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Editorial

Fluid composition and propagation in the deep crust: Case studies from the Limpopo Complex, South Africa

The important role of fluids in regional metamorphism has been highlighted by numerous geophysical, geochemical, and petrological studies aimed at understanding their key role in crustal evolution (e.g. Le Fort et al., 1987; Vrolijk, 1987; Koons and Craw, 1991; Hollister, 1992; Koons et al., 1998; Jamtveit and Yardley, 1997; Yardley, 2009; Jamtveit and Austrheim, 2010; Newton and Manning, 2010; Harlov and Austrheim, 2013; Touret and Huizenga, 2012; Aranovich et al., 2013; Harlov, 2014; Yardley and Bodnar, 2014). These studies have established the prominence of large-scale geodynamic processes as important sources of externally derived fluids, and the role of fluids in various tectonic processes. The contentious issue of fluids in crustal rocks has also been discussed in relation to metasomatism in the evolution and stabilization of the Earth's continental crust (Harlov and Austrheim, 2013; Harlov, 2014; Yardley and Bodnar, 2014).

Although many researchers concur that fluid infiltration plays an important role in the evolution of the Earth's continental crust, the composition and role of such fluids in the genesis of granulite facies terranes remains a contentious issue, with different schools of thought arguing strongly in favor of either a "fluid-absent" deep crust (e.g. Stevens, 1997; Yardley and Valley, 1997; Rigby and Droop, 2011) or a "fluid-present" deep crust (e.g. Hyndman and Shearer, 1989; Newton, 1989; Newton et al., 1998; Newton and Manning, 2010; Putnis and Austrheim, 2010; Harlov, 2012, 2014; Harlov and Austrheim, 2013; Touret and Huizenga, 2011, 2012; Aranovich et al., 2013; Safonov and Aranovich, 2014).

The granulite-fluid controversy was initially heavily influenced (Newton et al., 1980) by the earlier discovery (Touret, 1971) of dense CO_2-rich fluid inclusions in granulites, along with the later discovery of concentrated brine inclusions in quartz (e.g. Touret, 1985, Crawford and Hollister, 1986). The results of recent experimental studies (e.g. Newton and Manning, 2010; Aranovich et al., 2013) have emphasized the importance of (Na,K)Cl brines coexisting with an immiscible CO_2-rich fluid during the evolution of high-grade metamorphic terranes. In fact, experimental studies at high P and T have shown that the low wetting angle of brines along mineral grain boundaries, in contrast to CO_2, is much more efficient in promoting mineral reactions as well as enhancing mass transport in the mid to deep parts of the crust (e.g. Watson and Brenan, 1987; Newton and Manning, 2010). Concentrated brines not only provide a low H_2O activity fluid, which is necessary for the high-grade, solid state conversion of hydrous minerals such as biotite and amphibole to anhydrous ortho- and clinopyroxenes, but also have a high solubility for silicate, phosphate, and oxide minerals, which makes them effective agents of mass transfer (Newton and Manning, 2010).

The source of the H_2O responsible for mid-crustal migmatization and granite production during high-grade metamorphism is likewise the subject of continuing debate in the literature (e.g. Aranovich et al., 2013). This debate concerns the query of fluid-absent or dehydration melting, where the major source of H_2O is from the micas and amphibole (e.g. Thompson, 1983, 2001; Waters, 1988; Stevens and Van Reenen, 1992; Stevens and Clemens, 1993; Stevens, 1997; Nair and Chacko, 2002; Rigby and Droop, 2011), versus fluid-present or rehydration melting where H_2O-bearing fluids infiltrate the granulites from an external source (e.g. Newton and Manning, 2010; Touret and Huizenga, 2011, 2012; Harlov, 2012; Aranovich et al., 2013). A recent study by Aranovich et al. (2013) has shown that there is a large range in P-T space where subsolidus deep crustal metasomatism may take place at low H_2O activities due to infiltrating brine-rich fluids. Brine-fluxed anatexis also offers a viable explanation for the origin of large volumes of leucocratic anatectic granitoids that always post-date the main metamorphic and deformational events within high-grade gneiss terranes. On the other hand, experimental melting studies (Johannes and Holtz, 1991) have shown that only small amounts of granitic liquid can be obtained by dehydration melting at temperatures lower than 900 °C. The important conclusion from recent mineral solubility studies involving (Na,K)Cl brines at high P-T is that, regardless of their origin, these brines were not only important agents during metasomatism and mass transfer in granulite-facies terranes, but might also have been responsible for mid-crustal migmatites and large-scale anatexis (e.g. Newton and Manning, 2010; Aranovich et al., 2013).

Compelling evidence in the Southern Marginal Zone (SMZ) of the Limpopo Complex for regional and channeled fluid-rock interaction under granulite-facies conditions provided the motivation for bringing together experts in related areas to report their recent studies that directly relate to the role and composition of fluids in the evolution of Precambrian granulite-facies terranes. For this purpose, the special issue of *Precambrian Research* is organized into three sections:

The *first three papers* focus on new experimental data that pertain to different aspects on the role and composition of metamorphic fluids during high-grade metamorphism and anatexis. *The following two papers* revisit the interesting topic of

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fluid inclusions in incipient charnockite formation at different type localities that provide direct evidence for high-temperature fluid-rock interaction. *The final six papers* focus on the SMZ of the Limpopo Complex as a classical example of a Precambrian granulite-facies field laboratory within which virtually all aspects of high-temperature fluid-rock interaction, including the source of these fluids, brine-fluxed anatexis, regional high-temperature rehydration of granulite, and shear zone-hosted high-temperature metasomatic alteration involving formation of gold mineralization, can be demonstrated in the field and studied in the laboratory (e.g. Van Reenen, 1986; Van Reenen et al., 1992, 1994, 2011; Roering et al., 1995; Smit and Van Reenen, 1997; Huizenga et al., 2014). These studies determine that proposals for pervasive and channeled fluid flow during granulite facies metamorphism should be contingent with three important conditions: (i) strong field evidence, supported by laboratory studies, for high temperature fluid-rock interaction on both the regional and local (shear zone-hosted) scales that necessitates a persistent source of infiltrating fluids derived at depth below the metamorphic complex, (ii) evidence for a tectonic plumbing system and related mechanisms (i.e. shear deformation and hydraulic fracturing) to facilitate fluid transport into overriding hot, ductile granulite, and (iii) a persistent deep-seated source for infiltrating fluids. Such preconditions require a clear understanding of the geological control of the rock mass undergoing metamorphism and thus of successfully integrating tectonic/mechanical and metamorphic processes as the basis to study deep crustal fluid-rock interaction.

In the following sections, we summarize the highlights of the contributions in this special issue.

In the opening article, Manning and Aranovich (in this issue) deal with the thermodynamic, petrological, and geochemical effects of highly saline fluids (brines) at high *P-T* conditions. Experimental data show that the thermodynamic mixing properties of brines differ greatly from those of water-non-polar gas mixtures, causing contrasting melting behavior and unmixing of ternary H_2O-CO_2-salt fluids. New experiments on granitoid-brine equilibria furthermore indicate that alkali consumption by the melt produces HCl-rich magmatic fluids. Mineral solubility depends strongly on salt concentration in the coexisting fluid, and, with the exception of quartz, are higher in a brine than in pure H_2O at a given pressure and temperature. Partitioning of REE between rock forming minerals and brines, and between felsic melts and brines, strongly differs from that between minerals, melts, and water (±non-polar gas) indicating that brines are effective metasomatic agents for REE.

Newton et al. (in this issue) provide insight into the significance of fluid inclusions in minerals from incipient charnockites characteristic of granulite facies transitions in several Precambrian gneiss terranes. They show that NaCl-dominated brine inclusions, such as those in Kabbal–Satnur–Halagur area of the Archean Shield of southern India, have salt concentrations in agreement with the stabilization of orthopyroxene relative to calcic amphibole and biotite, while closely associated CO_2-rich inclusions can have substantial H_2O contents at incipient granulite-facies conditions (700–750 °C and 5–7 kbar). This strongly suggests that CO_2-H_2O fluids might have been in equilibrium with the associated brines as immiscible metamorphic fluids during incipient granulite formation. Fluid inclusion evidence is in accordance with the data accumulated from experimental petrology and thermodynamic calculations based on detailed studies of natural occurrences that the H_2O activity in the area of regional amphibolite–granulite boundaries in high-grade Precambrian gneiss terranes is considerably higher (0.4–0.6) than previously thought (0.15–0.3). The authors conclude that the important role of fluids in granulite-facies metamorphism of the deep crust has been greatly under-emphasized compared to the role of fluid-deficient partial melting.

Aranovich et al. (in this issue) discuss new experimental data pertaining to the concept of dehydration melting that has guided granite-granulite relations for more than half a century. In this model, granitic magmas triggered by dehydration melting at depth, are emplaced at shallow levels leaving behind a more mafic, volatile poor residue that is depleted in H_2O, alkalis, and heat-producing elements. This paper shows that the preferred model of granite genesis does not adequately explain important aspects of granite formation. For example, the heat sources responsible for melting large amounts of the lower crust require heat and mass input from mantle-derived mafic magmas. At the same time the H_2O resident in micas and amphibole is insufficient to account for large volumes of granitic rocks. This is seen in migmatitic complexes, which reveal many features that cannot be satisfied by a simple dehydration-melting model. Lastly, depletion of large-ion lithophile elements in granulites cannot be explained by dehydration melting. The authors show that CO_2 and Cl are important agents in deep crustal metamorphism and partial melting. Since CO_2 is highly insoluble in granitic liquids, and Cl is largely expelled during granite crystallization, granite generation naturally leads to production or separation of a metasomatic agent with low water activity that coexists with granulite assemblages. Such a fluid could be capable of dehydration, alkali exchange, and LILE extraction to explain chemical processes during granulite-facies metamorphism that are not readily explained by dehydration melting.

Newton and Tsunogae (in this issue) employ mass balance calculations for charnockites and host hydrous gneisses from two type localities in southern India. They conclude that metasomatism at the Kabbal-type locality at Karnataka is related to hornblende breakdown and production of orthopyroxene in orthogneiss accompanied by loss of FeO, CaO, and H_2O. At the Ponmudi-type locality in Kerala, the breakdown of biotite in paragneiss is related to the loss of FeO and MgO, and the increase of SiO_2 and alkalis. Alteration at both type localities also occurred at mid-crustal levels of 5–7 kbar and 700–750 °C. Based on fluid inclusion evidence, the authors furthermore argue that charnockitic alteration and formation of orthopyroxene-bearing anhydrous assemblages at both type localities are open system processes caused by the infiltration of coeval CO_2-rich and ultrasaline fluids. Fluid/rock ratios for both localities were small (about 10 mass percent of the rocks) and the fluids largely rock-governed. Carbon isotopes from fluid inclusions seem to indicate an igneous or primitive mantle source for both localities, but ultrasaline fluids are more plausibly explained by metasedimentary sources.

Harlov et al. (in this issue) lay out a synopsis regarding the general characteristics of localized orthopyroxene-bearing dehydration zones in granitoid rocks. This includes a relatively complete bibliography on localized dehydration zones in granitoid rocks in general starting with the earliest observed localities. Their work specifically focuses on two localized dehydration zones associated with the magmatic Varberg charnockite, SW Sweden, which are probably related to the emplacement of the charnockite. This includes a complete petrographic, mineral chemistry, and fluid inclusion study along with estimations of the pressure and temperature of formation. These results are then tied into the formation of the neighboring Varberg charnockite. Speculations are then made regarding the fluids involved in the formation of the two charnockite patches and their origins and transport from the Varberg charnockite. These fluids most likely consisted of a CO_2-rich fluid co-existing with a $CaCl_2$-dominated brine. The paper finishes with a general discussion regarding what the next step is regarding further research in the area – specially a regional survey of all charnockite patches within a 10–15 km radius of the Varberg charnockite followed by a detailed petrographic, mineralogical, fluid inclusion, and isotopic study of three to four of the best patches coupled with high precision dating (zircon, garnet etc.) of their formation and relating these dates with that for the emplacement of the Varberg charnockite.

Van Reenen et al. (in this issue) document compelling field evidence, supported by petrological, fluid inclusion, and O-isotopic fractionation data, that H_2O-CO_2-salt fluids with greatly reduced water activity infiltrated into and interacted with a cooling granulite resulting in a diversity of high-temperature fluid-assisted phenomena. They argue that infiltrating fluids triggered the main pulse of anatexis that resulted in the production of large volumes of largely undeformed, granodioritic–tonalic melts that intruded and interacted with an already migmatized metapelitic granulite over a *P-T* interval that ranged from $P > 7.5$ kbar, $T > 900$ °C to $P = \sim 6$ kbar, $T = \sim 600$ °C. Similar fluids are also shown to have initiated shear zone-hosted metasomatic alteration of quartzo-feldspathic gneisses at high temperatures ($T = \sim 900$ °C). At lower *P-T* conditions ($P = \sim 6$ kbar, $T = \sim 600$ °C), the infiltrating H_2O-CO_2-salt fluids established a regional retrograde (Orthopyroxene-out, Anthophyllite-in) isograd expressed within metapelitic granulite, and an associated zone of retrogressed, rehydrated granulite that occupies 4500 km^2 of crust located in the hanging wall section of the north-dipping Hout River Shear Zone that bounds the SMZ in the south. Shear zone-hosted, metasomatic alteration of quartzo-feldspathic gneisses and associated gold deposits that characterizes the rehydration zone also formed at *P-T* conditions of $P = \sim 6$ kbar, $T = \sim 600$ °C. The source (underthrusted greenstone belt material) and a mechanism for controlling persistent fluid influx is the topic of the next paper.

Smit et al. (in this issue) discuss field, structural, petrological, geochronological, and geophysical data that support a fluid-assisted tectonic mechanism (termed a conveyor belt mechanism) that managed persistent influx of H_2O-CO_2-salt fluids derived from devolatilization of underthrusted greenstone material as the result of the near-isobaric ($P = \sim 6$ kbar) emplacement (horizontal channeling) of a hot SMZ granulite nappe. This 18 km thick nappe, consisting of a basal hot granulite tongue (SMZ) overlain by stiff upper crustal material, was emplaced onto the underthrusted granite-greenstone terrane of the northern Kaapvaal Craton along a major thrust plane (the Hout River Shear Zone) in the interval ~2.69–2.62 Ga. Near vertical channeling of melt-weakened (migmatitic) crust in the Central Zone of the Limpopo Complex was the driving force that pushed horizontal channeling of the hot SMZ granulite nappe, with its steep structures, over a distance of more than 60 km southwards onto the adjacent granite-greenstone craton. Near vertical channeling is indicated by the fact that all linear elements, associated with major sheath folds and associated granitic diapirs in the Central Zone, plunge at steep angles to the SW, suggesting that the Central Zone was forced bodily upwards in the direction defined by the orientation of these structural elements. Deep crustal fundamental geological processes that could be observed and studied in the Limpopo Complex are shown to have been strongly dependent on the role of infiltrating fluids/melts. Fluid-assisted processes not only provided the necessary mechanical weakening and buoyancy that allowed vertical channeling and horizontal displacement of deep seated granulite-facies material onto the cool granite-greenstone craton, but also controlled brine-fluxed anatexis at high temperatures and the pervasive rehydration of the cooling granulite. This included shear zone-hosted metasomatic alteration and the formation of quartz vein-hosted gold mineralization.

Kramers et al. (2014) present detailed geology and geochronological data of a section through the Giyani greenstone belt at the northern edge of the Kaapvaal Craton. These data show that the south-eastern domain of the greenstone belt retains a memory of a metamorphic episode at about 2850 Ma, while at the northwestern edge this memory has been erased by metamorphism at about 2720 Ma, associated with overthrusting of the Southern Marginal Zone (SMZ) of the Limpopo Complex. The authors compare lithologies as well as new and published geochronological data of the Giyani, Pietersburg and Rhenosterkoppies greenstone belts, and argue the case for an orogeny at ca. 2850 Ma in which both the Pietersburg and Giyani greenstone belts underwent north-verging thrusting and metamorphism of presently exposed rock units associated with amphibolite facies conditions. This contribution does not recognize a link between north-verging thrusting in the Pietersburg and Giyani greenstone belts on the one hand, and the high grade metamorphism of the SMZ and associated tectonics along the Hout River Shear Zone (HRSZ), that bounds the SMZ in the south, on the other. 40Ar/39Ar amphibole ages along the HRSZ are shown to be mostly 60–80 Ma younger than the peak metamorphism in the Southern Marginal Zone, and thermal modeling indicates that these younger ages could reflect a delayed response to overthrusting: supracrustal units of the Kaapvaal Craton, buried at mid- to lower crustal level, would have heated up slowly due to the low regional content of radioactive heat producing elements, leading to fluid release over a long period.

Safonov et al. (in this issue) record evidence from the SMZ for the interaction between a fluid-rich, trondhjemitic magma, which originated from fluid-fluxed partial melting of metabasic material at the base of the granulite complex or the top of the underthrusted greenstone blocks, with peraluminous (orthopyroxene–cordierite–biotite–plagioclase–quartz) metapelitic granulites from a high-grade shear zone (the Petronella shear zone). Using detailed petrological data from both the metapelites and trondhjemites, which include fluid inclusions, conventional thermobarometry (TWQ method), and the Gibbs energy minimization method (pseudosections by PERPLE_X), the authors conclude that the hot ($T = \sim 1000$ °C) trondhjemitic magma transferred heat and transported large volumes of exotic aqueous-carbonic-salt fluids from the lower (28–24 km) to the middle (18–20 km) crust during exhumation of the SMZ at 2.667 ± 0.9 Ga. The hot magma, which cooled and solidified isobarically from $T = \sim 900$ °C to $T = \sim 600$ °C, expelled low water activity fluids ($a_{H_2O} < 0.3$) that participated in rehydration of SMZ granulites. The authors also discuss evidence that the hot magma triggered local decompression melting of the host metapelite. This study shows that large volumes of granitic magma in the SMZ did not originate through dehydration melting of mica-bearing metapelites, but that such melts are the first direct evidence that hot "foreign" (i.e. derived from a deep-seated source) granitoid magmas played a critical role in the exhumation and emplacement of SMZ granulites onto the adjacent Kaapvaal granite-greenstone craton.

Koizumi et al. (in this issue) provide new petrological data for a partially hydrated pelitic granulite from the retrograde (Orthopyroxene-out) isograd developed in the SMZ of the Limpopo Complex in South Africa, and discuss the *P-T*-fluid evolution based on mineral equilibrium modeling in the NCKFMASH system. Their results demonstrate that the first rehydration event that formed anthophyllite replacing orthopyroxene took place at 630–730 °C and $M(H_2O) = 3.0$–3.5 mol%. This was followed by further increase in $M(H_2O)$ (4.5–6.0 mol%) and cooling to <630 °C that triggered a second rehydration event, which formed kyanite/sillimanite + orthoamphibole + quartz and garnet + kyanite/sillimanite + quartz assemblages after cordierite. They concluded that such an increase in the molar H_2O content and formation of rehydrated granulites during the exhumation stage might be related to the influx of an H_2O-bearing fluid along the Hout River Shear Zone that marks the major crustal break between the Southern Marginal Zone and the adjacent Kaapvaal Craton.

Tsunogae and Van Reenen (in this issue) examine fluid-rock interaction and high-temperature metasomatism recorded in a tonalitic charnockite and metasomatized equivalents from the granulite zone of the SMZ of the Limpopo Complex, South

Africa. Detailed petrological studies demonstrate that bulk K_2O increases, and CaO, MgO, FeO, and TiO_2 decrease continuously from the tonalitic charnockite (plagioclase + quartz + K-feldspar + orthopyroxene) to the intensely metasomatized rock (mesoperthite/antiperthite + quartz + garnet + sillimanite) derived from the charnockite. Based on phase equilibrium modeling in the system NCKFMASH and supported by fluid inclusion microthermometry, and ternary-feldspar geothermometry, the authors conclude that addition of K to, and removal of Ca and Mg from, the tonalitic charnockite gave rise to the metasomatized mineral assemblages. Metasomatism was caused by infiltration of high-salinity aqueous fluids along the high-temperature Petronella Shear Zone during granulite-facies metamorphism, probably derived from devolatilization of low-grade greenstone materials from the Kaapvaal Craton as the result of the thrusting of hot granulite over cool granite-greenstone belts.

The 11 papers presented in this special issue of *Precambrian Research* represent the results of recent studies undertaken by different experts with the singular purpose to emphasize the important role of H_2O-CO_2-salt fluids of greatly reduced water activity in the evolution of Precambrian granulite-facies terranes, including gold mineralization, with special emphasis on the Limpopo Complex of southern Africa. We hope that these contributions will provide useful information and encouragement to researchers to continue to study the unique outcrops exposed in the Limpopo Complex.

The guest editors wish to thank the Editor of Precambrian Research (Prof Guochun Zhao) for overseeing our editorial work for this special issue and for his encouragement. A project of this nature and dealing with the contentious issue of the role of fluids during granulite metamorphism could never have been successfully concluded without the support of the different experts who have agreed to contribute their time and expertise to prepare papers for this special issue. This included a trip to South Africa in June–July 2013 to participate in a fluid seminar that was held in the Geology Department, University of Johannesburg and to attend a Limpopo field workshop during which all participants were introduced to different aspects of high-temperature fluid-rock interaction as expressed on outcrop. A special word of appreciation to Prof Robert C Newton who at 80 years of age was not only prepared to travel from UCLA to Johannesburg in 2013, but also for his unmatched enthusiasm and special insights into the study of fluid-related phenomena in the outcrops during the field workshop. The guest editors would also like to thank the reviewers who added value to the special issue by providing critical reviews of the different papers in a timely manner.

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Dirk van Reenen*
Department of Geology, University of Johannesburg, P.O. Box 524, Auckland Park 2006, South Africa

M. Santosh
School of Earth Sciences and Resources, China University of Geosciences Beijing, No. 29 Xueyuan Road, Haidian District, Beijing 100083, China

Leonid Aranovich
Institute of Geology of Ore Deposits, Petrography, Mineralogy and Geochemistry (IGEM), Russian Academy of Sciences, Staromonentny 35, 119017 Moscow, Russia

Daniel Harlov
GeoForschungs Zentrum, Telegrafenberg, D-14473 Potsdam, Federal Republic of Germany

Oleg Safonov
Institute of Experimental Mineralogy (IEM), Russian Academy of Science, Academician Ossipian Street 4, 142432 Chernogolovka, Moscow Area, Russia

* Corresponding author. Tel.: +27 11 559 3933; fax: +27 11 489 2191.
E-mail address: dirkvr@uj.ac.za (D. van Reenen)

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