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Supplementary Materials for

Titanium isotopes constrain a magmatic transition at the Hadean-Archean boundary in the Acasta Gneiss Complex

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Supplementary Materials
Iron isotopic fractionation

Interpretation of Fe isotope variations in igneous rocks is complicated by the fact that two Fe oxidation states often coexist in magmas. The decrease in δ^{56}Fe values with increasing SiO_2 measured in the 4.02 Ga ITG is reminiscent of what was observed by Sossi et al. (*69*) in the tholeiitic differentiation series found in the Red Hill intrusion, Tasmania (Fig. S1). We interpret the Fe isotopic evolution of the ITG samples to possibly represent the post-magnetite crystallization limb of the magmatic differentiation trend analogous to that documented in the Red Hill suite, however small variations within measurement error make this interpretation difficult.

Following the interpretative framework of Sossi et al. (*69)* and Dauphas et al. (*70*), the heavy Fe isotope enrichments with increasing SiO_2 documented in the <3.9 Ga AGC TTGs could reflect crystallization in a system that was open to exchange with the surrounding medium such that buffering of oxygen fugacity could have mitigated the increase in Fe^{3+}/Fe_{tot} expected for closed-system crystallization. In that context, the oxides that crystallized could have been characterized by lower Fe^{3+}/Fe_{tot} than those that crystallized in the 4.02 Ga ITG. The study of Roskosz et al. (*71*) on spinels shows that the redox state of Fe in oxides has a strong influence on the strength of Fe bonds and Fe isotopic fractionation. It is thus conceivable that crystallization of low-Fe^{3+} oxides, in concert with the crystallization of other Fe^{2+}-bearing phases, drove the residual magma towards heavy isotope enrichments.

Figure S1b illustrates how the ITG and AGC samples may be defined by different trends in δ^{56}Fe *vs.* δ^{49}Ti, however small δ^{56}Fe variations make this interpretation ambiguous. As discussed above, these trends could reflect the different paths of Fe^{3+}/Fe^{2+} evolution experienced by the parental magmas during magmatic differentiation. Data is missing at the present time to quantitatively model those trends but the systematics revealed by the present study illustrate the potential of combining Fe and Ti isotopic analyses to tease out the oxide crystallization history of igneous rocks.

Age and geochemical history of Acasta Gneiss Complex

The AGC has been extensively geochemically characterized (*21*), and dated using U-Pb isotope methods, with ages ranging from 4.03-3.4 Ga *(27, 72-74)*. Reimink et al. (*22*) argued that the AGC saw a transition from shallow level melting of hydrated mafic crust at 3.96-3.75 Ga to greater melting depths at ~3.6 Ga (*23*). The ~3.6 Ga granitoids were believed to be the result of thrusting beneath a Hadean mafic plateau and subsequent melting at depths >50 km (*26*). These constraints on the formation depth are difficult to reconcile without invoking a horizontal tectonic regime (*22*).

Recently Reimink et al. (*22*) utilized whole-rock $^{142}Nd/^{144}Nd$ ($\mu^{142}Nd$) ratios to infer that the 4.02 Ga crust was extracted from a bulk-Earth source whereas the Eoarchean rocks from the AGC were interpreted to be the result of partial melting of a hydrated, Hadean age mafic crust at relatively shallow depths. By 3.6 Ga, granitic-rocks formed from partial melting of hydrated Archean mafic crust at increased depths within the garnet stability field. The $\mu^{142}Nd$ signatures *(22, 75)* and zircon Hf isotope work *(24, 25, 76-78)* ruled out the hypothesis of gradual thickening and melting of an ancient mafic plateau previously hypothesized in Reimink et al. (*23*), and supports the idea that the felsic crust observed here at 3.75 Ga was produced via plate subduction rather than melting of hydrated basaltic material at the base of a thickened crust (*79*).

Based on geochemical data and U-Pb geochronology, the AGC is identified to have undergone six major episodes of magmatism at 4.02, 3.96, 3.75, 3.55-3.6, 3.37, and 2.94 Ga (*26*). The oldest evolved silicic rocks are the Įdì whaà tonalitic gneiss (4.02 Ga), which formed by shallow fractional crystallization of a basaltic magma. Hafnium isotope data from the 4.02 samples in the AGC show no indication of derivation or interaction with an older Hadean continental crust, and instead suggest that the oldest evolved crust from the AGC was generated from an older ultramafic or mafic reservoir that was present on the surface of the early Earth (*10*).

The later granitoids (3.96-2.94 Ga) are thought to have formed *via* partial melting of mafic rocks that underwent alteration from surface waters at low temperatures (*10*). The dehydration melting process is thought to have occurred at progressively deeper levels reaching the stability field of garnet at ~3.6 Ga at a depth of at least 50 km, and may have occurred in a subduction-like setting with tectonic under-thrusting or by burial or gravitational sinking of a thick oceanic plateau comprised of hydrated mafic rocks to depths of >50 km (*10*).

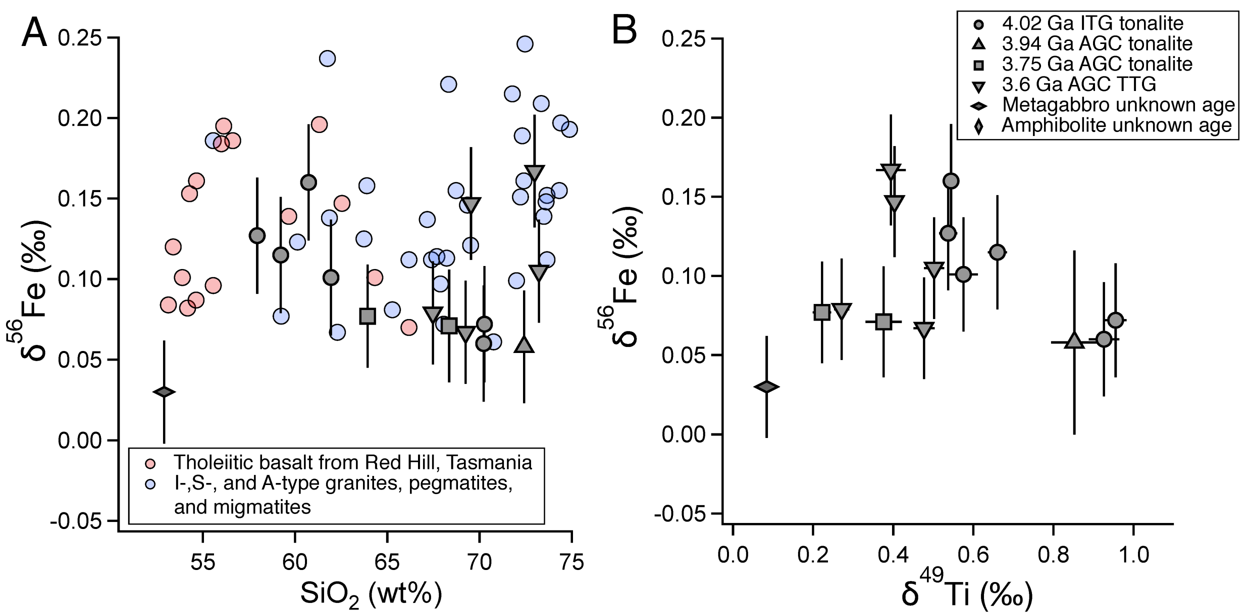

Fig. S1. δ^{56}Fe compositions as a function of SiO_2 content and the relationship of δ^{49}Ti with respect to δ^{56}Fe. Samples from the AGC are plotted as gray symbols in **a** and **b**. **a)** δ^{56}Fe compositions vs. SiO_2 of rocks from Įdì whaà tonalite gneiss (this study) and Red Hill, Tasmania (*69*) and I-, S- and A-type granites, pegmatites, and migmatites (*80*). Plume-derived rocks show a rapid depletion in δ^{56}Fe composition, whereas rocks from convergent settings slightly increase in δ^{56}Fe composition. **b)** δ^{56}Fe and δ^{49}Ti isotopic compositions of rocks from AGC.

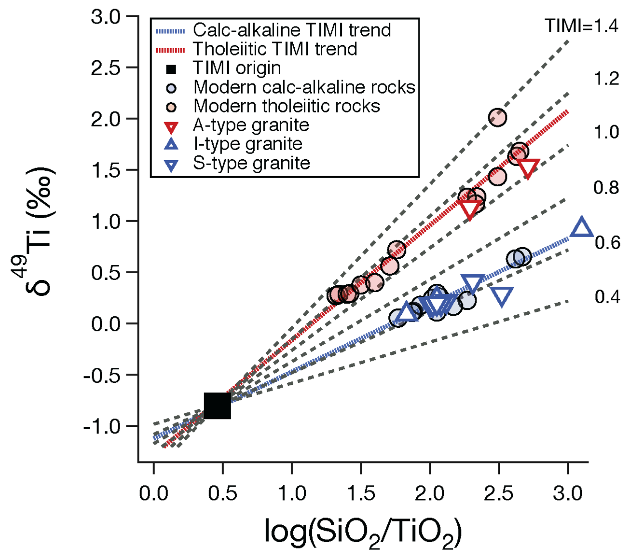

Fig. S2. Calculated TIMI for plutonic rocks and igneous rocks. Calc-alkaline rocks (blue circles) from Agung and Kos (*29, 36*), tholeiitic rocks (red circles) from Iceland, Afar and Hawaii (*30, 31*), A-type (this study) and I-and S-type granites (*36*).

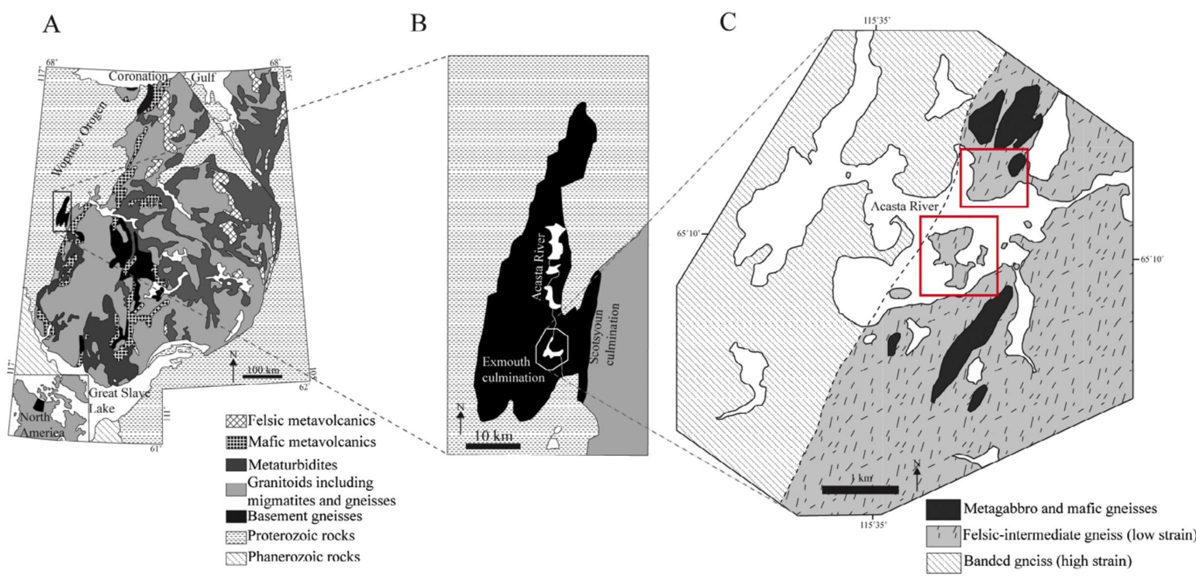

Fig. S3. Map of Acasta Gneiss Complex region modified from (*26*). a) Map of Slave Province with location of AGC noted. b) A close up view of the ~1300 km^2 exposure that encompasses the AGC. c) Simplified geologic map of the area highlighted in b).

Dataset S1 (separate file). Titanium isotope compositions and sample information for rocks analyzed in this study and plotted in Figs. 1, 2, and 3 in main text.

Dataset S2 (separate file). Iron isotope compositions and sample information for rocks analyzed in this study and plotted in Fig. S1.

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