta Gneiss Complex, Northwest Territories, Canada. Geosci. Can. 25 (1), 27–31.
 - Thompson, A.B., 1983. Fluid-absent metamorphism. J. Geol. Soc. 140, 533–547.
- **Touret, J.L.R.,** 1985. Fluid regime in Southern Norway: the record of fluid inclusions, in: Tobi, A.C., Touret, J.L.R. (Eds.), The Deep Proterozoic Crust in the North Atlantic Provinces. Reidel, Dordrecht, pp. 517–549. **Touret, J.L.R.,** 2001. Fluids in metamorphic rocks. Lithos 55, 1–25.
- **Touret, J.L.R., Huizenga, J.M.,** 2011. Fluids in granulites, in: Van Reenen, D.D., Kramers, J.D., Mc-Court, S., Perchuk, L.L. (Eds.), Origin and Evolution of Precambrian High-Grade Terranes, with Special Emphasis on the Limpopo Complex of South Africa. Geol. Soc. Am. Mem. 207, pp. 25–38.
- **Touret, J.L.R., Huizenga, J.M.,** 2012a. Charnockite microstructures: from magmatic to metamorphic. Geosci. Front. 3, 745–753.
- **Touret, J.L.R., Huizenga, J.M.,** 2012b. Fluid-assisted granulite metamorphism: a continental journey. Gondwana Res. 21, 224–235.
- **Touret, J.L.R., Nijland, T.G.,** 2013. Prograde, peak and retrograde metamorphic fluids and associated metasomatism in upper amphibolite to granulite facies transition zones, in: Harlov, D.E., Austrheim, H. (Eds.), Metasomatism and the Chemical Transformation of Rocks. Lecture Notes in Earth System Science. Springer-Verlag, Berlin, Heidelberg, pp. 411–465.
- **Touret J.L.R., Santosh, H.M., Huizenga, J.M.,** 2016. High-temperature granulites and supercontinents. Geosci. Front. 7, 101–113.
- Touret, J.L.R., Newton, R.C., Cuney, M. 2019a. Incipient charnockites from southern India: The role of brines. Geosci. Front. 10, 1789–1801.
- **Touret, J.L.R., Huizenga, J.M., Kehelpannala, K.V.W., Piccoli, F.,** 2019b. Vein-type graphite deposits in Sri Lanka: the ultimate fate of granulite fluids. Chem. Geol. 508, 167–181.
- **Tsunogae, T., Santosh, M., Osana, Y. Owada, M. Toyoshima, T., Hokada, T.,** 2002. Very high density carbonic fluid inclusions in sapphirine-bearing granulites from Tonagh Island in the Archean Napier Complex, East Antarctica: implications for CO_2 infiltration during ultrahigh-temperature (T > 1110 °C) metamorphism. Contrib. Mineral. Petrol. 143 (3), 279–299.
 - Van den Kerkhof A., Thiéry, R., 2001. Carbonic inclusions. Lithos 55, 49–68.
- **Veevers, J.J.,** 2007. Pan-Gondwanaland post-collisional extension marked by 650–500 Ma alkaline rocks and carbonatites and related detrital zircons: A review. Earth Sci. Rev. 83, 1–47.
- **Watson, E.B., Brenan J.M.,** 1987. Fluids in the lithosphere, I. Experimentally determined wetting characteristics of CO₂–H₂O fluids and their implications for fluid transport, host rock physical properties, and fluid inclusion formation. Earth Planet. Sci. Lett. 85, 497–515.
- **Weerakoon, M.W.K., Miyazaki, T., Shuto, K., Kagarni, H.** 2001. Rb-Sr and Sm-Nd geochronology of the Eppawala rock and carbonatite, Wanni Complex, Sri-Lanka. Gondwana Res. 4–3, 409–420.
- Weiss, Y., McNeill, J., Pearson, D.G., Nowell, G.M., Ottley, C.J., 2015. Highly saline fluids from a subducting slab as the source for fluid-rich diamonds. Nature 524, 339–342.
- Wickham, S.M., Janardhan, A.S., Stern, R.J., 1994. Regional carbonate alteration of the crust by mantle-derived magmatic fluids, Tamil Nadu, South India. J. Geol. 102, 379–398.
- Zedgenizov, D.A., Ragozin, A.L., Shatsky, V.S., Griffin, W.L., 2018. Diamond formation during metasomatism of mantle eclogite by chloride-carbonate melt. Contrib. Mineral. Petrol. 173 (10), Article 84.