

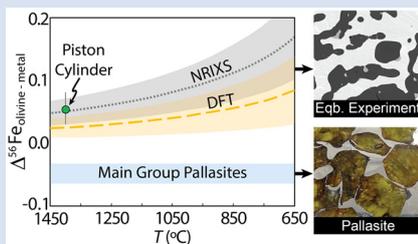
Iron isotope evidence of an impact origin for main-group pallasites

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Abstract



Despite decades of work, the origin of pallasite meteorites has remained enigmatic. Long thought to be samples of the core-mantle boundary of differentiated asteroids, more recent studies have suggested a range of mechanisms for pallasite formation. These include olivine-metal mixing during a planetesimal collision and the intrusion of over-pressured core liquids into a planetesimal mantle. Establishing if the olivine and metal that comprise pallasites were once equilibrated at high temperature remains key to discriminating between these hypotheses. To this end, we determined the iron isotope compositions of olivine and metal in eleven main-group pallasites and found, in all cases, that olivine is isotopically lighter than metal. To interpret these data, we constrained the olivine-metal equilibrium Fe isotope fractionation with *ab initio* calculations and high temperature experiments. These independent approaches show that olivine preferentially incorporates the heavy isotopes of iron relative to metal. Our results demonstrate that pallasitic olivine and metal never achieved isotopic equilibrium with respect to iron. This precludes extended cooling at high temperature and is best reconciled with an impact origin for the main-group pallasites.

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Introduction

Pallasites are predominately composed of Fe-Ni metal and olivine. Traditionally, they are interpreted to be remnants of core-mantle boundary material from differentiated asteroids, an idea which continues to merit discussion (e.g., McKibbin *et al.*, 2019). Alternatively, pallasites may form in the shallow mantle of an asteroid either by ferrovulcanism (Johnson *et al.*, 2020), impact-induced local heating and igneous differentiation (Mittlefehldt, 1980), or injection of exogeneous metal into the mantle of the pallasite parent body (Tarduno *et al.*, 2012). One way to distinguish these varied formation mechanisms is by constraining the thermal history of pallasites. A core-mantle boundary origin suggests a protracted period of cooling at high temperature, during which elemental and isotopic equilibrium between phases would be achieved. In contrast, a shallow mantle or impact mixing origin may be accompanied by more rapid cooling from high temperature, thus limiting re-equilibration. The low temperature thermal history of pallasite meteorites is well constrained by metallographically determined cooling rates, however, there remains little information on their high temperature thermal history. Here, we constrain this high temperature history by combining 1) Fe isotope measurements of main-group pallasites (MGP), 2) the equilibrium olivine-metal Fe isotope fractionation determined from high temperature experiments and *ab initio* calculations, and 3) models of planetesimal cooling and olivine-metal equilibration.

Several studies have attempted to determine equilibration temperatures for pallasites from the apparent partitioning of elements between olivine and metal (e.g., Seifert *et al.*, 1988). However, these studies assume equilibrium rather than test for it. Iron stable isotope data measured in olivine and metal have also been evaluated previously for geothermometry, but the results are highly variable (Poitrasson *et al.*, 2005; Weyer *et al.*, 2005; Chernozhkin *et al.*, 2016). These large variations result from unrepresentative sampling of the metal, which is composed of taenite (face-centred cubic or fcc Fe-Ni) and kamacite (body-centred cubic or bcc Fe-Ni) that form Widmanstätten patterns. At high temperature, Fe-Ni alloy is present with an fcc crystal structure (Yang *et al.*, 2010). As temperature decreases, this phase unmixes into coexisting Fe-rich and Fe-poor metallic phases; kamacite and taenite, respectively. Kamacite growth is diffusion-limited by the supply of iron from taenite (Dauphas, 2007). As the light isotopes of iron diffuse faster, kamacite is slightly enriched in light isotopes while heavy isotopes concentrate in taenite. Iron diffusion in taenite is 3 orders of magnitude slower than in kamacite. Taenite therefore retains significant gradients in isotope composition. As a result, preferential sampling of taenite will displace the Fe isotope composition to heavy values. In contrast, kamacite contains the bulk (>85 %) of the metallic Fe in pallasites and diffusion within kamacite is sufficiently rapid to homogenise Fe isotopes (Dauphas, 2007). The result is that kamacite closely approximates the bulk metal isotopic composition. Accordingly, we sampled kamacite to

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obtain a representative measurement of the bulk metal Fe isotope compositions of pallasite meteorites.

Sound interpretation of this Fe isotope data relies on access to a robust olivine-metal fractionation factor. However, the direction of fractionation is debated (Polyakov and Mineev, 2000; Dauphas *et al.*, 2014), complicating the interpretation of data collected on natural samples. To resolve this issue, we employed multiple techniques to determine Fe isotope fractionation between olivine and metal. As different metal phases are present in the pallasites, it is imperative to determine which phase could have equilibrated with olivine. Although the metal phase presently in contact with olivine is kamacite (bcc-Fe), olivine-metal isotopic equilibrium could not have been achieved in the stability field of kamacite. For the mean Ni contents of MGP metal (10.6 wt. % Ni; Wasson and Choi, 2003), kamacite formation begins at ~650 °C. For an average pallasite cooling rate of 7 °C/Myr (Yang *et al.*, 2010) and a median olivine diameter of 4 mm (Wasson and Choi, 2003), the closure temperature for iron in olivine is also ~650 °C (see Supplementary Information). Accordingly, if there were any chance of equilibration between olivine and metal it would have occurred when olivine was in contact with a single fcc-metal phase or metallic liquid (*i.e.* at higher temperatures). Therefore, our *ab initio* calculations and equilibration experiments focused on fcc-Fe and liquid metal phases.

Results and Discussion

Eleven MGP were analysed by micro-drilling kamacite and adjacent olivine crystals (Fig. S-1). To illustrate the range of data that can be obtained by untargeted sampling, metal and olivine were also handpicked and processed. All samples were analysed for their Fe isotope compositions by multi-collector inductively coupled plasma mass spectrometry (MC-ICPMS). Iron isotope compositions are reported in δ notation:

$$\delta^{56}\text{Fe}_{\text{smp}}(\text{‰}) = 1000 \times \left(\frac{{}^{56}\text{Fe}/{}^{54}\text{Fe}_{\text{smp}}}{{}^{56}\text{Fe}/{}^{54}\text{Fe}_{\text{std}}} - 1 \right) \quad \text{Eq. 1}$$

where smp denotes the sample (*i.e.* bulk metal, olivine) and std denotes the Fe isotopic standard IRMM-524a. Isotopic fractionations are expressed in Δ notation:

$$\Delta^{56}\text{Fe}_{\text{olivine-metal}} = \delta^{56}\text{Fe}_{\text{olivine}} - \delta^{56}\text{Fe}_{\text{metal}} \quad \text{Eq. 2}$$

In all eleven MGP, the olivine fractions are consistently lighter than the metal fractions (Fig. 1). The range of measured isotope fractionation is reduced by a factor of ~3 compared to literature data (Fig. S-2). Data for our handpicked samples reproduced the range of $\Delta^{56}\text{Fe}_{\text{olivine-metal}}$ obtained in previous studies, showing that the variability in $\Delta^{56}\text{Fe}_{\text{olivine-metal}}$ reported in previous studies reflects biased sampling of the metal fractions. Based on the rationale that all MGP come from the same parent body (Greenwood *et al.*, 2015), we report the weighted average $\Delta^{56}\text{Fe}_{\text{olivine-metal}}$ of $-0.049 \text{‰} \pm 0.016 \text{‰}$.

Using density functional theory (DFT), we calculated force constants for the iron sublattices in olivine ($159 \pm 10 \text{ N/m}$) and fcc-metal ($134 \pm 6 \text{ N/m}$). Due to difficulty in modelling the magnetic structure of taenite, the fcc-metal modelled here is awaruite (FeNi_3). To evaluate the effect of Ni on the force constant for Fe in fcc-metal, tetrataenite (FeNi), which has an fcc-like structure (Clarke and Scott, 1980), was also modelled. The force constant for tetrataenite is $142 \pm 6 \text{ N/m}$, which is slightly higher but identical within error to awaruite, suggesting that Ni has no significant effect on the force constant of iron in fcc-metal. As olivine has a larger force constant, $\Delta^{56}\text{Fe}_{\text{olivine-metal}}$ is always positive.

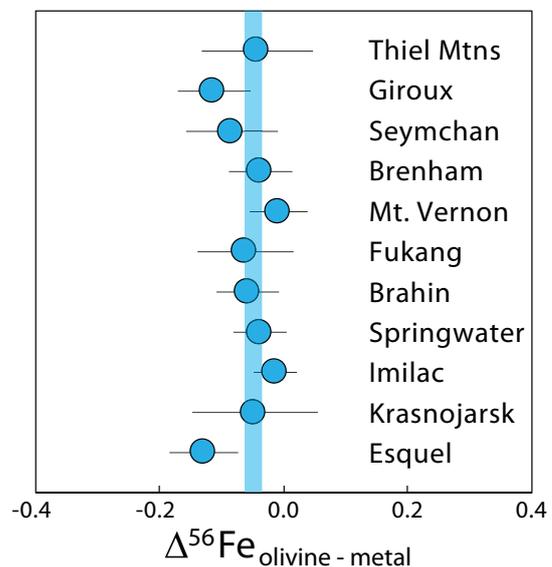


Figure 1 Measured Fe isotope fractionations between olivine and bulk metal in MGP. The blue band shows the weighted average for the 11 MGP measured in this study, plotted in order of cooling rate (Esquel slowest, Giroux fastest and Thiel Mtns. unknown; Yang *et al.*, 2010).

To provide another independent constraint on $\Delta^{56}\text{Fe}_{\text{olivine-metal}}$, we performed piston cylinder experiments that directly equilibrate olivine and Fe \pm Ni metal at high temperature. Olivine-metal assemblages were enclosed in graphite capsules, pressurised to 1 GPa, then heated to 1400 °C for 3.5–95 hr before quenching. These conditions are similar to those estimated for the core-mantle boundary of the pallasite parent body (1330 °C; Wasson and Choi, 2003, and ~0.1 GPa). Although our experimental pressure is higher than estimated for pallasites, Clayton *et al.* (1975) showed that between 0.1 and 2.0 GPa, pressure has no effect on equilibrium isotope fractionation factors. The bonding environment for molten metal in these experiments is similar to that of fcc-metal (see Supplementary Information). As with our DFT calculations, we did not observe an effect of Ni on the olivine-metal Fe isotope fractionation. The weighted average of three equilibrated experiments yields $\Delta^{56}\text{Fe}_{\text{olivine-metal}} = +0.054 \pm 0.027 \text{‰}$ at 1400 °C. This value is in excellent agreement with our DFT calculations, as well as the prediction made using nuclear resonant inelastic X-ray scattering data (NRIXS; Dauphas *et al.*, 2014; Krawczynski *et al.*, 2014).

To our knowledge, this is the first time that a single Fe isotope fractionation factor has been corroborated using all three of these independent approaches. We are thus confident that at conditions relevant to asteroidal core formation, heavy Fe isotopes preferentially partition into olivine relative to metal at equilibrium. The data obtained for MGP, however, show the opposite sense of fractionation (Fig. 2), suggesting that coexisting olivine and metal in pallasites never achieved Fe isotope equilibrium. Kinetic and equilibrium processes occurring at low temperature may potentially overprint high temperature equilibrium isotopic signatures. These processes, however, cannot reverse the direction of olivine-metal Fe isotope fractionation (see Supplementary Information).

Having established that olivine and metal are not in Fe isotope equilibrium, it is possible to determine parent body conditions where olivine-metal annealing could have taken place without leading to isotopic equilibrium. We coupled a 1 D

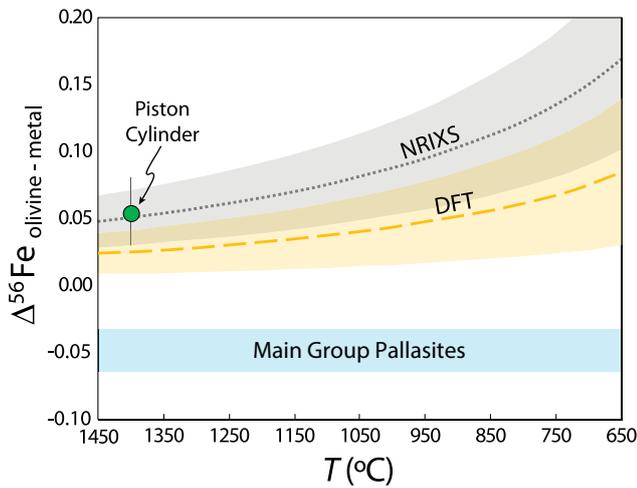


Figure 2 Grey and yellow bands show predicted olivine-metal Fe isotope fractionations at equilibrium, based on NRIXS and our DFT calculations, respectively. The piston cylinder datum is the weighted average of three isotopically equilibrated laboratory experiments. All methods show the same direction of equilibrium olivine-metal fractionation. MGP display the opposite sense of fractionation, suggesting that MGP olivine and metal are not in Fe isotopic equilibrium.

planetesimal cooling model with simultaneous diffusive Fe exchange between olivine and metal to place constraints on the depth and timing of olivine-metal mixing in the MGP parent body (see [Supplementary Information](#)). [Figure 3a](#) displays the results of one of these models, which was performed at conditions that also satisfy previous palaeomagnetic constraints on MGP. The black dashed curve marks the depth and timing of olivine-metal mixing above which Fe isotopic disequilibrium would be maintained. The key takeaway from this model is that even for late-stage olivine-metal mixing, core-mantle boundary

temperatures remain too high to prevent isotopic equilibrium. This result persists for a wide range of core diameters and mantle thicknesses ([Fig. 3b](#)). Our modelling therefore demonstrates that a core-mantle boundary, or other deep mantle, origin for pallasites is not compatible with the observed Fe isotope disequilibrium.

We next turn our attention to pallasite formation models that allow for a shallow to mid-mantle origin for MGP: 1) ferro-volcanism ([Fig. 4a](#); [Johnson et al., 2020](#)), and 2) impact-induced mixing of olivine and metal on the MGP parent body, involving either an endogenous (e.g., [Mittlefehldt, 1980](#)) or exogenous (e.g., [Kruijer et al., 2022](#); [Windmill et al., 2022](#)) source of metal ([Fig. 4b,c](#)).

[Johnson et al. \(2020\)](#) proposed that pallasites form when evolved, over-pressured, core liquids intrude the overlying mantle and entrain olivine in the process. The required over-pressure is facilitated by light element enriched (*i.e.* more buoyant) core liquid compositions. Light element (namely S) enrichment of planetesimal core liquids is achieved *via* fractional crystallisation. [Ni et al. \(2020\)](#) demonstrated that fractional crystallisation of Fe-S alloys is accompanied by enrichment of the S-rich liquid in light Fe isotopes. The heavy Fe isotope compositions of the bulk metals measured in this study ([Fig. S-2](#)) are not indicative of an evolved S-rich liquid, and thus do not lend support for the ferro-volcanism model.

An alternative is impact-induced mixing of endogenous olivine and metal reservoirs without complete parent body disruption. An impact into a cold body can raise the local temperature sufficiently to facilitate melting if the bodies have high porosities ([Davison et al., 2012](#)). Partial melting of a chondritic composition requires high temperatures (> 1100 °C; [Jurewicz et al., 1993](#)), that must be sustained long enough to permit igneous differentiation into discrete metal and silicate (olivine) reservoirs that are not isotopically equilibrated with each other. These reservoirs must then be mixed and cooled quickly enough to prevent isotopic re-equilibration. The plausibility of this

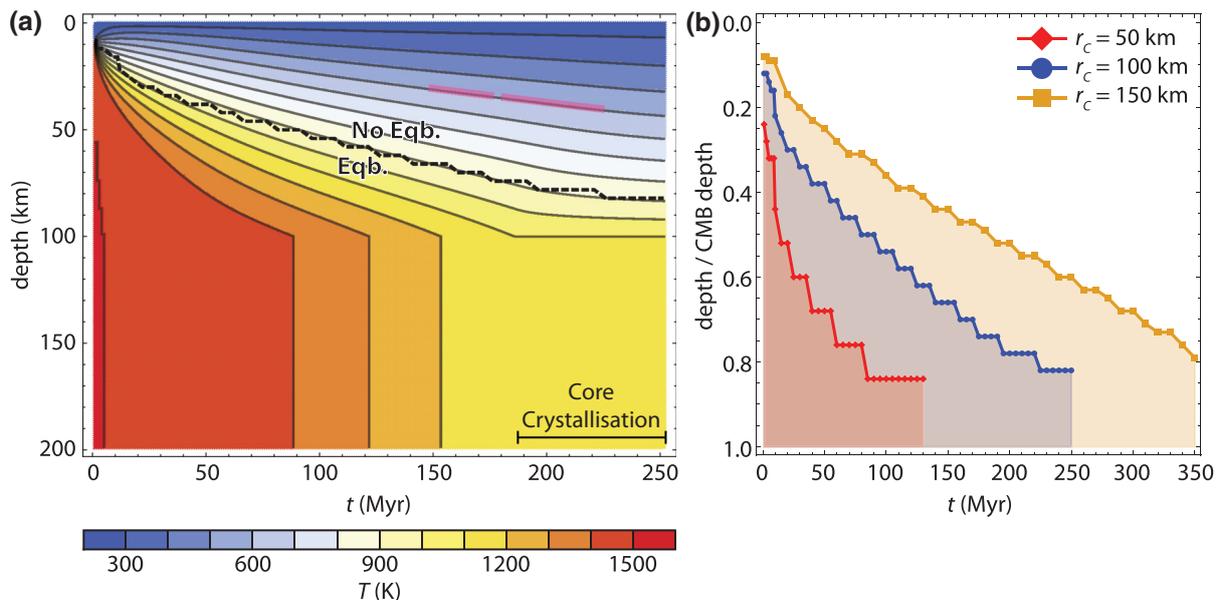


Figure 3 (a) Cooling and olivine-metal mass exchange model for a 200 km radius body with the core mantle boundary located at 100 km depth. The black dashed curve shows the time and depth of olivine-metal mixing above which Fe isotope equilibrium is inhibited. MGP formed above this curve. Also shown are the emplacement depths and periods of cloudy zone formation for the Imilac and Esquel meteorites, determined following [Bryson et al. \(2015\)](#). (b) Equilibrium curves for the same planetary radius but with different core radii (r_c). The shaded regions indicate where olivine-metal equilibrium will occur for each model. Regardless of core size, olivine and metal always equilibrate when mixed near the core-mantle boundary. Note that in all cases models are terminated once the core is fully crystallised.

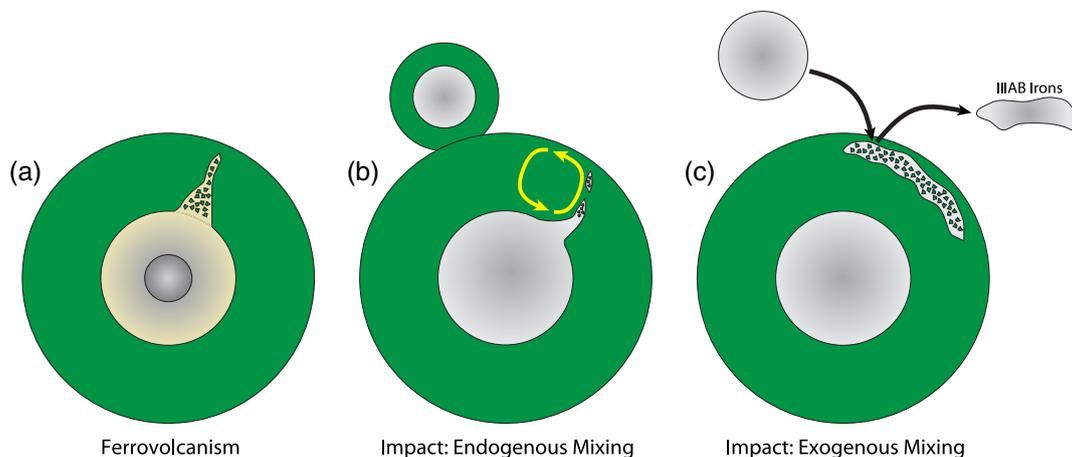


Figure 4 MGP formation models that account for shallow depths of olivine-metal mixing. Our Fe isotope data are most readily explained by model (c).

scenario is difficult to assess, particularly given our limited knowledge of the mineral-melt Fe isotope fractionation factors that control igneous differentiation. Measurements of O isotopes in pallasites, however, suggest that olivine and metal were sourced from different planetesimal bodies, precluding such an endogenous origin (Windmill *et al.*, 2022).

Kruijjer *et al.* (2022) provided W and Mo isotopic evidence that the metal from MGP is genetically related to the IIIAB irons, verifying an idea that originated from the trace element compositions of these meteorite groups (Scott, 1977). Furthermore, Yang *et al.* (2010) showed that IIIAB irons cooled significantly faster than MGP. These observations can be explained if the IIIAB core collided with the MGP parent body in a hit-and-run collision, leading to the rapid cooling of run-away metal that was exposed to space (Fig. 4c). This scenario also accommodates olivine-metal mixing in the shallow to mid-mantle of the MGP parent body, where Fe isotope equilibration between these phases is inhibited. Emplacement at these depths also permits MGP metal to cool below its blocking temperature before complete solidification of the parent body core and concomitant extinction of the dynamo (Tarduno *et al.*, 2012; Bryson *et al.*, 2015; Nichols *et al.*, 2021).

The subsequent slow cooling phase, as required by MGP metallographic cooling rates (3–18 °C/Myr; Yang *et al.*, 2010), must begin at a temperature below ~650 °C – low enough to inhibit iron diffusion in olivine, but high enough for Widmanstätten patterns to develop. While this two stage cooling history for MGP has previously been suggested on the basis of deformation experiments (Walte *et al.*, 2020), our study provides the corroborating geochemical evidence needed to cement this hypothesis and an impact origin for MGP.

Conclusion

There now exists multiple lines of evidence that MGP did not form near a core-mantle boundary: 1) The genetic link between IIIAB irons and MGP metals, and their difference in cooling rates, can only be explained by the impact of the IIIAB core into the MGP parent body, 2) palaeomagnetic data requires MGP metal crystallisation during an extant core dynamo, and 3) the Fe isotope data presented here demonstrate that olivine and metal are not in isotopic equilibrium, requiring rapid cooling to the olivine closure temperature. Our iron isotope measurements on pallasites, coupled with new constraints on the olivine-metal fractionation factor, demonstrate that pallasites

originated from shallow to mid-mantle depths in the parent body. Our findings support a growing consensus that main-group pallasites formed *via* an impact that provided an exogenous source of metal.

Author Contributions

NRB, CKS, and AS conceived the study. NRB performed the piston cylinder experiments, electron microprobe analyses, and coupled thermal and diffusion modelling. CKS performed the ICPMS and MC-ICPMS analyses and guided the diffusion calculations. ES performed the DFT calculations and wrote the relevant methods section. NRB and CKS wrote the manuscript with input from all co-authors.

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Additional Information

Supplementary Information accompanies this letter at <https://www.geochemicalperspectivesletters.org/article2229>.



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