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The oldest zircons of Africa—Their U–Pb–Hf–O isotope and trace element systematics, and implications for Hadean to Archean crust–mantle evolution

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ABSTRACT

More than 450 detrital zircon grains from two Limpopo Belt quartzite samples were investigated by a combination of scanning electron imaging, U–Pb dating, δ^{18}O, Lu–Hf isotope and trace element analyses in order to get robust information about the early Earth's crust–mantle evolution. The detrital zircon grains have crystallization ages between 3.95 Ga and 3.18 Ga, show εHf_t between +1 to −15 (±1 ε-unit), and $\delta^{18}O_{VSMOW}$ mostly between +5.5 and +8.1‰ (±0.2‰). Pristine zircon domains reveal Ti-in-zircon temperatures between 700 and 865 °C, and Th/U of 0.3–2.3. Trace elements point to zircon formation in predominately granitoid rocks. Metamorphic zircon rims have ages ≤2.65 Ga, $\varepsilon Hf_{2.65Ga} \sim -15$, $\delta^{18}O = 7.0$–8.1‰, and Th/U mostly <0.1. Nine zircon grains define an εHf_t-age array (I), which starts from a chondritic uniform reservoir (CHUR) at about 4.5 Ga, and requires $^{176}Lu/^{177}Hf = 0.020$, indicative for mafic crust. Most zircon grains, however, plot on or above an εHf_t-age array (II), which runs parallel to array I, $^{176}Lu/^{177}Hf = 0.021$, and starts from CHUR at 4.01 Ga. Oxygen isotope compositions of $\delta^{18}O > 5.5$ indicate that the magmatic host rocks of the zircons have been formed either by melting of altered mafic crust, which interacted with cold water prior to granitoid formation, and/or that ancient sedimentary and/or magmatic rocks were involved in the melting process. The new U–Pb–Hf–δ^{18}O datasets together with compiled data from worldwide sources indicate a significant gap of about 5 epsilon units between arrays I and II. Furthermore, they illustrate that many Hadean zircon analyses plot well below array I, and some above CHUR. These findings support an interpretation that the Hadean Earth was covered by a long-lived, mafic protocrust, perhaps forming a partially open "stagnant lid". This protocrust was affected by internal reworking, but also injected and overlain by (ultra)mafic rocks derived from chondritic and (highly) depleted mantle sources. At <4.3 Ga, the mafic protocrust was locally transformed into a TTG crust, perhaps caused by enhanced lower crust foundering, related to enhanced volcanic resurfacing and secular cooling of the Hadean lithosphere. Eventually, this heterogeneous Hadean protocrust became completely substituted by a new crust, which started to evolve from the mantle at <4.01 Ga

1. Introduction

Our current knowledge about the early Earth's crust–mantle evolution during the Hadean and Early Archean remains fragmentary and controversial for at least two main reasons. First, there is only limited material to provide direct evidence for the early Earth's crustal evolution, and second, it is challenging to extract pristine information from Hadean/Archean materials. The oldest rocks preserved on Earth, the Acasta Gneisses in western Canada, formed at <4.03 Ga (Iizuka et al., 2006, 2007, 2009), *i.e.*, about 550 Ma after formation of the proto-Earth from the solar nebula at about 4.567 Ga (Amelin et al., 2002), and the subsequent giant impact (ca. 60 Ma later), which caused the formation of the Earth–Moon system (*e.g.*, Bourdon et al., 2008). The oldest direct source of information for the early Earth's crust–mantle evolution includes detrital zircon grains in meta-conglomerates of the Jack Hills of Western Australia with U–Pb ages up to 4.4 Ga (*e.g.*, Wilde et al., 2001). These zircon grains have been extensively investigated over the past 15 years, comprising studies on zircon U–Pb ages and Lu–Hf isotopes (*e.g.*, Maas et al., 1992; Amelin et al., 1999, 2000; Harrison et al., 2005, 2008; Blichert-Toft and Albarède, 2008; Kemp et al., 2010; Nebel-Jacobsen et al., 2010), oxygen isotopes (Peck et al., 2001; Cavosie et al., 2005; Nemchin et al., 2006; Harrison et al., 2008; Bell et al.,

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2012), trace elements (Cavosie et al., 2006; Harrison et al., 2008), mineral inclusions (e.g., Harrison et al., 2005; Menneken et al., 2007; Hopkins et al., 2008, 2010; Rasmussen et al., 2011), and Li-isotopes (Ushikubo et al., 2008).

However, despite the large number of investigations of the Jack Hills zircons, comprising more than 100,000 grains (Holden et al., 2009), the formation and evolution of the early Earth's crust and mantle remain controversial. Presently, there are two contrasting models, designated here as **HACCAP (Ha**dean **C**ontinental **C**rust Formation **a**nd **P**late tectonics) and **LOLIHP (Lo**ng **li**ved **H**adean **P**rotocrust) models. The HACCAP model postulates that (abundant) continental crust made up of TTG's (tonalities–trondhjemites–granodiorites) and a supplementary depleted mantle reservoir existed early during the Earth's history, along with (local?) highly LILE-depleted mantle reservoirs and extremely LILE-enriched, non-radiogenic (Nd, Hf) crust, and that subduction-driven plate tectonics (similar to present day) operated during the Hadean Eon (Harrison et al., 2005, 2008; Blichert-Toft and Albarède, 2008; Hopkins et al., 2008, 2010). The HACCAP model is mainly based on the observations that Hadean (Jack Hills) detrital zircon grains reveal extremely positive or negative εHf_t (*e.g.*, Harrison et al., 2005), and contain quartz, K-feldspar and muscovite inclusions (*e.g.* Hopkins et al., 2008, 2010). Furthermore, they reveal steep εHf_t-age trends, which point to the formation and reworking of TTG crust, with low $^{176}Lu/^{177}Hf < 0.01$ between 4.3 and 4.1 Ga (*e.g.*, Blichert-Toft and Albarède, 2008), or even earlier (Harrison et al., 2005, 2008).

The LOLIHP model, on the other hand, suggests that the early Earth was covered by a volumetrically insignificant, relatively stable 'KREEP'-like mafic protocrust (KREEP: high potassium, REE and phosphorus), and that this protocrust was continuously reworked by re-melting over nearly 400 Ma (without the addition of new material from the depleted mantle), and finally disappeared with the onset of new crust formation during the Eoarchaean at about 4.0 Ga, perhaps by the onset of modern style plate tectonics. This model was suggested by Kemp et al. (2010) and comprises many concepts proposed previously by Kamber et al. (2003, 2005) and Shirey et al. (2008). The LOLIHP model is based mainly on data from two very careful LA–ICP–MS studies on detrital and magmatic zircons from the Jack Hills and Greenland (Kemp et al., 2009, 2010), and on zircon data from a lunar breccia (Taylor et al., 2009). It is furthermore supported by solution U–Pb–Hf isotope data of Jack Hills zircons (Amelin et al., 1999, 2000), and by the observation that zircon grains with quartz and K-feldspar inclusions can occur also in mafic rocks (Darling et al., 2009), and that mineral inclusions in zircon are not always primary, but can be formed or altered during subsequent metamorphism (Rasmussen et al., 2011). The datasets of Kemp et al. (2009, 2010) and Amelin et al. (1999, 2000) neither provide evidence for the existence of a significantly depleted (global scale) mantle reservoir during the Hadean to Eoarchean, nor for the existence of (abundant) continental (TTG) crust. Modern style plate tectonics during the Hadean is not required, and data which plot significantly below or above the εHf_t-age trend for the Hadean mafic protocrust are interpreted to result from analytical problems (*i.e.*, mixed zircon core-rim analyses during *in-situ* LA–ICP–MS dating and/or zircon dissolution studies), and/or from alteration processes (*e.g.* zircon re-crystallization), causing uncoupling between the U–Pb zircon age and initial $^{176}Hf/^{177}Hf$.

Thus, the contrasting models proposed for the early Earth's crust–mantle evolution result from a different interpretation of the same datasets, and whether or not these datasets are believed to reflect pristine information and, thus, the composition of the Earth at the time of zircon formation (also see discussions in Cavosie et al., 2006; Valley et al., 2006; Kemp et al., 2010; Hopkins et al., 2012; Rasmussen et al., 2012). The major concern revolves around combined U–Pb and Lu–Hf isotope datasets, which are most critical for the interpretation of the early Earth's crust–mantle evolution. In general there may be two main problems related to such datasets. The first problem results from the behaviour of zircon which may preserve initial Hf isotope composition during multiple episodes of metamorphism, but which may undergo resetting of the U–Pb system. This uncoupling between the U–Pb and the Lu–Hf isotope systems (=Age-Hf isotope uncoupling) has been demonstrated by many field based and experimental studies in the past (*e.g.*, Zeh et al., 2007, 2010a, 2011; Gerdes and Zeh, 2009; Lenting et al., 2010). The second problem is that Hadean/Archean zircon grains are often affected by multiple (over)growths and re-crystallization with large time gaps in between (up to several 100 Ma), causing the formation of zircon grains with complex zoning, whereby different zones can reveal different U–Pb ages and different initial $^{176}Hf/^{177}Hf$ (for example see Gerdes and Zeh, 2009; and this study). For such grains, greatest analytical care must be taken to avoid mixed core-rim analyses. Zircon alteration and multiple zircon growth, however, have not only implications for the interpretation of coupled U–Pb and Lu–Hf isotope datasets, but also for the interpretation of related trace element and oxygen isotope data. In fact, fluid infiltration into fractured and/or metamict zircon domains may lead to a significant change of the initial Ti-content (used for Ti-in-zircon thermometry, *e.g.* Harrison and Schmitt, 2007; Fu et al., 2008), or to a shift of the primary $\delta^{18}O$ (*e.g.*, Booth et al., 2005; Zeh et al., 2012; Pidgeon et al., 2013), or of the Li isotope composition (Ushikubo et al., 2008). Variations of trace elements and isotopes can also result from multiple zircon growth.

For Archean orthogneisses, zircon alteration can be detected easily in most cases by the analysis of a large number of zircon grains, whereby the oldest U–Pb age and lowest initial $^{176}Hf/^{177}Hf$ reflect the time of crystallization and the initial Hf isotope composition of the magma (*e.g.*, Gerdes and Zeh, 2009). Subsequently, trace element and $\delta^{18}O$ analyses can be obtained from the most pristine grains/domains in order to gain additional genetic information, like crystallization temperatures (Ti-contents), the state of melt fractionation (Sm/Yb_N), or fluid rock interactions ($\delta^{18}O$; *e.g.*, Hiess et al., 2011). For Hadean/Archean detrital zircon grains, the situation is much more difficult, as each grain can originate from a different source and, therefore, can have different ages, isotope and trace element compositions. Thus, alteration related Hf isotope-age uncoupling is difficult to quantify, in particular if just a single U–Pb and Lu–Hf isotope analysis ($\pm\delta^{18}O$, ±trace element analysis) is obtained from each detrital zircon grain. Faithful information, however, can be obtained by multiple analyses (or even mapping) of U–Pb and Lu–Hf isotopes ($\pm\delta^{18}O$ and trace element) on single zircon grains which are well characterized by CL/BSE imaging (*e.g.*, Cavosie et al., 2006). This procedure is applied during this study to a large number of detrital zircon grains from two quartzite samples in order to get new and unambiguous information about the crust–mantle evolution in the hinterland of the Limpopo Belt. In combination with data from worldwide sources these new data also place new constraints on the global crust–mantle evolution and crust re-working during the Hadean to Archean Eon.

2. Geological setting and samples

The Beit Bridge quartzite of the Limpopo Belt's Central Zone in South Africa is known to contain the oldest detrital zircon grains discovered so far in Africa (Zeh et al., 2008). However, due to their scarcity (just two grains >3.8 Ga have been found so far), and the lack of $\delta^{18}O$ and trace element data, their correlation with Hadean to Paleoarchean zircon grains from other worldwide sources is very limited at the moment. A detailed geological description of the Limpopo Belt's Central Zone is given in Zeh et al. (2004, 2008, 2010a), and only a few important points will be repeated here.