Hadean zircon formed due to hydrated ultramafic protocrust melting

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ABSTRACT

Hadean zircons, from the Jack Hills (Western Australia) and other localities, are currently the only window into the earliest terrestrial felsic crust, the formation of which remains enigmatic. Based upon new experimental results, generation of such early crust has been hypothesized to involve the partial melting of hydrated peridotite interacting with basaltic melt at low pressure (<10 km), but it has yet to be demonstrated that such liquids can indeed crystallize zircons comparable to Jack Hills zircon. We used thermodynamic and geochemical modeling to test this hypothesis. The predicted zircon saturation temperatures of <750 °C, together with the model zircon Th, U, Nb, Hf, Y, and rare earth element (REE) contents at 700 °C, δ18OVSMOW (Vienna standard mean ocean water) signatures, and co-crystallizing mineral assemblage were compared to those of the Jack Hills zircon. This comparison was favorable with respect to crystallization temperature, most trace-element contents, and mineral inclusions in zircon. The discrepancy in δ18OVSMOW signatures may be explained by hotter conditions of Hadean protocrust hydration. Our work supports the idea that felsic magma generation at shallow depths involving a primordial weathered ultramafic protocrust and local basaltic intrusions is indeed a viable mechanism for the formation of felsic crust on early Earth.

INTRODUCTION

The conditions and mechanisms that led to formation of the earliest felsic (Si- and Al-enriched) crust on Earth and Mars are the subject of debate (e.g., Kemp et al., 2010; Burnham and Berry, 2017; Harrison et al., 2017; Turner et al., 2020). In terms of general context, one possibility is that the Hadean Eon was very similar to the modern Earth; i.e., a voluminous oceanic basaltic residuum with high abundance of incompatible elements, with some similarity to lunar KREEP basalts (e.g., Cronberger and Neal, 2018). Borisova et al. (2021) performed experiments on the interaction between serpentinite and basaltic magma that demonstrated that tonalite to granodiorite compositions can be formed in this way at shallow depths (<10 km). In this scenario, partial melting is triggered by dehydration driven by intrusion of basaltic magmas or by meteorite impacts. The most common approach to address the question of the origin of the felsic Hadean crust is to investigate the possibility to crystallize zircons analogous to the Hadean zircons, the only mineral remnants of Earth’s Hadean
crust identified so far. Most samples are detrital zircons from the Jack Hills (JHZ), Western Australia (Harrison, 2009), but other localities are also known (Harrison et al., 2017), including a recent discovery in South Africa (Drabon et al., 2021). While the Hadean zircons provide valuable information about their crystallization environment, no clear consensus exists for the genesis conditions of their parental magmas. In any case, mechanisms for the generation of Earth’s earliest felsic crust should be compatible with the mineralogical and geochemical features of the Hadean detrital zircons.

Our study tested the hypothesis that an early felsic crust may have formed by low-pressure, fluid-present melting of hydrated peridotite interacting with basaltic magma through assessment of the capacity of experimental felsic melts (Borisova et al., 2021) to crystallize zircon (referred to here as “model zircon”) and then comparison of the temperature conditions, co-crystallizing mineral assemblages, and isotopic and trace-element patterns of these model zircons with data from detrital Hadean zircons.

ANALYTICAL AND MODELING METHODS
Experiments performed at 1250–1300 °C, 0.2 GPa (corresponding to depth of ∼5 km), and redox conditions from ΔQFM +1.8 to ΔQFM +4.4 (expressed in log_{10} fO2 relative to the quartz-fayalite-magnetite [QFM] oxygen fugacity [fO2] buffer) indicate that partial melts produced at these conditions are enriched in SiO2 (average 66 ± 4 wt% SiO2; Figs. 1A and 1B; Tables S1 and S2 in the Supplemental Material1) and other lithophile elements (Al, alkalis), with abundances broadly similar to those of continental crustal rocks (Borisova et al., 2021).

Details of the modeling methods and new analytical data on the experimental products are summarized in the Supplemental Material and Tables S2–S5 therein. We performed four types of modeling: (1) simulations of equilibrium and fractional crystallization of the experimentally produced felsic liquids to determine the mineral phases crystallizing during cooling, using the rhyolite-MELTS thermodynamic calculator (version 1.2.0, http://melts.ofm-research.org/; Gualda et al., 2012); (2) simulations of temperature conditions of zircon saturation/crystallization of the produced felsic liquids; (3) modeling of zircon oxygen isotope compositions; and (4) geochemical modeling of the model zircon compositions using available zircon-melt partitioning coefficients for Th, U, Nb, Hf, Y, and REEs (Table S2; Claiborne et al., 2018).

MODEL CRYSTALLIZATION OF EXPERIMENTAL FELVIC MELTS
The fluid-present interaction of hydrated peridotite with basaltic melt at 0.2 GPa and 1250 °C and ΔQFM +1.8 to +4.4 results in formation of interstitial water-saturated, silica-rich, peraluminous (average aluminum saturation index [ASI] = 1.2 ± 0.2; n = 21) melts in association with high-Mg olivine (Fo91–94) and chromite (Fig. 1A; Tables S1 and S2). Model crystallization sequences using the rhyolite-MELTS...
Figure 2. Zircon saturation expressed in zircon contents ($C_{sat}$ ppm) versus temperature ($T$, °C) according to Supplemental equations 1 (Boehnke et al., 2013; Bindeman and Melnik, 2016), 2 (Harrison et al., 2007), and 3 (Watson and Harrison, 1983), and the M factors (computational factor $M = (Na + K + 2Ca)/(Al \times Si)$ (1.4–2.9) (see the Supplemental Material; Table S4 [see footnote 1]). Dashed red horizontal lines correspond to minimum (49 ppm) and maximum (110 ppm) zircon contents in experimental felsic glasses. Red field indicates conditions (below 750 °C) of zircon crystallization above solids (~700 °C).

Figure 3. $\delta^{18}O_{VSMOW}$ (VSMOW—Vienna standard mean ocean water) in the model (Model) zircon (Zn) crystallized from felsic melts at 700–750 °C versus temperature of hydrous protolith (serpentinite) formation. Lines demonstrate the composition of model zircon crystallized from felsic liquids produced due to two mixtures of serpentinite (50 wt% and 80 wt%) with basalt (at 1250–1300 °C and 0.2 GPa). Gray field represents the most common compositions of Jack Hills zircon (JHZ; Cavosie et al., 2006; Whitehouse et al., 2017). Pink field represents the model composition of zircon that can crystallize from experimental felsic liquids. To calculate $\delta^{18}O_{VSMOW}$ in the model zircon, Supplemental equations 4–7 (see the Supplemental Material and Tables S1 and S5 [see footnote 1]) were applied (according to the fractionation factors of Savin and Lee [1988]).

ZIRCON TRACE-ELEMENT CHEMISTRY

The trace-element composition of model zircon was calculated based on the experimental felsic glass composition, zircon-melt partitioning coefficients at 700 °C from Claiborne et al. (2018) (Fig. 4; Table S2) and the Ti-in-zircon equation of Ferry and Watson (2007) assuming activities of SiO$_2$ and TiO$_2$ in the melt of $\varphi_{SiO_2} = \varphi_{TiO_2} = 1$. The assumption of $\varphi_{TiO_2} = 1$ is the source of uncertainty for the zircon Ti and the model trace-element contents, as illustrated in Figure 4 and in Table S3, but it does not affect the principal conclusions reached. In contrast to Turner et al. (2020), we used zircon-melt partitioning data obtained at much lower temperatures (700 °C versus 1300 °C). This choice is critical because of the strong dependence of the mineral-melt partitioning on temperature (Claiborne et al., 2018). We used multi-element diagrams of Grimes et al. (2015) and normalization to the MOR zircon to compare the model zircon composition to that of Hadean detrital zircons. Most elements, especially REEs, Nb, Y, and Hf contents, are similar to those of the median MOR zircon. The Nb/Yb (0.01–0.03) and U/Yb (0.01–0.15) ratios of the model zircon (Table S2) are variable and characteristic of the mantle array after Grimes et al. (2015). The mantle array signatures of the model zircons are controlled by the composition of the starting tholeitic MOR basaltic glass (Borisova et al., 2021), and by the conditions of parental felsic melt equilibration.

COMPARISON WITH HADEAN ZIRCONS, AND DISCUSSION

The calculated zircon saturation temperatures demonstrate that the experimentally produced felsic melts may precipitate zircon upon cooling below ~750 °C, in agreement with low-temperature estimates of ~680 °C of products. The $\delta^{18}O_{VSMOW}$ values ranging from 6.2‰ ± 1.4‰ to 6.6‰ ± 0.9‰ were measured in situ by secondary ion mass spectrometry in the olivines in equilibrium with the experimentally produced felsic melts (Tables S1 and S5). The corresponding bulk $\delta^{18}O$ signature of the samples and the experimental phases were a function of the relative contributions from serpentinite and basalt, with the former being a function of the temperature of hydration. Taking into consideration the fractionation factors between olivine and zircon of 1.0‰–1.1‰ at 750–700 °C (Chiba et al., 1989; Valley et al., 2003), and assuming protolith hydration at 80 °C to ~120 °C, the calculated $\delta^{18}O$ enrichment in model zircons ranged from 7.2‰ to 9.9‰ (Fig. 3), above the mantle value of 5.3‰. These calculations thus indicate that zircon crystallizing from felsic melts generated in this scenario inherits the $\delta^{18}O$-enriched signature of the serpentinite protolith.

OXYGEN ISOTOPE SIGNATURES

In the experimental study of Borisova et al. (2021), modern tholeiitic mid-ocean ridge (MOR) basalt ($\delta^{18}O_{VSMOW} = 5.8‰$, VSMOW—Vienna standard mean ocean water) and a serpentinite with 9.7‰ $\delta^{18}O$ (VSMOW—Vienna standard mean ocean water) and a serpentinite with 9.7‰ $\delta^{18}O$ (VSMOW) were used as the starting composition to that of Hadean detrital zircons. Most elements, especially REEs, Nb, Y, and Hf contents, are similar to those of the median MOR zircon. The Nb/Yb (0.01–0.03) and U/Yb (0.01–0.15) ratios of the model zircon (Table S2) are variable and characteristic of the mantle array after Grimes et al. (2015). The mantle array signatures of the model zircons are controlled by the composition of the starting tholeitic MOR basaltic glass (Borisova et al., 2021), and by the conditions of parental felsic melt equilibration.

The predicted zircon saturation temperatures according to the liquid M factors (computational factor $M = (Na + K + 2Ca)/(Al \times Si)$ (1.4–2.9) calculated after Watson and Harrison (1983), Harrison et al. (2007), Boehnke et al. (2013), and Bindeman and Melnik (2016) imply that zircon would crystallize on cooling at temperatures in the range of 750–700 °C (Fig. 2; see the Supplemental Material and Tables S3 and S4).

ZIRCON TRACE-ELEMENT CHEMISTRY

The trace-element composition of model zircon was calculated based on the experimental felsic glass composition, zircon-melt partitioning coefficients at 700 °C from Claiborne et al. (2018) (Fig. 4; Table S2) and the Ti-in-zircon equation of Ferry and Watson (2007) assuming activities of SiO$_2$ and TiO$_2$ in the melt of $\varphi_{SiO_2} = \varphi_{TiO_2} = 1$. The assumption of $\varphi_{TiO_2} = 1$ is the source of uncertainty for the zircon Ti and the model trace-element contents, as illustrated in Figure 4 and in Table S3, but it does not affect the principal conclusions reached. In contrast to Turner et al. (2020), we used zircon-melt partitioning data obtained at much lower temperatures (700 °C versus 1300 °C). This choice is critical because of the strong dependence of the mineral-melt partitioning on temperature (Claiborne et al., 2018). We used multi-element diagrams of Grimes et al. (2015) and normalization to the MOR zircon to compare the model zircon composition to that of Hadean detrital zircons. Most elements, especially REEs, Nb, Y, and Hf contents, are similar to those of the median MOR zircon. The Nb/Yb (0.01–0.03) and U/Yb (0.01–0.15) ratios of the model zircon (Table S2) are variable and characteristic of the mantle array after Grimes et al. (2015). The mantle array signatures of the model zircons are controlled by the composition of the starting tholeitic MOR basaltic glass (Borisova et al., 2021), and by the conditions of parental felsic melt equilibration.
Figure 4. (A) Zircon composition normalized to median mid-ocean ridge (MOR) zircon. (B) U/Yb versus Nb/Yb and Gd/Yb ratios. Median MOR zircon is after Grimes et al. (2015). The model zircon composition was calculated at 700 °C. Gray circles represent the entire compositional range for Jack Hills zircon (JHZ): 4.37–4.02 Ga type-1 crystals (Cavosie et al., 2006) and several ≥4 Ga Jack Hills crystals (Turner et al., 2020). The result of 10% of KREEP addition to the parental basalt is shown (where KREEP indicates residuum with high abundance of incompatible elements such as K, rare earth elements [REEs], and P). Present-day zircon types, mantle array, and Green Sandstone Bed (GSB) zircon composition are after Grimes et al. (2015) and Drabon et al. (2021). Model data and references are given in Table S2 (see footnote 1).

The trace-element composition of the model zircons is similar to those of the ≥4 Ga JHZ (Cavosie et al., 2006; Turner et al., 2020) and the Green Sandstone Bed zircons (Figs. 4A and 4B; Drabon et al., 2021). In detail, the REEs, Nb, Y, and Hf contents are in agreement with those of the ≥4 Ga zircons, while there is nevertheless a noticeable depletion in Th and U contents of the model zircon compared to the Hadean ones. In this respect, we note that the starting experimental basalt was a tholeiite with composition similar to that of Archean tholeiite. Nevertheless, the discrepancy between model and Hadean zircons cannot be explained only by tapping a mantle reservoir with primitive composition. It requires a contribution of an enriched KREEP-like reservoir, corresponding to very late-stage liquids derived from magma ocean crystallization that may reach Th contents >10 ppm (Snyder et al., 1995; Neal and Kramer, 2006). We note that production of KREEP-like basalts at the onset of the Hadean Eon was also assumed by Kemp et al. (2010). However, our model differs by stating that Earth’s protocrust was made of peridotites and not of KREEP-like basalts that remained under the protocrust. From a geochemical perspective, the participation of 10% of KREEP-like basalts is sufficient to explain the Th discrepancy (Fig. 4A), but it does not seem to fit the heaviest REEs (Yb and Lu), which are slightly too high, resulting in lower U/Yb (Fig. 4B). However, the strong partitioning of the heavy (H) REEs in amphibole (e.g., Sisson, 1994) implies that crystallization of amphibole from the experimental felsic melt could offset this effect on the HREE budget.

The quartz, plagioclase, apatite, amphibole, and biotite that crystallize from the felsic melt during model cooling from 750 to 700 °C (Table S3) are common JHZ-hosted mineral inclusions (Cavosie et al., 2004). We demonstrate here that the felsic melts produced by Borisova et al. (2021) are peraluminous, similar to those inferred to be the hypothetical parental melts for JHZ (Harrison, 2009). Muscovite inclusions are also abundant in the JHZ, although their primary or secondary origin is debated (Rasmussen et al., 2011; Bell et al., 2015). Rhylolite-MELTS modeling shows that muscovite may crystallize in near-solidus conditions (Table S3). Furthermore, the aqueous fluid coexisting with the melt is highly enriched in Si, Al, and alkalis, according to calculations by Borisova et al. (2021), and therefore could crystallize muscovite as a late- to postmagmatic phase.

Thus, our results and comparisons collectively support the proposed mechanism (Borisova et al., 2021) of formation of the first Hadean felsic crust via aqueous fluid-assisted partial melting of hydrated peridotite induced by reaction with basaltic melt at depths of ~10 km. A minor contribution of KREEP-like basalts is required to provide the best fit to the trace-element signatures. When taken together, we conclude from these comparisons that the experimental felsic liquids could be good analogs of parental melts that formed JHZ crystals. Their formation thus does not seem to require an environment associated with plate tec-tonics, and such a mechanism could also explain the formation of a Noachian felsic crust on Mars.

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