



Recovering the primary geochemistry of Jack Hills zircons through quantitative estimates of chemical alteration

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Abstract

Despite the robust nature of zircon in most crustal and surface environments, chemical alteration, especially associated with radiation damaged regions, can affect its geochemistry. This consideration is especially important when drawing inferences from the detrital record where the original rock context is missing. Typically, alteration is qualitatively diagnosed through inspection of zircon REE patterns and the style of zoning shown by cathodoluminescence imaging, since fluid-mediated alteration often causes a flat, high LREE pattern. Due to the much lower abundance of LREE in zircon relative both to other crustal materials and to the other REE, disturbance to the LREE pattern is the most likely first sign of disruption to zircon trace element contents. Using a database of 378 (148 new) trace element and 801 (201 new) oxygen isotope measurements on zircons from Jack Hills, Western Australia, we propose a quantitative framework for assessing chemical contamination and exchange with fluids in this population. The Light Rare Earth Element Index is scaled on the relative abundance of light to middle REE, or $LREE-I = (Dy/Nd) + (Dy/Sm)$. LREE-I values vary systematically with other known contaminants (e.g., Fe, P) more faithfully than other suggested proxies for zircon alteration (Sm/La, various absolute concentrations of LREEs) and can be used to distinguish primary compositions when textural evidence for alteration is ambiguous. We find that zircon oxygen isotopes do not vary systematically with placement on or off cracks or with degree of LREE-related chemical alteration, suggesting an essentially primary signature. By omitting zircons affected by LREE-related alteration or contamination by mineral inclusions, we present the best estimate for the primary igneous geochemistry of the Jack Hills zircons. This approach increases the available dataset by allowing for discrimination of on-crack analyses (and analyses with ambiguous or no information on spot placement or zircon internal structures) that do not show evidence for chemical alteration. It distinguishes between altered and unaltered samples in ambiguous cases (e.g., relatively high Ti), identifying small groups with potentially differing provenance from the main Jack Hills population. Finally, filtering of the population using the LREE-I helps to more certainly define primary correlations among trace element variables, potentially relatable to magmatic compositional evolution.

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1. INTRODUCTION

Detrital zircons from Jack Hills, Western Australia range up to nearly 4.4 Ga in age and record an abundance

of geochemical information about the crust during Earth's first few hundred million years (Holden et al., 2009; Harrison, 2009). High $\delta^{18}O$ among some zircons has been interpreted as evidence for low-temperature rock–water interactions during this time (e.g., Mojzsis et al., 2001; Peck et al., 2001; Harrison et al., 2008), and low average Ti-in-zircon crystallization temperatures (T^{zln}) suggest near

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granitic minimum melting conditions for the source magmas of the zircons (Watson and Harrison, 2005). The granitic primary mineral inclusion suite, dominated by quartz and muscovite (Hopkins et al., 2008, 2010; cf. Rasmussen et al., 2011; Bell et al., 2015b), further supports granitic source rocks for the zircons. Their relatively unradiogenic Lu-Hf systematics have been interpreted as indicating continental-like material in the source throughout the Hadean (>4 Ga) eon (e.g., Harrison et al., 2005, 2008; cf. Kemp et al., 2010; Bell et al., 2014) and among the more numerous younger zircons (e.g., Amelin et al., 1999; Bell et al., 2011).

Zircon incorporates many petrologically useful minor and trace elements, such that they are a potentially rich yet currently underexploited source of petrologic information on Earth's first few hundred million years. However, the extent to which chemical alteration may have obscured the primary chemistry of the Jack Hills population, and thus its implications for the Hadean, has been a source of controversy. Although many studies have used the trace element contents of the zircons to argue for various igneous or metamorphic origins (e.g., Peck et al., 2001; Crowley et al., 2005; Watson and Harrison, 2005; Cavosie et al., 2006; Coogan and Hinton, 2006; Grimes et al., 2007; Bell and Harrison, 2013), some aspects of zircon trace element chemistry are susceptible to chemical alteration during metamorphism or metasomatism (e.g., Hoskin, 2005 for Jack Hills zircons; Vavra et al., 1999; Hoskin and Black, 2000; Hoskin and Schaltegger, 2003 for zircon in general). For instance, hydrothermally altered zircon often shows a high, flat light rare earth element (LREE) pattern relative to the normal, high ratios of the heavy rare earth elements (HREE) to the LREE in magmatic zircons (e.g., Hoskin and Schaltegger, 2003). Such apparently altered LREE patterns are seen among a minority of Jack Hills zircons (Maas et al., 1992; Peck et al., 2001; Hoskin, 2005; Cavosie et al., 2006), prompting Hoskin (2005) to ascribe them to hydrothermal alteration rather than to high LREE contents among Hadean magmas. Cavosie et al. (2006) attributed similar high-LREE patterns among a minority of zircons to alteration processes other than hydrothermal, given their lack of correlation with anomalous $\delta^{18}\text{O}$. They instead attributed such patterns to contamination by phosphate inclusions or local LREE enrichment at very low water/rock ratios in radiation-damaged regions.

It is known that a variety of metamorphic events affected the ca. 3 Ga host conglomerate of the Jack Hills zircons and its surrounding environs during the Archean and Proterozoic (e.g., Myers, 1988a,b), with several recognized chemical effects on the zircons themselves and their host rocks (e.g., Rasmussen et al., 2011; Pidgeon, 2014). Metasediments at the Jack Hills discovery site fail the paleomagnetic conglomerate test, indicating that they were remagnetized after deposition (Weiss et al., 2015; cf. Tarduno et al., 2015). In addition to Archean and Proterozoic metamorphic events (Myers, 1988a,b; Fletcher et al., 1988), granitoids intruded the region at ca. 3.0 and 2.6 Ga (Myers, 1988a,b; Spaggiari et al., 2007) along with a phase of volcanism at 1.8 Ga (Wilde, 2010), and mafic dikes associated with the Marnda Moorn and Warrakurna Large

Igneous Provinces intruded the region at ca. 1.2 Ga (Spaggiari et al., 2007). Any of these tectonothermal events could potentially create conditions conducive to zircon alteration.

Many Jack Hills zircons contain cracks which are associated with higher Ti and Fe contents relative to the magmatic zircon lattice (Harrison and Schmitt, 2007) and appear to contain slightly lower $\delta^{18}\text{O}$ values (Trail et al., 2007). Cracks in the zircons are often mineralized (Rasmussen et al., 2011; Caro et al., 2008; Bell et al., 2015b), with the dominant phases being quartz, xenotime, Fe oxides, muscovite, and minor monazite (Bell et al., 2015b). These phases are common in the host metaconglomerate (Rasmussen et al., 2010, 2011). Some but not all inclusions in contact with cracks in the zircons have been altered by fluid ingress over geologic time (Bell et al., 2015b). It has so far been unclear, however, to what extent these crack-related zones of alteration may affect the Jack Hills zircon geochemical record as a whole and to what extent the zircons may indeed retain their primary igneous chemistry.

To address these issues, we present a database of published and new trace element and oxygen isotope compositions on a variety of Jack Hills zircon samples with U–Pb dates from 4.2 to 2.2 Ga which range from uncracked and magmatically zoned to highly cracked and/or metamict grains. We investigate the relationship of various trace element concentrations and $\delta^{18}\text{O}$ values to likely alteration features such as cracks, metamict regions, or disrupted zoning as shown by cathodoluminescence (CL) in order to define quantitative estimates for the degree and mechanisms of chemical alteration of Jack Hills zircons, and potential structural markers for such alteration.

2. TRACE ELEMENTS IN ZIRCON

Considerable effort has been expended in developing various aspects of zircon trace element chemistry as proxies for the conditions of zircon formation or for discriminating the provenance of detrital zircons. Several trace element contents and ratios in zircon are quantitative proxies for useful properties of magmas, including the Ti content as a proxy for crystallization temperature (Watson and Harrison, 2005; Ferry and Watson, 2007) and Ce/Ce* as a proxy for f_{O_2} (Trail et al., 2011). Other trace element concentrations and ratios, especially of the rare earth elements (REE), provide less certain and less quantitative information about igneous zircon origins. They may record useful information about magmatic compositional evolution or source region (e.g., cf. Hoskin et al., 2000; Claiborne et al., 2010; Barth et al., 2013). Their usefulness as provenance indicators is a subject of debate (e.g., Hoskin and Ireland, 2000; Belousova et al., 2002). However, in order to properly assess the potential of trace element chemistry as a discriminant of zircon provenance, it must first be established that the effects of later alteration can be distinguished from the original chemistry of the zircon as inherited from the magma (hereafter, 'primary chemistry').

Although normally robust, under certain circumstances zircons can be chemically altered. Damage to or

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