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1 Hadean zircon formed due to hydrated ultramafic
2 protocrust melting

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26 **ABSTRACT**

27 Hadean zircons, from the Jack Hills (**Western Australia**) and other localities, are
28 currently the only window into the earliest terrestrial felsic crust, the formation **of which**
29 remains enigmatic. Based upon new experimental results, generation of such early crust
30 has been hypothesized to involve the partial melting of hydrated peridotite interacting
31 with basaltic melt at low pressure (<10 km), but it has yet to be demonstrated that such
32 liquids can indeed crystallize zircons comparable to Jack Hills zircon. **We used**
33 thermodynamic and geochemical modeling to test this hypothesis. The predicted zircon
34 saturation temperatures of <750 °C, together with the model zircon Th, U, Nb, Hf, Y, and
35 rare earth element (REE) contents at 700 °C, $\delta^{18}\text{O}_{\text{VSMOW}}$ (**Vienna standard mean ocean**
36 **water**) signatures, and co-crystallizing mineral assemblage **were** compared to those of the
37 Jack Hills zircon. This comparison **was** favorable with respect to crystallization
38 temperature, most trace-element contents, and mineral inclusions in zircon. The
39 discrepancy in $\delta^{18}\text{O}_{\text{VSMOW}}$ signatures may be explained by hotter conditions of Hadean
40 protocrust hydration. **Our** work supports the idea that felsic magma generation at shallow
41 depths involving a primordial weathered ultramafic protocrust and local basaltic
42 intrusions is indeed a viable mechanism for the formation of felsic crust on early Earth.

43 **INTRODUCTION**

44 The conditions and mechanisms that led to formation of the earliest felsic (Si- and
45 Al-enriched) crust on Earth and Mars are the subject of debate (e.g., Kemp et al., 2010;
46 Burnham and Berry, 2017; Harrison et al., 2017; Turner et al., 2020). In terms of general
47 context, one possibility is that the Hadean **Eon** was very similar to the modern Earth; i.e.,
48 a voluminous granitic continental crust shaped by plate tectonics (Armstrong, 1981;
49 Harrison, 2009). A contrasting scenario is a single-plate basaltic protocrust that formed
50 after the solidification of the global magma ocean (Kramers, 2007). In either case,
51 various scenarios have been proposed to account for Hadean felsic crust formation, either
52 through processes resembling those of Archean **tonalite-trondjemite-granodiorite (TTG)**
53 formation, such as melting of a garnet-bearing igneous protolith at depth (Burnham and
54 Berry, 2017), or **through** shallow-level hydration of an igneous mafic protolith before
55 subsequent melting to generate Hadean felsic crust (e.g., Drabon et al., 2021). For
56 example, Kemp et al. (2010) suggested that a thin KREEP-like crust accumulated at
57 Earth's surface just after crystallization of the magma ocean and interacted with the
58 hydrosphere (**where KREEP indicates residuum with high abundance of incompatible**
59 **elements such as K, rare earth elements [REEs], and P**). This hydrated protocrust was
60 subsequently buried by later komatiitic and basaltic lava flows and finally remelted due
61 to radioactive self-heating to generate the magmas that crystallized the Hadean zircons.
62 As an alternative scenario, Borisova et al. (2021) assumed that a primitive protocrust
63 quenched on top of the magma ocean, as observed in a permanent lava lake (Helz, 2009).
64 In the case of the Hadean, this protocrust would have been of peridotite composition and
65 become rapidly hydrated (e.g., serpentinized) in its uppermost part by the readily
66 available liquid hydrosphere (Valley et al., 2002; Elkins-Tanton, 2012), as proposed by

67 Albarède and Blichert-Toft (2007). The residual part of the magma ocean then evolved
68 beneath the early quenched crust to yield basaltic magmas, likely enriched in
69 incompatible elements, with some similarity to lunar KREEP basalts (e.g., Cronberger
70 and Neal, 2018). Borisova et al. (2021) performed experiments on the interaction
71 between serpentinite and basaltic magma that demonstrated that tonalite to granodiorite
72 compositions can be formed in this way at shallow depths (<10 km). In this scenario,
73 partial melting is triggered by dehydration driven by intrusion of basaltic magmas or by
74 meteorite impacts.

75 The most common approach to address the question of the origin of the felsic
76 Hadean crust is to investigate the possibility to crystallize zircons analogous to the
77 Hadean zircons, the only mineral remnants of Earth's Hadean crust identified so far.
78 Most samples are detrital zircons from the Jack Hills (JHZ), **Western** Australia (Harrison,
79 2009), but other localities are also known (Harrison et al., 2017), including a recent
80 discovery in South Africa (Drabon et al., 2021). While the Hadean zircons provide
81 valuable information about their crystallization environment, no clear consensus exists
82 for the genesis conditions of their parental magmas. In any case, mechanisms for the
83 generation of Earth's earliest felsic crust should be compatible with the mineralogical and
84 geochemical features of the Hadean detrital zircons.

85 **Our** study tested the hypothesis that an early felsic crust may have formed by low-
86 pressure, fluid-present melting of hydrated peridotite interacting with basaltic magma
87 through assessment of the capacity of experimental felsic melts (Borisova et al., 2021) to
88 crystallize zircon (referred to here as "model zircon") **and** then comparison of the

89 temperature conditions, co-crystallizing mineral assemblages, and isotopic and trace-
90 element patterns of these model zircons with data from detrital Hadean zircons.

91 **ANALYTICAL AND MODELING METHODS**

92 Experiments performed at 1250–1300 °C, 0.2 GPa (corresponding to depth of ~5
93 km), and redox conditions from $\Delta\text{QFM}+1.8$ to $\Delta\text{QFM}+4.4$ (expressed in $\log_{10}f_{\text{O}_2}$ relative
94 to the quartz-fayalite-magnetite [QFM] oxygen fugacity [f_{O_2}] buffer) indicate that partial
95 melts produced at these conditions are enriched in SiO_2 (average 66 ± 4 wt% SiO_2 ; Figs.
96 1A and 1B; Tables S1 and S2 in the Supplemental Material¹) and other lithophile
97 elements (Al, alkalis), with abundances broadly similar to those of continental crustal
98 rocks (Borisova et al., 2021).

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

109 **MODEL CRYSTALLIZATION OF EXPERIMENTAL FELSIC MELTS**

110 The fluid-present interaction of hydrated peridotite with basaltic melt at 0.2 GPa
111 and 1250 °C and $\Delta\text{QFM}+1.8$ to +4.4 results in formation of interstitial water-saturated,

112 silica-rich, peraluminous (average aluminum saturation index [ASI] = 1.2 ± 0.2 ; $n = 21$)
113 melts in association with high-Mg olivine (Fo₉₁₋₉₄) and chromite (Fig. 1A; Tables S1 and
114 S2). Model crystallization sequences using the rhyolite-MELTS calculator at conditions
115 of Δ QFM+2 and 0.2 GPa predict equilibrium crystallization of quartz (800–700 °C),
116 plagioclase (800–700 °C), and apatite (800–700 °C) along with amphibole (775–700 °C)
117 and biotite (760–700 °C) in the presence of aqueous fluid (H₂O) (Table S3). The
118 predicted zircon saturation temperatures according to the **liquid M factors** [**compositional**
119 **factor $M = (\text{Na} + \text{K} + 2\text{Ca})/(\text{Al} \times \text{Si})$**] (1.4–2.9) calculated after Watson and Harrison
120 (1983), Harrison et al. (2007), Boehnke et al. (2013), and Bindeman and Melnik (2016)
121 imply that zircon would crystallize on cooling at temperatures in the range of 750–700 °C
122 (Fig. 2; see the Supplemental Material **Correct? Or did you mean to just cite the**
123 **supplemental tables?]]**; Tables S3 and S4).

124 OXYGEN ISOTOPE SIGNATURES

125 In the experimental study of Borisova et al. (2021), modern tholeiitic mid-ocean
126 ridge (MOR) basalt ($\delta^{18}\text{O}_{\text{VSMOW}} = 5.8\text{‰}$, **VSMOW—Vienna standard mean ocean water**)
127 and a serpentinite with 9.7‰ $\delta^{18}\text{O}_{\text{VSMOW}}$ (suggesting hydration below ~120 °C; Fig. 3;
128 see the Supplemental Material) were used as the starting products. The $\text{O}^{18}_{\text{VSMOW}}$ values
129 ranging from $6.2\text{‰} \pm 1.4\text{‰}$ to $6.6\text{‰} \pm 0.9\text{‰}$ were measured *in situ* by secondary ion
130 mass spectrometry in the olivines in equilibrium with the experimentally produced felsic
131 melts (Tables S1 and S5). The corresponding bulk $\delta^{18}\text{O}$ signature of the samples and the
132 experimental phases **were** a function of the relative contributions from serpentinite and
133 basalt, with the former being a function of the temperature of hydration. Taking into
134 consideration the fractionation factors between olivine and zircon of 1.0‰–1.1‰ at 750–

135 700 °C (Chiba et al., 1989; Valley et al., 2003), and assuming protolith hydration at 80 °C
136 to ~120 °C, the calculated ^{18}O enrichment in model zircons ranged from 7.2‰ to 9.9‰
137 (Fig. 3), above the mantle value of 5.3‰. These calculations thus indicate that zircon
138 crystallizing from felsic melts generated in this scenario inherits the ^{18}O -enriched
139 signature of the serpentinite protolith.

140 **ZIRCON TRACE-ELEMENT CHEMISTRY**

141 The trace-element composition of model zircon was calculated based on zircon
142 and/or melt partitioning coefficients at 700 °C from Claiborne et al. (2018) (Fig. 4; Table
143 S2) and the Ti-in-zircon equation of Ferry and Watson (2007) assuming activities of SiO_2
144 and TiO_2 in the melt of $\alpha_{\text{SiO}_2} = \alpha_{\text{TiO}_2} = 1$. The assumption of $\alpha_{\text{TiO}_2} = 1$ is the source of
145 uncertainty for the zircon Ti and the model trace-element contents, as illustrated in Figure
146 4 and in Table S3, but it does not affect the principal conclusions reached. In contrast to
147 Turner et al. (2020), we used zircon/melt partitioning data obtained at much lower
148 temperatures (700 °C versus 1300 °C). This choice is critical because of the strong
149 dependence of the mineral/melt partitioning on temperature (Claiborne et al., 2018). We
150 used multi-element diagrams of Grimes et al. (2015) and normalization to the MOR
151 zircon to compare the model zircon composition to that of Hadean detrital zircons. Most
152 elements, especially REEs, Nb, Y, and Hf contents, are similar to those of the median
153 MOR zircon. The Nb/Yb (0.01–0.03) and U/Yb (0.01–0.15) ratios of the model zircon
154 (Table S2) are variable and characteristic of the mantle array after Grimes et al. (2015).
155 The mantle array signatures of the model zircons are controlled by the composition of the
156 starting tholeiitic MOR basaltic glass (Borisova et al., 2021), and by the conditions of
157 parental felsic melt equilibration.

158 **COMPARISON WITH HADEAN ZIRCONS, AND DISCUSSION**

159 The calculated zircon saturation temperatures demonstrate that the experimentally
160 produced felsic melts may precipitate zircon upon cooling below ~750 °C, in agreement
161 with low-temperature estimates of ~680 °C of crystallization of Hadean JHZ crystals
162 (Cavosie et al., 2006; Harrison et al., 2017).

163 Available data on the JHZ show $\delta^{18}\text{O}_{\text{VSMOW}}$ values (6.0‰–7.3‰; Cavosie et al.,
164 2006; Whitehouse et al., 2017) plotting above the mantle zircon field and displaying
165 heterogeneities. Our model values also plot above the mantle zircon field, but they
166 display higher values, probably as a result of the elevated $\delta^{18}\text{O}_{\text{VSMOW}}$ value of the starting
167 serpentinite (9.7‰). Indeed, if the Hadean altered ultramafic protocrust was hydrated at
168 higher temperatures (>120 °C) than present-day oceanic peridotites, then $\delta^{18}\text{O}$ signatures
169 would be lower (Fig. 3; Table S5), yielding zircon modeled $\delta^{18}\text{O}$ values similar to the
170 JHZ. A testable hypothesis of involvement of seawater-altered serpentinite will be
171 relatively high $\Delta^{17}\text{O}$ signatures in the Hadean zircons (e.g., Bindeman,
172 2021).

173 The trace-element composition of the model zircons is similar to those of the ≥ 4
174 Ga JHZ (Cavosie et al., 2006; Turner et al., 2020) and the Green Sandstone Bed zircons
175 in the Jack Hills (Figs. 4A and 4B; Drabon et al., 2021). In detail, the REEs, Nb, Y, and
176 Hf contents are in agreement with those of the ≥ 4 Ga zircons, while there is nevertheless
177 a noticeable depletion in Th and U contents of the model zircon compared to the Hadean
178 ones. In this respect, we note that the starting experimental basalt was a tholeiite with
179 composition similar to that of Archean tholeiite. Nevertheless, the discrepancy between
180 model and Hadean zircons cannot be explained only by tapping a mantle reservoir with

181 primitive composition. It requires a contribution of an enriched KREEP-like reservoir,
182 corresponding to very late-stage liquids derived from magma ocean crystallization that
183 may reach Th contents ≥ 10 ppm (Snyder et al., 1995; Neal and Kramer, 2006). We note
184 that production of KREEP-like basalts at the onset of the Hadean Eon was also assumed
185 by Kemp et al. (2010). However, our model differs by stating that Earth's protocrust was
186 made of peridotites and not of KREEP-like basalts that remained under the protocrust.
187 From a geochemical perspective, the participation of 10% of KREEP-like basalts is
188 sufficient to explain the Th discrepancy (Fig. 4A), but it does not seem to fit the heaviest
189 REEs (Yb and Lu), which are slightly too high, resulting in lower U/Yb (Fig. 4B).
190 However, the strong partitioning of the **heavy (H)** REEs in amphibole (e.g., Sisson, 1994)
191 implies that crystallization of amphibole from the experimental felsic melt could offset
192 this effect on the HREE budget.

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

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

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342 **FIGURE CAPTIONS**

343 Figure 1. (A) Scheme of hybrid and mixed experiments in basalt (BAS)–serpentinite
344 (SERP) systems producing felsic glasses (FL), after Borisova et al. (2021). (B) Total
345 alkali oxides versus SiO₂ contents (in wt%) in felsic glasses produced during our
346 experiments (Tables S1–S2 ~~[see footnote~~  Diagram is after Le Maitre (2002). BA—
347 basaltic andesite.

348 **[[Figure edits: Fix spelling of interstitial in the legend in panel A. Put units in**
349 **parentheses in axis labels in panel B: (wt%).]]** 

350

351 Figure 2. Zircon saturation expressed in Zr contents (C_{sat} , ppm) versus temperature (T ,
352 °C) according to **Supplemental** equations 1 (Boehnke et al., 2013; Bindeman and Melnik,
353 2016), 2 (Harrison et al., 2007), and 3 (Watson and Harrison, 1983), and the **M factors** 
354 **[compositional factor $M = (\text{Na} + \text{K} + 2\text{Ca})/(\text{Al} \times \text{Si})$ (1.4–2.9) (see the Supplemental**
355 **Material; Table S4 [see footnote 1]). Dashed red horizontal lines correspond to minimum**
356 **(49 ppm) and maximum (110 ppm) Zr contents in experimental felsic glasses. Red field**
357 **indicates conditions (below 750 °C) of zircon crystallization above solidus (~700 °C).**

358 **[[Figure edits: Put units in parentheses in the axis labels: e.g., (ppm); (°C). Italicize**

359 **T (for temperature) and C (in C_{sat}).]]**

360

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

371 **[[Figure edits: Put units in parentheses in axis labels: e.g., (°C); (%). Italicize T (for**
372 **temperature) and C (in C_{sat}).]]** 

373

374 Figure 4. (A) Zircon composition normalized to median mid-ocean ridge (MOR) zircon.
375 (B) U/Yb versus Nb/Yb and Gd/Yb ratios. Median MOR zircon is after Grimes et al.
376 (2015). The model zircon composition was calculated at 700 °C. Gray **circles represent**
377 **the whole** **[[entire?]]** compositional range for Jack Hills zircon (JHZ): 4.37–4.02 Ga
378 type-1 crystals (Cavosie et al., 2006) and several ≥ 4 Ga Jack Hills crystals (Turner et al.,
379 2020). The result of 10% of KREEP addition to the parental basalt is shown (**where**
380 **KREEP indicates residuum with high abundance of incompatible elements such as K,**

381 rare earth elements [REEs], and P). Present-day zircon types, mantle array, and Green
382 Sandstone Bed (GSB) zircon composition are after Grimes et al. (2015) and Drabon et al.
383 (2021). Model data and references are given in Table S2 (see footnote 1).

384

385 ¹Supplemental Material. ~~[[Please provide a brief caption here.]]~~ Please visit
386 ~~<https://doi.org/10.1130/XXXXX>~~ to access the supplemental material, and contact
387 editing@geosociety.org with any questions.