



Applications of biotite inclusion composition to zircon provenance determination

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ABSTRACT

Detrital zircons are the only confirmed surviving remnants of >4.03 Ga crust while younger detrital zircons provide a parallel record of more recent crustal evolution to that preserved in crystalline rocks. Zircons often preserve inclusions that may provide clues as to the origins of out-of-context grains in the sedimentary record. Previous studies have established that inclusions of biotite in magmatic zircon are compositionally well-matched to biotite in the source rock matrix, although a direct application to ancient detrital zircons has not been made. A number of studies have documented variations in the Fe, Mg, and Al contents of magmatic biotite from different source rocks and tectonic settings, suggesting that biotite inclusions may indeed serve as provenance indicators for detrital zircons. Consistent with earlier studies, we find that the FeO^*/MgO ratio of magmatic biotite from continental arcs, collisional, and within-plate settings varies with relative oxidation state as well as whole-rock FeO^*/MgO , while its $\text{Al}_2\text{O}_3/(\text{FeO}^* + \text{MgO})$ varies with whole-rock Al/CNK (molar $\text{Al}/(2 \cdot \text{Ca} + \text{Na} + \text{K})$). Biotite from oxidized metaluminous and reduced S-type granitoids can be readily distinguished from each other using FeO^*/MgO and $\text{Al}_2\text{O}_3/(\text{FeO}^* + \text{MgO})$, while biotite from reduced I-type and oxidized peraluminous granites may in some cases be more ambiguous. Biotite from peralkaline and reduced A-type granites are also distinguishable from all other categories by $\text{Al}_2\text{O}_3/(\text{FeO}^* + \text{MgO})$ and FeO^*/MgO , respectively. Biotite inclusions in Hadean zircons from Jack Hills, Western Australia indicate a mixture of metaluminous and reduced S-type host rocks, while inclusions in 3.6–3.8 Ga detrital zircons from the Nuvvuagittuq Supracrustal Belt indicate more oxidized peraluminous magmas. These results highlight the diversity of felsic materials on the early Earth and suggest that biotite inclusions are applicable to zircon provenance throughout the sedimentary record.

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1. Introduction

The absence of a known rock record prior to 4.03 Ga (Bowring and Williams, 1999; cf. O'Neil et al., 2008) drove development of geochemical proxies in detrital zircon to obtain insights into the planet's first few hundred million years. Various sources of isotopic and petrologic evidence have revealed that Hadean (>4.03 Ga) detrital zircons from the Jack Hills locality (Compston and Pidgeon, 1986) show evidence for sedimentary cycling in the presence of water (high $\delta^{18}\text{O}$; Mojzsis et al., 2001; Peck et al., 2001; Cavosie et al., 2004, 2006; Trail et al., 2007; Harrison et al., 2008) and origin in low-temperature granitic melts (low Ti-in-zircon crystallization temperature; Watson and Harrison, 2005; hereafter T^{zln}). At least some are derived from peraluminous granitoids due to

the ubiquity of primary igneous muscovite inclusions (Hopkins et al., 2008, 2010; Bell et al., 2015; cf. Rasmussen et al., 2011) and rare high-Al zircons (Trail et al., 2017) while many resemble I-type zircons in either trace chemistry (Burnham and Berry, 2017; Trail et al., 2017) or inclusion assemblage (Hopkins et al., 2008, 2010). While the extensive efforts in cataloguing the geochemistry of the zircons has revealed a very different early Earth than the arid and inhospitable world that was long assumed (e.g., Cloud, 1976), many questions remain as to the character of Hadean source rocks and geodynamic settings.

Inclusions in zircon have a great deal of potential as indicators of source rock character. They provide a (likely non-exhaustive) snapshot of phases present in the magma during zircon growth. The presence of rutile and other oxide inclusions, for instance, provides a constraint on α_{TiO_2} . The chemistry of some inclusions may also reveal more about the setting in which the magma formed. For instance, Hopkins et al. (2008, 2010) used the Si^{PFU} of Hadean muscovite inclusions combined with T^{zln} from their host zircon

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to estimate the P–T conditions of crystallization for muscovite-bearing Jack Hills zircons, finding unexpectedly low geotherms that may point to formation of the zircons in an under-thrust tectonic environment.

Biotite is the third most abundant inclusion phase among Hadean (>4.03 Ga) Jack Hills zircons (Hopkins et al., 2010), making up approximately 12% of inclusions isolated from cracks in the host zircon (Bell et al., 2015). Magmatic biotite composition varies with magma character (e.g., Abdel-Rahman, 1994; Ague and Brimhall, 1988; Dodge et al., 1969; Burkhard, 1991; Czamanske et al., 1981; Lalonde and Bernard, 1993; Neiva, 1981; Shabani et al., 2003; Anderson and Bender, 1989; Barker et al., 1975; Kinnaird, 1985; Lalonde and Martin, 1983; Parsons, 1981; Platt and Woolley, 1986; Schneiderman, 1991). Furthermore, a study of granitoids from Dronning Maud Land, Antarctica showed that biotite inclusions in magmatic zircon have nearly identical compositions to biotite in the rock matrix, meaning that biotite inclusions show great promise as a tool for detrital zircon provenance (Jennings et al., 2011). In this paper we present biotite compositions from granitic (*sensu lato*) rocks with a range of tectonic settings and bulk compositions. We compare these biotites to inclusions in Hadean and Eoarchean detrital zircons from the Jack Hills and Nuvvuagittuq Supracrustal Belt (NSB) localities, respectively, in order to infer provenance and the implications for chemical variability of magmas on early Earth.

2. Magmatic biotite composition

Various studies have related magmatic biotite chemistry to both the f_{O_2} (given knowledge of other chemical activities) under which it formed and the A/CNK of the host melt. Both quantities should be useful for characterizing the sources of detrital materials, as they correlate both with magma source composition and the geologic setting in which the magma formed. Whether the information recorded in biotite is useful out of a rock context (i.e., absent knowledge of other chemical activities) must first be established.

In general, lower biotite Fe/Mg ratios are characteristic of more oxidized conditions (e.g., Burkhard, 1991; Abdel-Rahman, 1994; Hayama, 1959; Wones and Eugster, 1965; Lalonde and Bernard, 1993; Shabani et al., 2003). This is attributed to the higher magmatic Fe^{3+}/Fe^{2+} leading to crystallization of magnetite, and consequently less Fe available for substitution into biotite. Burkhard (1991) calibrated a biotite Fe/Mg oxybarometer, but quantitative f_{O_2} estimates using these methods require knowledge of crystallization temperature and the activities of magnetite and sanidine. While there are probably some detrital zircons for which this information is available (e.g., those containing magnetite and K-feldspar), the vast majority of biotite-including detrital zircons will likely only contain some of this information. In the absence of this petrologic context, can general trends in biotite Fe/Mg be useful for provenance?

Abdel-Rahman (1994) formulated a discriminant scheme to distinguish magmatic biotite from different source rocks based on Al_2O_3 , FeO^* , and MgO contents. This study found that calc-alkaline, peraluminous, and alkaline A-type granitoids yield compositionally distinct biotite: biotite from peraluminous granitoids has higher Fe/Mg than biotite from calc-alkaline granitoids, consistent with the f_{O_2} trends observed in S- and I-type granitoids, respectively (Chappell and White, 1974). S-type granitoids tend to be more reducing, falling generally into Ishihara's (1977) ilmenite series. I-type granitoids vary in relative oxidation state but are more likely to fall into the more oxidized magnetite series (Ishihara, 1977). In the study of Abdel-Rahman (1994), alkaline A-type granitoids were very enriched in Fe over the other granitoid categories, although the heterogeneity of rocks in the A-type category, espe-

cially in f_{O_2} , leads to a wide range in biotite compositions (e.g., Anderson and Bender, 1989; Barker et al., 1975; Kinnaird, 1985; Lalonde and Martin, 1983; Parsons, 1981; Platt and Woolley, 1986; Schneiderman, 1991).

Fe/Mg ratio of biotite may also be correlated with whole-rock Fe/Mg, as shown by Czamanske et al. (1981) for various granitoids from Japan. These whole-rock Fe/Mg values typically also followed the rule of low Fe/Mg values for oxidized magmas and higher Fe/Mg for reduced magmas, although the correlation between Fe/Mg and SiO_2 varied between their oxidized and reduced samples. Aluminum contents in biotite may also have significance for whole-rock chemistry, with Abdel-Rahman (1994) finding the highest Al contents among biotites from peraluminous, S-type (Chappell and White, 1974) granites and Shabani et al. (2003) finding a trend of higher biotite Al with whole-rock SiO_2 , suggestive of progressive sediment assimilation in several Appalachian granitoids.

3. Materials and methods

Biotite was chemically characterized from one Neoproterozoic and 20 Phanerozoic granitoids ranging in composition from tonalite to granite and deriving from continental arc, collisional, and continental rift settings (see Table 1; more details on sampling location and setting in SOM-1). Biotite from three granitoids was analyzed *in situ* in thick section, and for 18 granitoids the biotite was picked and mounted following crushing to <450 μm . Additionally, zircons were separated from crushed powders of these granitoids, a fuchsite quartzite (Aqf of Cates et al., 2013; cf. Darling et al., 2013) from the Nuvvuagittuq Supracrustal Belt (NSB), and an orthogneiss (Ag of Cates and Mojzsis, 2007, 2009) from the NSB, following elution in water, density separation with heavy liquids, and magnetic separation. Whole-rock major and trace element chemistry were determined by X-ray fluorescence spectroscopy of unseparated powders at Pomona College following double fusion of a 2:1 lithium tetraborate:rock powder mixture in graphite crucibles (Poletti et al., 2016).

Zircons and biotite were separately mounted in epoxy and polished to expose an interior surface. U–Pb ages for NSB zircons were analyzed on the CAMECA *ims1270* ion microprobe at UCLA following the protocol of Quidelleur et al. (1997) using a ca. 15 nA primary O^- beam focused to a ca. 20 μm spot. Rare earth elements (REE), Ti, and other trace elements in NSB zircons were also analyzed on the *ims1270* under similar primary beam conditions, with a –100 V energy offset to suppress molecular interferences (see Wielicki et al., 2012 for more details). Biotite compositions were determined by electron probe microanalysis (EPMA) using a Jeol-8600 electron microprobe at UCLA with a 15 nA beam. For measurements of matrix biotite, the beam was defocused to a ~5 μm -diameter spot to limit K loss, while the small size of most inclusions in zircon necessitated a focused beam <1 μm . Several measurements were also made of matrix biotite using the focused beam to demonstrate the lack of effect on other aspects of measured stoichiometry (analyses designated “small” in SOM-3). Secondary fluorescence during inclusion measurements was monitored by measuring for ZrO_2 , which was used to subtract any SiO_2 contribution from the host zircon. Several biotite analyses display low X-site occupancy due either to K loss from the electron beam or to prior chloritization; analyses with X-site occupancy <0.75 were not considered further due to the possibility of alteration. All analyses were renormalized to 95 wt.% (excluding the unmeasured hydroxide and halogen components). Jack Hills biotite inclusions investigated in this study were identified by Hopkins et al. (2010) from grains dated by Holden et al. (2009).

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