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An impact melt origin for Earth's oldest known evolved rocks

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Earth's oldest evolved (felsic) rocks, the 4.02-billion-year-old Idiwhaa gneisses of the Acasta Gneiss Complex, northwest Canada, have compositions that are distinct from the felsic rocks that typify Earth's ancient continental nuclei, implying that they formed through a different process. Using phase equilibria and trace element modelling, we show that the Idiwhaa gneisses were produced by partial melting of iron-rich hydrated basaltic rocks (amphibolites) at very low pressures, equating to the uppermost ~3 km of a Hadean crust that was dominantly mafic in composition. The heat required for partial melting at such shallow levels is most easily explained through meteorite impacts. Hydrodynamic impact modelling shows not only that this scenario is physically plausible, but also that the region of shallow partial melting appropriate to formation of the Idiwhaa gneisses would have been widespread. Given the predicted high flux of meteorites in the late Hadean, impact melting may have been the predominant mechanism that generated Hadean felsic rocks.

The Hadean and earliest Archaean eons (~4.5 to 3.9 billion years ago (Ga)) witnessed a barrage of asteroid impacts that caused global-scale melting and recycling of Earth's surface[1–3]. Although ~3.7-Gyr-old rocks are known to have existed within most continental nuclei, and there are rare vestiges of still older Archaean crust[4], there is almost no crustal material preserved from the Hadean, Earth's formative eon.

Much of our understanding of the Hadean Earth comes from studies of tiny grains (<<1 mm) of the mineral zircon, which have radiometric ages that extend back to ~4.4 Ga (ref. [5]). The trace element and isotopic composition of Hadean zircons provide compelling evidence for the existence of surface water[5–7]. Although some also interpret chemical and mineral inclusion data from Hadean zircon as evidence for significant volumes of evolved (felsic) continental crust[8–10], others posit that Earth's crust in the Hadean was dominantly of basaltic (mafic) composition[11–13].

The only known felsic rocks of Hadean age occur within the Acasta Gneiss Complex (AGC), part of the Slave Craton of northwest Canada[14,15]. Its oldest dated components, the 4.02-Gyr-old Idiwhaa tonalite gneisses[11,16,17], are magnetite-rich felsic gneisses with chemical compositions clearly distinct from rocks of the tonalite–trondhjemite–granodiorite (TTG) suite that dominate exposed Archaean crust worldwide (Fig. 1)[18]. These compositional differences suggest a different petrogenesis[17]. The AGC also contains abundant amphibolite, hydrated mafic rocks from which the felsic rocks may have been derived through partial melting[11,19,20]. In particular, the Idiwhaa gneisses are spatially associated with Fe-rich amphibolites, which now contain garnet that formed during a subsequent episode of metamorphism at 3.65–3.60 Ga (refs [19,21]). Although the age of the mafic components of the AGC is unknown, some are intruded by felsic rocks and are considered to be Hadean in age[19,21].

Compared with TTGs (average SiO_2 = 69 wt%, Al_2O_3 ~15 wt%, average FeO_T = 2.7 wt%, where FeO_T = total Fe expressed as Fe^{2+}), which include most of the younger (3.93–2.94 Ga) Acasta felsic gneisses, the Idiwhaa gneisses range to lower silica contents (70–58 wt%) and Mg# (most are <20; Mg# = atomic Mg/(Fe + Mg)), marginally lower Al_2O_3 (~13 wt%) and much higher FeO_T (8–15 wt%)[17] (Fig. 1a,b and Supplementary Table 1). The high FeO contents, low Mg#, negative Sr and Eu anomalies, and flat rare earth element (REE) patterns of the Idiwhaa gneisses (Fig. 1c) are similar to rocks known as icelandites[22]. Like icelandites, the Idiwhaa gneisses are interpreted by some to record shallow-level fractional crystallization of low-H_2O ('dry') basaltic magmas in which plagioclase was a major fractionating phase[11,16]. However, others propose that icelandites were produced by partial melting of hydrothermally altered ('wet') basalt[23,24]. As the age of the Idiwhaa gneisses falls near the end of the period of intense meteorite bombardment, and impacts are an efficient means of melting target rocks, either partially or completely[1,2,12,25–30], an origin via impact-induced partial melting of hydrated mafic host rocks seems a plausible petrogenetic model.

Here we combine phase equilibria and trace element modelling with hydrodynamic simulations to test the hypothesis that the Idiwhaa gneisses had an origin through impact-induced partial melting. Our phase equilibria modelling uses thermodynamic models appropriate for the partial melting of mafic rocks[31], and allows calculation of the abundance and composition of minerals, melts and fluids as a function of pressure (P), temperature (T) and bulk composition. Trace element modelling uses these data and published mineral/melt partition coefficients[32] to predict the trace element composition of melt as a function of P, T and melt fraction (F). In modelling the petrogenesis of the Idiwhaa gneisses, we take the composition of an average Fe-rich amphibolite host rock from the Acasta gneisses[19] as a putative source (see Methods).

Phase equilibria modelling of the Idiwhaa gneisses

A simplified P–T phase diagram calculated in the range 0.05–0.80 GPa, 650–950 °C is shown in Fig. 2, on which calculated melt fractions (F, as mol%, ~vol%) are shown as red dotted lines (see Supplementary Fig. 1 for the full phase diagram). The temperature of the onset of partial melting (the H_2O-saturated solidus) decreases

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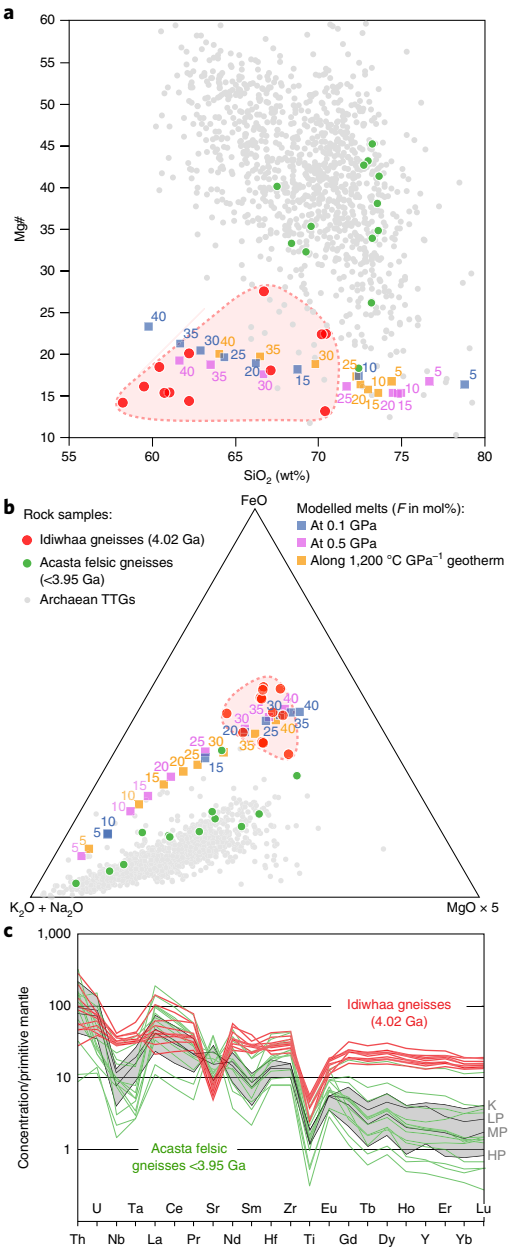

Fig. 1 | Major element oxide and trace element geochemistry. a, SiO_2 (wt%) versus Mg# (molar Mg/(Mg + Fe^{2+})). **b**, Ternary diagram with apices FeO, MgO and $K_2O + Na_2O$ (all wt%). The dots show the measured composition (all normalized to 100% in the NCKFMASTO (that is, anhydrous) compositional system) of rocks. Grey dots, Archaean TTGs and potassic rocks[18]; green dots, Acasta felsic gneisses <3.95 Ga in age[17]; red dots, the 4.02-Gyr-old Idiwhaa gneisses[17]. Squares show the composition of modelled melts at 0.1 GPa (blue squares), 0.5 GPa (pink squares) and along a 1,200 °C GPa^{-1} geotherm (yellow squares), for which the coloured numbers are melt fractions (*F*) in mol% (~vol%). **c**, Primitive mantle normalized trace element compositions of the Idiwhaa gneisses (red) and younger Acasta felsic gneisses (green). The grey field encompasses the average compositions of the different baric types (high (HP), medium (MP) and low (LP) pressure) of Archaean TTG, including potassic (K) rocks, worldwide[18].

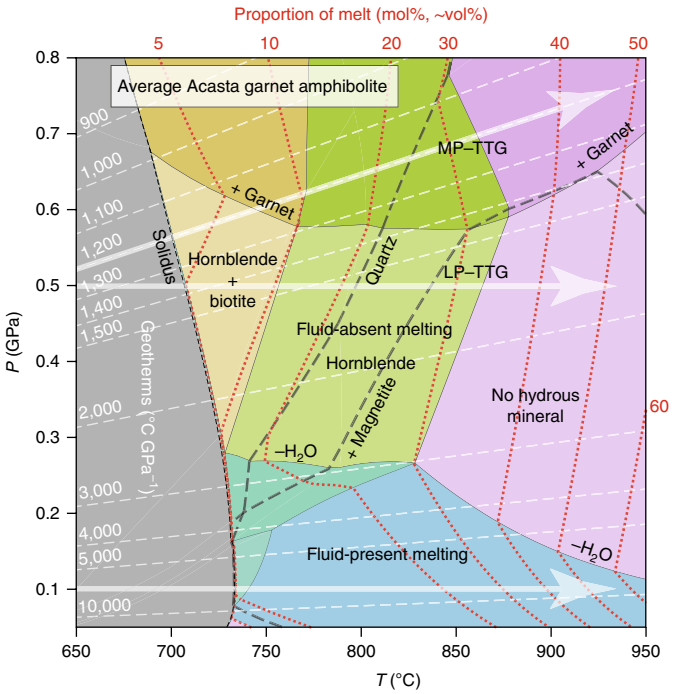

Fig. 2 | Phase equilibria modelling. Simplified *P*–*T* phase diagram for an average Acasta garnet amphibolite[19] ($n = 6$). The red dotted lines show the proportion of melt (mol% on a one oxide basis, ~vol%); the white dashed lines are linear geotherms (°C GPa^{-1}) assuming a surface *T* of 25 °C. Within the yellow fields, H_2O-undersaturated melting occurs by reactions consuming biotite and hornblende (±quartz) to produce granitic to granodioritic melts. Within the green fields, H_2O-undersaturated melting occurs by reactions consuming hornblende (±quartz) to produce low pressure TTG (LP-TTG) melts at $P < 0.6$ GPa and medium pressure TTG (MP-TTG) melts at higher *P*. The region within the blue fields is characterized by H_2O-saturated ('wet') melting mainly consuming plagioclase and orthopyroxene (and H_2O). The stability of garnet, quartz and magnetite are indicated. The three large white arrows show the modelled melting paths (isobaric at 0.1 GPa and 0.5 GPa and along a 1,200 °C GPa^{-1} geotherm).

from ~730 °C at low pressures (<0.1 GPa) to ~670 °C at 0.8 GPa (Fig. 2). Heating beyond the solidus at pressures above 0.3 GPa permits minimal fluid-present melting (< or <<10 mol%). Thereafter, partial melting proceeds by fluid-absent reactions consuming biotite, hornblende and quartz until biotite is exhausted at around 730–770 °C. Within this *P*–*T* region (shaded yellow on Fig. 2), melt compositions are granite evolving to granodiorite. Thereafter, fluid-absent melting reactions consume hornblende (with or without quartz; green shaded area) to produce melts with lower K_2O/Na_2O ratios typical of TTGs[33]. Within the *P*–*T* window modelled, low-pressure TTGs (in which garnet is not stable) are predicted at $P < 0.6$ GPa and medium pressure TTGs, in which garnet but not rutile is stable, at higher *P*.

At low pressure ($P < 0.27$ GPa) the system remains H_2O-saturated ('wet'; blue region in Fig. 2) to temperatures in excess of 800 °C, and to temperatures in excess of 1,000 °C at 0.1 GPa, corresponding to depths of around 3 km. At these low pressures, magnetite is ubiquitous, suprasolidus hornblende is absent and quartz is exhausted almost immediately as the solidus is crossed (Fig. 2 and Supplementary Fig. 1). Low-pressure melting proceeds by H_2O-saturated reactions consuming mainly plagioclase and orthopyroxene to produce melts that become successively depleted in SiO_2 and enriched in FeO and MgO as temperature (and *F*) increases.

The calculated major element composition of modelled melts (recalculated on an anhydrous basis) produced at melt fractions of 5–40 mol% (~vol%) along isobaric heating paths at 0.1 GPa (blue squares) and 0.5 GPa (pink squares) and along a geothermal gradient of 1,200 °C GPa^{-1} (orange squares) are shown in Fig. 1 (see also

Fig. 2 and Supplementary Fig. 1), and compared with the compositions of the Idiwhaa gneisses and younger Acasta felsic rocks (assuming $Fe^{3+}/\Sigma Fe = 0.1$). These three paths were chosen as proxies for potential impact-induced melting in the uppermost crust (depth of ~3 km) and mid/lower crust (depth of ~15 km), and for (non-impact) melting along a plausible early Archaean crustal geotherm[33,34], respectively.

Due to the higher thermal stability of quartz with increasing pressure (Fig. 2), melts at 0.5 GPa and along the 1,200 °C GPa^{-1} geotherm remain silica rich ($SiO_2 > 70$ wt%) until melt fractions exceed 25 mol%, whereas the 0.1 GPa melts fall below this value at melt fractions of 10–15 mol% (Fig. 1a). In each case, the Mg# of the melt increases with increasing F (and T). Compositionally the modelled melts are unlike Archaean TTGs and the younger Acasta felsic rocks[17], but match well with the Idiwhaa gneisses. In terms of SiO_2 and Mg#, the modelled melts are a good fit with the Idiwhaa gneisses at $F = 15$–35% at 0.1 GPa (produced at ~810–890 °C) and $F = 30$–40 mol% at 0.5 GPa and along the 1,200 °C GPa^{-1} geotherm (~850–900 °C) (Fig. 1a). A similar or slightly more restricted range in F (20–35 mol% at 0.1 GPa; 30–40 mol% at 0.5 GPa; 35–40 mol% along the geotherm), requiring $T > 850$ °C, is indicated based on the relative proportions of FeO, MgO and $K_2O + Na_2O$ (Fig. 1b).

Trace element modelling of the Idiwhaa gneisses

Using the calculated abundance of residual minerals formed in equilibrium with melt at different melt fractions (Supplementary Table 2), and mineral/melt partition coefficients appropriate for partial melting of hydrated mafic rocks[32] (Methods and Supplementary Table 3), we model the trace element composition of melts formed at 0.1 GPa, 0.5 GPa and along the 1,200 °C GPa^{-1} geotherm (Fig. 3). Relative to the measured composition of the Idiwhaa gneisses (pale blue field in Fig. 3), the modelled trace element compositions of melts produced at 0.5 GPa and along the 1,200 °C GPa^{-1} geotherm are depleted in Sr and in all but the lightest REE, and exhibit no pronounced Eu anomaly (Fig. 3). Other than at $F = 5$ mol%, melts modelled along the 1,200 °C GPa^{-1} geotherm show a pronounced depletion in heavy over middle REE due to the presence of garnet in the residuum (Fig. 3). By contrast, the modelled trace element composition of melts formed at 0.1 GPa at $F \geq 10$ mol% are an excellent match with the measured composition of the Idiwhaa gneisses.

Our modelled melts produced at 0.1 GPa show a good major element oxide and excellent trace element correspondence with the measured composition of the Idiwhaa gneisses. Thus, the Idiwhaa gneisses could have been the result of 20–35 mol% (~vol%) partial melting of hydrated mafic source rocks similar in composition to Fe-rich amphibolites within the AGC, in the uppermost few kilometres of a hydrated mafic crust, requiring temperatures of ~800–900 °C. The occurrence of Fe-rich (now garnet-bearing) amphibolite is restricted to the northeastern part of the AGC, broadly coincident with the Idiwhaa gneisses (Fig. 1 in ref. [19]). Such Fe-rich mafic rocks are consistent with protracted reworking of thick plateau-like ultramafic to mafic primary crust that is predicted if mantle temperature were significantly warmer than at present[33], and/or the modelled composition of the uppermost 15 km of a fractionated terrestrial magma ocean[35].

Hydrocode impact modelling

To test whether our predicted P and T conditions for formation of the Idiwhaa gneisses are consistent with impact-induced melting, we used the Simplified Arbitrary Lagrangian–Eulerian shock physics hydrodynamic code[36] to simulate impact crater formation. For example, the results of models considering a 10-km-diameter dunite projectile with velocities ranging from 12 to 17 km s^{-1} impacting a basaltic crust of variable thickness (20–40 km) and variable geothermal gradient (20–40 °C km^{-1}) and underlain by dunitic mantle are shown in Fig. 4 (see Methods and Supplementary Movies 1–8). These models show formation of a ~120-km-diameter crater and

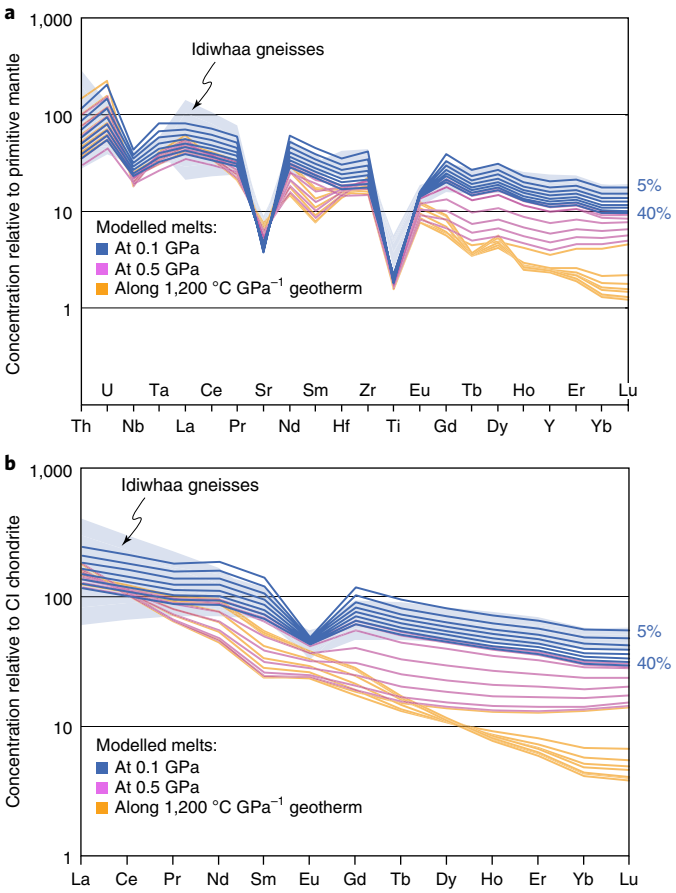

Fig. 3 | Trace element modelling. a,**b**, Primitive mantle normalized trace element (**a**) and chondrite normalized REE (**b**) compositions of modelled melts at 0.1 GPa (blue), 0.5 GPa (pink) and along a 1,200 °C GPa^{-1} geotherm (yellow) for melt fractions ranging from 5 to 40 mol%. In each case, the pale blue field shows the range of compositions of the Idiwhaa gneisses. For details see Methods.

a hot impact melt pool extending 10–15 km from the impact site. At greater distances, the models predict a layer at temperatures of 800–900 °C and at shallow depths (<3 km) extending up to ~50 km radial distance (Fig. 4). This layer is at conditions appropriate to the formation of the Idiwhaa gneisses according to our phase equilibria and trace element modelling.

Although no material with conditions appropriate for formation of the Idiwhaa gneisses is predicted in a model considering a projectile of 1 km diameter (that forms a ~15-km-diameter crater; Supplementary Fig. 2a and Supplementary Movies 9 and 10), the model for a 100-km-diameter projectile, which forms a crater ~1,000 km in diameter, predicts a shallow layer at temperatures between 800 and 900 °C extending between ~200 and 1,000 km radial distance from the impact site (Supplementary Fig. 2b and Supplementary Movies 11 and 12). Recent dynamic modelling of the terrestrial bombardment history suggested that >70% of Earth's surface experienced burial by impact-generated melt since 4.15 Gyr, with most impacts occurring ~4.15–3.8 Ga (ref. [1]). Our hydrodynamic modelling shows that, even in the waning stages of bombardment, layers of shallow partial melting appropriate for the formation of the Idiwhaa gneisses would have been extensive.

Petrogenesis of the Idiwhaa gneisses

The unusual major and trace element composition of the Idiwhaa gneisses has been interpreted to indicate they were formed by shallow

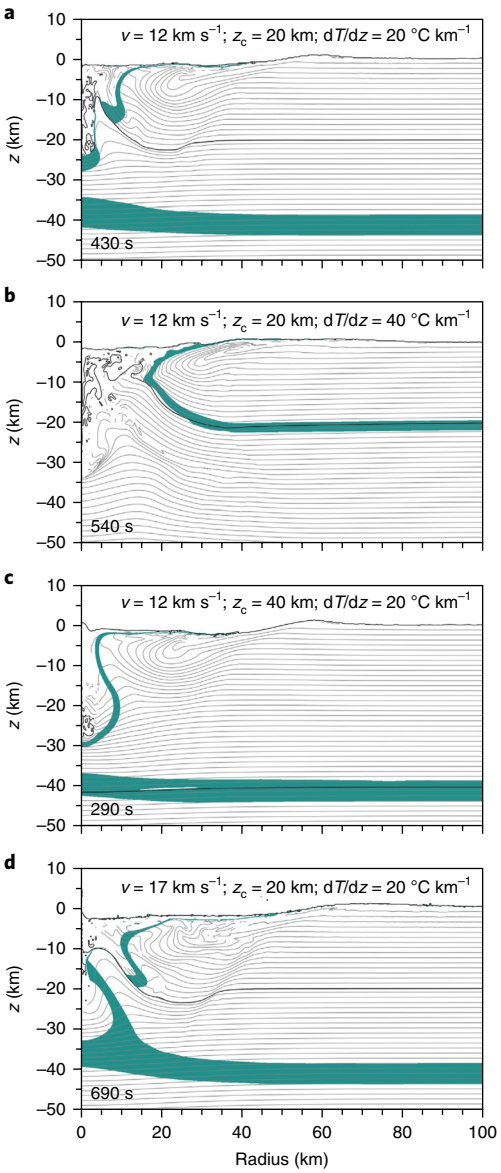

Fig. 4 | Hydrocode numerical simulation. a–d, The modelling considers formation of a Vredefort-like crater made by vertical impact of a 10-km-diameter projectile with a dunite composition travelling at a velocity (v) of 12 km s^{-1} (**a**–**c**) or 17 km s^{-1} (**d**), impacting basaltic crust with a thickness (z_c) of 20 km (**a**,**b**,**d**) or 40 km (**c**) that is underlain by dunite mantle. The target temperature gradient (dT/dz) was 20 °C km^{-1} (**a**,**c**,**d**) or 40 °C km^{-1} (**b**). The horizontal lines are simple stratigraphic markers. The blue areas show crust predicted to be at temperatures of 800–900 °C; those at shallow levels (<3 km) are plausible analogues of the Idiwhaa gneisses.

(low P) fractional crystallization of low-H_2O basaltic magmas, similar to petrogenetic models proposed for modern-day icelandites[16]. However, isotopic and trace element data suggest that an origin for icelandites through partial melting of hydrothermally altered basaltic rocks is also plausible[23,24,37], similar to our petrogenetic model for the Idiwhaa gneisses. Thus, we propose that Earth's oldest known evolved rocks formed through moderate degrees of partial melting of hydrated Fe-rich rocks in the uppermost few kilometres of a fractionated Hadean crust that was dominantly mafic[11,13,38]. Melting at such shallow levels requires temperatures of 800–900 °C or higher and geothermal gradients in excess of ~4,000 °C GPa^{-1} (Fig. 3). Such geotherms are much warmer than those recorded in regional metamorphic rocks of any age worldwide[34], and would require a source of heat beyond what is normally attainable in Earth's crust. We suggest that the required heat was delivered by meteorite impacts towards the end of the post-accretionary bombardment that characterized the first 600 Myr of Earth history[1,2]. The flux of meteorites bombarding the Earth during the late Hadean may mean that the predominant mechanism for generating felsic rocks during this time was through impact melting.

Methods

Methods, including statements of data availability and any associated accession codes and references, are available at https://doi.org/10.1038/s41561-018-0206-5.

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Author contributions

T.E.J. conceived the idea for the paper and did the phase equilibria modelling. T.E.J. and N.J.G. undertook the trace element modelling. K.M. performed the hydrodynamic impact modelling. T.E.J. wrote the manuscript draft. All authors contributed to discussions and the writing of the final paper.

Competing interests

The authors declare no competing interests.

Additional information

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Methods

Phase equilibria modelling. Phase equilibria were modelled in the Na_2O–CaO–K_2O–FeO–MgO–Al_2O_3–SiO_2–H_2O–TiO_2–O (NCKFMASHTO) chemical system using an average Acasta garnet amphibolite[19] ($n=6$) assuming an $Fe^{3+}/\Sigma Fe$ ratio of 0.1 (ref. [39]) and a H_2O content of 1.5 wt%. The composition, in terms of the mol% of the oxides Na_2O–CaO–K_2O–FeO–MgO–Al_2O_3–SiO_2–H_2O–TiO_2–O, is 2.365, 7.386, 0.405, 15.516, 5.520, 9.336, 51.192, 5.341, 2.167, 0.772. Calculations used THERMOCALC version 3.45i[40], the internally consistent thermodynamic dataset ds63[41] (updated 5 January 2015) and activity–composition solution models as follows: tonalitic melt, augite and hornblende[31] with a reduced DQF (Darken's quadratic formalism) value for the glaucophane end member of –3 kJ mol^{-1} from 0 kJ mol^{-1}; garnet, orthopyroxene, biotite and chlorite[42]; olivine and epidote[41]; magnetite–spinel[43]; ilmenite–haematite[44]; $C\bar{1}$ plagioclase and K-feldspar[45] and muscovite–paragonite with a reduced DQFvalue for the margarite end member of 5 kJ mol^{-1} from 6.5 kJ mol^{-1}. Quartz, rutile, sphene (titanite), and aqueous fluid (H_2O) are considered as pure end members. Results are shown in an isochemical P–T phase diagram (pseudosection) in the pressure range 0.05 to 0.80 GPa and the temperature range 650 to 950 °C (Supplementary Fig. 1).

The H_2O content in the modelled composition (1.5 wt%) is within the range inferred, based on loss on ignition from X-ray fluorescence analyses, for hydrated basaltic rocks sampled from drill core at depths of 2–3 km within hydrothermally altered oceanic crust in Iceland[46]. A quantity of 1.5 wt% H_2O produces minimal H_2O saturated melting (< or <<10 mol%) at $P>3$ kbar (Fig. 2 and Supplementary Fig. 1). Assemblage fields are labelled with stable phases, in which abbreviations are as follows: melt (L), garnet (g), augite (aug), orthopyroxene (opx), olivine (ol) hornblende (hb), biotite (bi), magnetite (mt), ilmenite (ilm), plagioclase (pl), K-feldspar (ksp), quartz (q), sphene = titanite (sph) and aqueous fluid (H_2O). All fields contain plagioclase and ilmenite. The depth of shading of assemblage fields reflects increasing variance. The highest variance fields, opx–hb–L (pl–ilm) and aug–opx–L (pl–ilm), have a variance of 7 (Supplementary Fig. 1). The software and data files used to generate the phase diagrams can be downloaded from http://www.metamorph.geo.uni-mainz.de/thermocalc.

Trace element modelling. Trace element modelling (Fig. 4) uses the average Acasta garnet amphibolite[19] as a starting composition and the calculated abundance of phases at melt fractions of 5–40 mol% along the three paths (0.1 GPa, 0.5 GPa and along a 1,200 °C GPa−1 linear geotherm; see Fig. 2, Supplementary Fig. 2 and Supplementary Table 2). Mineral/melt partition coefficients (Ds; Supplementary Table 3) follow Bédard[32], which were calculated for anatexis of metabasites[32], with a modified D_{Eu} for plagioclase of 1, based on the range of SiO_2 and MgO contents of the modelled melts[47], and a D_{Ti} for ilmenite of 150 (ref. [48]).

Numerical impact modelling. iSALE-2D (impact Simplified Arbitrary Lagrangian–Eulerian)[36,49,50] is a multi-material and multi-rheology finite difference shock physics hydrodynamic code (hydrocode) used for simulating impact processes in two dimensions. The code, which can be accessed at https://isale-code.github.io/, has been benchmarked against other hydrocodes[51]. A half-space target mesh was divided into two horizontal layers, the crust and the mantle, and a cylindrical symmetry was assumed. The numerical resolution was 20 cells per projectile radius, equating to a numerical cell size of 25 by 25 m for the model involving a 1 km projectile, 250 by 250 m for the model involving a 10 km projectile and 2.5 by 2.5 km for the model involving a 100 km projectile. The impactor speed was taken to be 12 or 17 km s^{-1}, to accommodate a wide distribution of expected impact speeds during the early terrestrial bombardment[52]––these impact speeds could also represent moderately oblique incidence angles.

The pre-impact crust thickness was fixed to either 20 or 40 km, and the temperature gradient was either 20 or 40 °C km^{-1}. The material models for the crust (basalt) and mantle and impactor (both dunite) use the appropriate ANEOS-derived equation of state (EOS) tables, as well as the strength[49], failure and thermal softening models that were used in previous studies for modelling large crater formation on the Moon[53,54] and Mars[55] (Supplementary Table 4). Simulations employ the block-oscillation model of acoustic fluidization to facilitate crater collapse[56]. The role of temperature is restricted to its effect on the shear strength. The material shear strength model in iSALE includes a description of thermal softening: $Y = Y_c \tanh[\varepsilon(T_m/T-1)]$, where Y_c is the cold shear strength, ε is a material constant, T is temperature and T_m is the melting temperature. We use the Simon approximation to fit the pressure dependence for the melting temperature of anhydrous basalt and KTB peridotite[56] for the crust and mantle, respectively: $T_m = T_{m,0}\,(P/a+1)^{1/c}$, where $T_{m,0}$ is the melt temperature at zero pressure, P is pressure, and a and c are material constants. The input parameters to our simulations are provided in Supplementary Table 4.

On the Earth, the 10 km projectile is predicted to form a peak-ring crater that is approximately 120 km diameter rim-to-rim. This is comparable to the size of the Vredefort crater[57], the largest confirmed terrestrial impact structure. In Fig. 4 (and Supplementary Fig. 2), horizontal lines show the structural morphology of the crater. Modest structural uplift is present within 20 km radial distance from the impact point. Structural crustal overturn of the top 5–10 km of crustal material is predicted to occur at 20–40 km radial distance. The blue zone denotes material at a temperature of 800–900 °C. The pre-impact horizontal blue layer at 20 or 40 km depth is a consequence of the imposed geothermal gradient. However, the blue zone occurring within the crater is a result of impact heating of the target. Specifically, the blue zone within the top 5 km of the crust, which is considered a good proxy for the Idiwhaa gneisses based on phase equilibria and trace element modelling, is a consequence of the process of peak-ring formation during the final phase of crater collapse. The diameter of the peak ring is about 60–80 km, in which material at shallow levels and at temperatures of 800–900 °C are predicted to occur along the bulk of its radial distance (Fig. 4a,b,d and, to a lesser extent, Fig. 4c).

Recent numerical modelling of terrestrial bombardment during the Hadean eon suggested that no substantial large region of the Earth's surface could have survived untouched by impact bombardment[1]. While the early Hadean eon is suggested to have experienced the largest impact bombardment[1], the later Hadean and early Archaean eons should have experienced a significant fraction of the so-called late heavy bombardment.

Code availability. The code used for simulating impact processes in 2-D can be accessed at: https://isale-code.github.io/. The software and data files used to generate the phase diagrams can be downloaded from: http://www.metamorph.geo.uni-mainz.de/thermocalc.

Data availability. All data supporting the findings of this study are available within the paper, Methods and Supplementary Information.

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