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Fluid-rock interaction during high-grade metamorphism: Instructive examples from the Southern Marginal Zone of the Limpopo Complex, South Africa

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ABSTRACT

The Southern Marginal Zone of the Limpopo Complex documents strong evidence that CO_2-rich ($X^{fluid}_{CO_2}$ = 0.7–0.9, $X^{fluid}_{H_2O}$ = 0.1–0.3) and brine fluids of greatly reduced water activity interacted with cooling metapelitic granulite during the thrust-controlled emplacement at 2.69–2.62 Ga onto the granite-greenstone terrain of the northern Kaapvaal Craton. Interaction of cooling metapelitic granulite with CO_2-rich fluids at T < 600–630 °C and P < ~6 kbar is recognized by the presence of a regional retrograde Opx-out/Ath-in isograd and an associated zone of retrograde hydrated granulites that occupies ~4500 km^2 of retrogressed crust located in the hanging wall section of the shallow north-dipping Hout River Shear Zone that bounds the Southern Marginal Zone in the south. On the other hand, brine fluids are considered to have triggered the main pulse of anatexis that resulted in production of large volumes of granodioritic-trondhjemitic melts that intruded and started to interact with metapelitic granulite in the deep crust at T > ~900 °C, P > ~7.5 kbar. Interaction of hot melt with metapelitic granulite continued until final emplacement in the middle crust (P = ~6 kbar, T = ~630 °C). Brine fluids also initiated shear zone-hosted metasomatism of quartzo-feldspathic gneisses at T between ~600 and ~900 °C and amphibolite-facies lode-gold mineralization. Available data implicate devolatilization of underthrusted greenstone material as the dominant deep crustal source for infiltrating CO_2-rich and brine fluids.
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1. Introduction

Interaction of granulite-facies rocks with externally derived fluids remains a contentious issue among metamorphic petrologists (e.g., Rigby and Droop, 2011; Touret and Huizenga, 2011, 2012; Yardley, 2013; Newton et al., 2014). This is true because most researchers agree that, in most cases, rocks undergoing high-grade metamorphism have a very low permeability and contain only small amounts of metamorphic fluids at near-lithostatic pressures during peak metamorphism (Yardley and Valley, 1997; Yardley, 2009, 2013). When the metamorphic rocks cool, they hardly back react with fluids to form a lower temperature mineral assemblage (retrogression) because fluids produced during prograde metamorphism escape and thus are no longer available.

However, in situ geophysical and geochemical studies carried out in active (modern) regional metamorphic terrains have shown that metamorphic fluids are an integral part of the tectonic system (e.g., Koons and Craw, 1991; Koons et al., 1998). Early studies done in the Himalaya, for instance, established the importance of aqueous fluids being driven out during metamorphism of sediments under the Main Central Thrust when overridden by the hot hanging wall of the Tibetan slab (Le Fort et al., 1987). These fluids triggered anatexis in the hanging wall, which resulted in the generation of leucocratic granite, which intruded into the upper levels of the Tibetan slab (Le Fort et al., 1987).

Different workers have also provided mineralogical and isotopic evidence for the existence of high time integrated fluid fluxes and fluid flow directions in many thermal aureoles and regional metamorphic belts (Rubenach, 2013). For example, Markl and Bucher (1998) and Gleeson et al. (2003) have documented evidence for amphibolitization of lower crustal granulites through brine influx, while others (Van Reenen, 1986; Ferry and Dipple, 1991; Ferry, 1994; Hoernes et al., 1995; Putnis and Austrheim, 2010) described evidence for infiltration-driven regional metamorphism.

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The reasons why researchers often doubt whether infiltration metasomatism is widespread during regional metamorphism appears to arise from concerns about the availability and source of fluids (e.g., Yardley, 2013). However, recent experimental data (e.g., Newton and Manning, 2010; Aranovich et al., 2013, 2014; Safonov et al., 2014) clearly show that, regardless of their origin, deep crustal fluids and specifically brines, must be important agents of metasomatism and mass transfer wherever such processes are recognized in the field or hand specimen (e.g., Harlov, 2012).

Wet versus dry middle to deep crust furthermore implicates the issue of fluid-absent (dehydration melting) versus fluid-assisted mechanisms of crustal anatexis. Proponents of the fluid-absent origin of granulite (e.g., Rigby and Droop, 2011) argue that granulite (and crustal anatexis) were formed in the mid to deep crust in the absence of a free fluid phase via high temperature metamorphism (dehydration melting/anatexis; e.g., Fyfe, 1973; Thompson, 1983; Clemens and Vielzeuf, 1987; Waters, 1988; Stevens and Clemens, 1993; Yardley and Valley, 1997). A strong argument usually used in favor of the fluid-absent model is that important features of granulite facies rocks support a dehydration melting origin, including the high-temperature nature of a number of granulite assemblages as well as the observation that many granulite-facies rocks preserve the protolith O-isotope signature, thus excluding involvement of a pervasive fluid flow in their origin (e.g., Valley, 1986; Vennemann and Smith, 1992).

On the other hand, proponents of the fluid-assisted model for granulite formation and for associated anatexis have shown that temperature estimates ($T < 850\,^{\circ}C$) obtained by geothermobarometry from numerous "normal" granulite facies rocks are in fact lower than the values required for the onset of fluid-absent dehydration melting (Aranovich et al., 1987, 2014; Newton, 1989; Johannes and Holtz, 1991; Newton et al., 1998, 2014; Nair and Chacko, 2002; Safonov et al., 2014). They ascribe the apparent undisturbed protolith O-isotope signatures of granulite to low fluid/rock ratios (e.g., Hoernes et al., 1995). The fluid-assisted model is also supported by the common occurrence of CO_2, brine and mixed CO_2-brine fluid inclusions (e.g., Touret and Huizenga, 2011, 2012; Aranovich et al., 2013; Newton et al., 2014; Safonov et al., 2014). Finally, the presences of large volumes of anatectic granitic melt in high-grade metamorphic terrains argue in favor of a mechanism of "water-fluxed melting" (e.g., Sawyer et al., 2011).

High-temperature metasomatism involving chemical and mineralogical changes are undisputable features of high-grade shear zones in granulite-facies terrains and indicates that they are sites of focused fluid flow (Skelton et al., 1995; Smit and Van Reenen, 1997; Glassly et al., 2010; Touret and Huizenga, 2012). Archaean lode-gold mineralization at different *P–T* conditions in high-grade gneiss terranes is clearly linked to metasomatism of precursor rocks by infiltrating fluids (e.g., Philips and Groves, 1983; Philips, 1985; Smith et al., 1984; Van Reenen et al., 1994; Groves et al., 2003). Such fluids involve H_2O-$CO_2 \pm CH_4$ fluids (e.g., Mikucki, 1998), with a maximum mole CO_2 mole fraction ($X^{fluid}_{CO_2}$) of ~0.25 derived from crystallizing magma at depth, metamorphic devolatilization reactions, or the mantle as is indicated by stable isotope data (Smith, 1986; Golding et al., 1987; Kerrich, 1987). These fluids will, depending on their prevailing temperatures and fluid-rock ratios, either metamorphose or metasomatose (and possibly mineralize) the rocks through which they pass (e.g., Vrolijk, 1987; Sisson et al., 1989; Moore et al., 1987; Hyndman, 1988; Hyndman and Shearer, 1989).

Given the complexity of the issues involved, it is imperative that strong field evidence in support of regional and local scale fluid-rock interaction should ideally be interpreted within the context of the overall geological evolution of the rocks in which such processes occur (Yardley, 2013; Huizenga et al., 2014; Smit et al., 2014). This paper contributes to specific aspects of this contentious issue by providing, for the first time, a comprehensive record compiled from published (Van Reenen, 1986; Van Reenen and Hollister, 1988; Baker et al., 1992; Hoernes and Van Reenen, 1992; Van Reenen et al., 1994, 2011; Van Schalkwyk and Van Reenen, 1992; Hoernes et al., 1995; Smit and Van Reenen, 1997; Huizenga et al., 2014) and unpublished data (Du Toit, 1994; Mokgatlha, 1995; Stefan, 1997) regarding interaction of low H_2O-activity ($a^{fluid}_{H_2O}$) fluids and cooling granulites in the Southern Marginal Zone of the Limpopo Complex. Although this paper recognizes devolatilization of underthrusted greenstone material as the primary source for infiltrating fluids, we will also discuss alternative fluid sources (crystallizing granitic magma, upper mantle) that might have contributed to the overall fluid budget of the SMZ during the thrust-controlled exhumation.

Fluids that infiltrate cooling granulites of the Southern Marginal Zone is expressed by a variety of high-temperature fluid-rock interaction phenomena including pervasive retrogression of granulite, anatexis, and shear zone-hosted metasomatism of high-grade rocks including gold mineralization. High-temperature metamorphic and metasomatic phenomena have shown not to be mutually exclusive but are closely linked processes involving CO_2-rich ($X^{fluid}_{CO_2} = 0.7–0.9, X^{fluid}_{H_2O} = 0.1–0.3$) fluids and brines. Metasomatism in this context is regarded as changes in the bulk chemical composition of non-volatile components (e.g., Putnis and Austrheim, 2010; Yardley, 2013).

Finally, this paper is a general introduction to different case studies in the present issue that deal with special aspects of pervasive and channeled fluid-rock interaction in the Southern Marginal Zone (Dubinina et al., under review; Koizumi et al., 2014; Tsunogae and Van Reenen, 2014; Safonov et al., 2014). Devolatilization of underthrusted greenstone material as a specific deep crustal fluid source and a tectono-metamorphic model that controlled persistent fluid flow into overriding hot granulite during exhumation, are the focus of a separate paper (Smit et al., 2014).

2. Tectonic setting of the Southern Marginal Zone and the northern Kaapvaal Craton

The high-grade Limpopo Complex in South Africa is a late Archean ENE-WSW trending zone located between the granite-greenstone terrains of the Zimbabwe and Kaapvaal Cratons (Fig. 1). The complex is subdivided into the Northern Marginal Zone, the Central Zone, and the Southern Marginal Zone (e.g., Van Reenen et al., 2011), of which the Northern and Southern Marginal Zones comprise high-grade granitoids and greenstone belt lithologies (inset in Fig. 1) (e.g., Kreissig et al., 2001; Van Reenen et al., 2011). The Northern and Southern Marginal Zone are juxtaposed against the Zimbabwe and Kaapvaal Craton, respectively, as a result of compression-related exhumation during 2.69–2.62 Ga (Van Reenen et al., 2011). The Central Zone is separated from the marginal zones by Paleoproterozoic crustal-scale strike slip shear zones (e.g., Roering et al., 1992b).

The origin and critical role that reactive fluids have played in the Southern Marginal Zone (SMZ) can only be appreciated if this process is considered within the context of the geotectonic relationship of the SMZ with the juxtaposed northern Kaapvaal Craton (NKVC) (Figs. 1 and 2). Smit et al. (2014) provide a comprehensive overview of the complex shared evolutionary histories of these two disparate juxtaposed terrains, and only pertinent aspects that impact directly on the focus of this paper will be briefly mentioned here.

A shallow north-dipping terrain bounding structure named the Hout River Shear Zone (HRSZ) (Figs. 1 and 2) separates two distinct geological terrains that, prior to the onset of the Limpopo Orogeny (~2.72 Ga), formed part of the granite-greenstone terrain of the NKVC. The SMZ of the Limpopo Complex presently occupies

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