



Tourmaline B-isotopes as tracers of fluid sources in silicified Palaeoarchaean oceanic crust of the Mendon Formation, Barberton greenstone belt, South Africa



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ARTICLE INFO

Article history:

Received 13 May 2015

Received in revised form 7 September 2015

Accepted 4 October 2015

Available online 8 October 2015

Keywords:

Tourmaline

SIMS

Boron isotopes

Barberton greenstone belt

Mendon Formation

ABSTRACT

Pervasive silicification is a common phenomenon in Early Archaean volcano-sedimentary sequences, reflecting the interaction between Si-rich fluids and the Archaean oceanic crust. In the Palaeoarchaean Barberton greenstone belt, South Africa, the silicified zones locally contain abundant tourmaline. In order to constrain the source of fluids responsible for the alteration, six tourmaline-bearing samples from the 3.3 Ga Mendon Formation were investigated by in-situ analyses of mineral chemistry and boron isotopes. The samples comprise a silicified, finely laminated sedimentary chert and five highly altered and silicified komatiites. Electron microprobe analysis (EMPA) indicates that tourmaline is mostly dravitic, with one sample between dravite and Mg-foitite. Tourmaline has a weakly-developed patchy colour- and chemical zonation, with variations in Al, Fe and Mg composition. Some samples show strong zoning in Cr-concentration with Cr enrichment in the core, and locally the high-Cr domains are at contacts with other Cr-rich phases such as magnesiochromite and chromian muscovite (“fuchsite”).

Overall, the boron isotope composition ($\delta^{11}\text{B}$) ranges from -20.7 to $+10.2\%$. Two groups of tourmaline are distinguished based on the variation in $\delta^{11}\text{B}$ ratios within single samples: 1) those with small ($<4\%$) range in $\delta^{11}\text{B}$ values (spinifex-textured komatiites and silicified sediment), and 2) those with a large range in $\delta^{11}\text{B}$ values of up to 18% (brecciated and foliated samples). Positive boron isotope values ($+6$ to $+10\%$) are found in one spinifex-textured komatiite and are interpreted as being derived from seawater interacting with Archaean oceanic crust in shallow hydrothermal systems. The intermediate boron isotope values (-5 to -8%) found in all other samples are similar to MORB and may represent boron derived from the Archaean oceanic crust. The rare and exceptionally light boron isotope values (-10 to -21%) were found in rims of group 2 tourmaline. These values cannot be produced by fractionation alone, and point to the presence of another fluid source. The source of the light boron has yet to be identified, but possible candidates are remobilized boron either derived from granitic crust or from marine evaporites that precipitated from seawater enriched in ^{10}B . In any case, the isotopically light boron appears to have been present during seafloor hydrothermal alteration and deposition of the Mendon Formation at ca. 3.3 Ga.

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1. Introduction

Many Palaeoarchaean greenstone belts contain pervasively silicified volcano-sedimentary sequences and ubiquitous chert layers (Hofmann and Harris, 2008; van den Boorn et al., 2010). The reason for the silicification is controversial, but numerous studies regard it as the result of hydrothermal fluid circulation in shallow sub-seafloor convection cells

(Duchac and Hanor, 1987; Hanor and Duchac, 1990; Hofmann and Harris, 2008; Farber et al., 2015). These low-temperature hydrothermal systems (<150 °C) may represent Archaean analogues to the diffusive venting of low-temperature fluids on modern oceanic plateaus (Hofmann and Harris, 2008). In the Onverwacht Group of the Barberton greenstone belt in South Africa, silicification is predominant at the top of mafic to ultramafic lava flows at the contact with sedimentary chert horizons (Hofmann and Harris, 2008). The silicified zones locally contain abundant tourmaline, reflecting exceptionally high contents of dissolved boron.

Minerals of the tourmaline supergroup are stable over pressure-temperature conditions ranging from diagenesis to high-grade metamorphism. They are also resistant to alteration, which makes them useful for

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extracting information about the evolution of rocks (e.g. Henry and Dutrow, 1996, 2012; Dutrow and Henry, 2011; Van Hinsberg et al., 2011). Furthermore, boron isotopes in tourmaline can be used to trace fluid sources since boron is highly mobile in fluids and tourmaline is the main boron-bearing mineral in most rocks (Palmer and Slack, 1989; Palmer and Swihart, 1996; Marschall and Jiang, 2011; Van Hinsberg et al., 2011).

Boron isotope ratios have a large natural variability in different geological reservoirs with $\delta^{11}\text{B}$ values ranging from ~ -30 to $+60\%$ (Barth, 1993; Palmer and Slack, 1989; Van Hinsberg et al., 2011 and references therein). The $\delta^{11}\text{B}$ values are expressed as per mil deviation from the standard SRM51 boric acid: $\delta^{11}\text{B} = ({}^{11}\text{B}/{}^{10}\text{B}_{\text{sample}} \text{ divided by } {}^{11}\text{B}/{}^{10}\text{B}_{\text{SRM951}}) - 1 * 1000$, with ${}^{11}\text{B}/{}^{10}\text{B}_{\text{SRM951}} = 4.04362$ (Catanzaro et al., 1970). Modern seawater has a constant $\delta^{11}\text{B}$ value of 39.6% (Foster et al., 2010), whereas the mean continental crust has a $\delta^{11}\text{B}$ value of -10.5% , and the mean mantle value is -7% (Chaussidon and Albarède, 1992; Chaussidon and Jambon, 1994; Marschall and Jiang, 2011). The highest $\delta^{11}\text{B}$ values are found in marine brines with $\delta^{11}\text{B} > 50\%$ (Vengosh et al., 1991), whereas $\delta^{11}\text{B}$ values as low as -30% are recorded in non-marine evaporites and granite-related veins (Swihart et al., 1986; Barth, 1993).

During mobilization and transport of boron, mixing of fluids from different source rocks is possible (e.g. Palmer and Slack, 1989; Palmer and Swihart, 1996), and such mixing will affect the final boron isotope composition preserved in tourmaline reflecting the isotope composition of pertinent reservoirs and the relative amount of boron contributed. Once formed, the isotopic and chemical record is well preserved in tourmaline, as only insignificant intracrystalline diffusion occurs and tourmaline is stable during most geological events (Henry and Dutrow, 1996; Dutrow and Henry, 2011; Van Hinsberg et al., 2011).

In the Barberton greenstone belt, tourmaline is found throughout the Onverwacht Group. Its boron isotope composition was previously studied using bulk analytical methods by Byerly and Palmer (1991). This study reported boron isotope analyses from a variety of rocks, including silicified mafic and ultramafic volcanic rocks, evaporite- and stromatolite-bearing sediments, as well as spatially associated quartz veins, with an overall range of $\delta^{11}\text{B}$ values between -10.5 and 2.2% . Byerly and Palmer (1991) proposed a two-stage boron enrichment model in which evaporitic boron was hydrothermally remobilized.

In the present study we revisit the B-isotope record in Barberton tourmaline using in-situ analyses by secondary ion mass spectrometry (SIMS). In-situ data are likely to be important because the Byerly and Palmer (1991) model is based on bulk mineral separates whereas the tourmaline grains record a wide range in chemical composition (Byerly and Palmer, 1991), and may also be zoned with respect to their $\delta^{11}\text{B}$ values. We present results of in-situ chemical and B-isotope analyses of hydrothermal tourmaline from silicified zones in the 3.3 Ga Mendon Formation and use these data to provide information on the source of the boron and to identify fluid sources that might be responsible for the alteration.

2. Geological setting

The Palaeoarchaeon Barberton Supergroup of the Barberton greenstone belt consists of the predominantly volcanic Onverwacht Group and the sedimentary Fig Tree and Moodies Groups (Fig. 1). The Onverwacht Group formed between 3.54 and 3.26 Ga (Kröner et al., 1991, 1996; Byerly et al., 1996; Decker et al., 2015) and consists of ultramafic to mafic rocks and sedimentary layers that were deposited during phases of volcanic quiescence in a marine environment (Lowe and Byerly, 1999; Lowe and Byerly, 2007).

The 3.33 to 3.26 Ga Mendon Formation is the youngest unit of the Onverwacht Group and consists of at least five volcano-sedimentary cycles of interbedded komatiites, komatiitic basalts, and silicified sediments ("banded cherts") (Byerly et al., 1996; Byerly, 1999; Lowe and Byerly, 1999; Decker et al., 2015). These cyclic members are subdivided into

M1 to M5 with an addition of v for volcanic and c for chert units (Lowe and Byerly, 1999). Silicification of the komatiites is most intense near the contact to the overlying silicified sediments, and is interpreted to have occurred in shallow, low-temperature subseafloor convection cells shortly after the deposition of the Mendon Formation (Hofmann, 2005; Hofmann and Harris, 2008). Serpentinised komatiites locally underlie the silicified komatiites, and spinifex textures are preserved in both types of rock (Fig. 2e; Byerly, 1999; Duchac and Hanor, 1987; Hofmann and Harris, 2008). Field evidence, as well as oxygen isotope ratios of chert and quartz separates indicate that the silicification of the ultramafic and sedimentary sequences occurred during low-temperature ($100\text{--}150\text{ }^\circ\text{C}$) hydrothermal alteration on the Archaean seafloor (Hofmann and Harris, 2008; Abraham et al., 2011; Farber et al., 2015). Additional to silica alteration, potassium and boron metasomatism occurred resulting in the formation of locally abundant mica and tourmaline (Byerly and Palmer, 1991; Toulkeridis et al., 1998; Byerly, 1999; Hofmann and Harris, 2008).

Silicified sediments that overlie the silicified komatiites are typically banded cherts. The banded cherts contain silicified accretionary lapilli and abundant carbonaceous matter that may have a biogenic origin (Hofmann, 2005; Hofmann and Bolhar, 2007; Walsh, 1992; Walsh and Lowe, 1999; Westall et al., 2001; Westall et al., 2015). Another variety of silicified sediment is a finely laminated chert showing domical features interpreted as stromatolites (Byerly et al., 1986) or as a laminated silica crust precipitated on the seafloor as a result of impact-induced evaporation of Archaean seawater (Lowe and Byerly, 2015). The laminae are completely replaced by fine-grained tourmaline (Fig. 2a, cf. Byerly and Palmer, 1991).

Silicified komatiites have a brown-reddish colour on weathered surfaces, whereas fresh surfaces are green due to the presence of chromian muscovite (Fig. 2). Stratiform and high-angle crosscutting veins of black chert are common in both silicified sediments and komatiites (Fig. 1b; Hofmann and Harris, 2008; Lowe, 1999a; Ledevin et al., 2015). Chert veins consist of microcrystalline quartz with minor amounts of sheet silicates, carbonaceous matter and/or volcanic ash (Lowe, 1999a; Hofmann and Bolhar, 2007). Vein size varies from cm to m scale with many chert veins containing a coarse-grained interior of crystalline quartz (Farber et al., 2015). Chert-clast breccias made up of angular fragments of silicified komatiite in a matrix of either black chert or carbonate locally occur in the silicified komatiites (Fig. 2b,c). The breccias and chert veins have been interpreted to have formed as a result of hydraulic fracturing (Hofmann and Bolhar, 2007; Ledevin et al., 2015) or due to impact-related seismic events (Lowe, 2013; Lowe et al., 2014; Lowe and Byerly, 2015).

The Mendon Formation units show evidence of regional metamorphism under greenschist facies conditions, as indicated by the presence of muscovite and chlorite. Temperatures in the Onverwacht Group during peak metamorphism are estimated at $\sim 260\text{--}350\text{ }^\circ\text{C}$ (Xie et al., 1997; Tice et al., 2004; Grosch et al., 2012; Farber et al., 2015). The peak of metamorphism has been related to regional scale folding and crustal convergence during the main accretionary (D_2) event at ca. 3230–3225 Ma (de Ronde and de Wit, 1994; Dziggel et al., 2002; Kisters et al., 2003). The subsequent D_3 deformation lasted from ca. 3230 to 3100 Ma, and was characterized by ongoing NW–SE oriented crustal shortening, followed by strike-slip and transtensional tectonics (Belcher and Kisters, 2006; de Ronde and de Wit, 1994; Schoene et al., 2008). The D_3 deformation was coeval with the emplacement of a major suite of sheet-like potassic granites dated at 3140–3100 Ma (de Ronde et al., 1991; Schoene and Bowring, 2007), although this event does appear not to have affected rocks in the study area.

3. Analytical methods

3.1. Electron microprobe

The major element composition of tourmaline was determined on polished thin sections and on grain mounts, using a JEOL JXA-8900R

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