Title: Olivine grain growth in partially molten Fe-Ni-S: A proxy for the genesis of pallasite meteorites

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Abstract:

The origin of pallasites has been the focus of a number of recent studies. Yet, their formation process remains elusive, while the mechanism leading to the genesis of the sub-group termed 'mixed type' pallasites (containing polygonal, rounded, and fragmental olivines simultaneously) is unclear. Here we test the hypothesis of mixing of olivine fragments with Fe-Ni-S after a non-destructive impact followed by annealing employing both experimental analogues and numerical models.

The experimental series evidenced that the addition of sulfur to olivine + Fe-Ni accelerates olivine grain growth, though the growth rate is reduced when Fe-Ni-S is not fully molten. This is shown to be the consequence of competing growth of olivine and Fe-Ni grains.

Numerical models satisfying available formation constraints from natural samples indicate that planetesimals with radii ≥200 km are favorable for the genesis of rounded olivine-bearing pallasites by annealing of fragments in partially molten Fe-Ni-S. Moreover, early mixing in the planetesimal can form regions containing olivine grains with different grain sizes that could explain the formation of mixed-type pallasites.

Key words: Olivine; Pallasite; 2D numerical model; Grain growth; Partially molten Fe-Ni-S; Silicate-metal texture

1. Introduction

Pallasites are stony-iron meteorites with an average mineralogical composition consisting of 64.9 vol% olivine (minimum-maximum range 37-85 vol%), 31.0% Fe-Ni metal (14-56 vol%), 2.3% troilite (FeS) (0.1-7.3 vol%) (Buseck 1977). Extra phases, have been recognized to be < 1.5 vol%, except for recent findings of five pyroxene bearing pallasites (up to 40 vol% orthopyroxene - North West Africa NWA 1911 – Bunch et al. 2005) inclusive of a sample that contained plagioclase as well (NWA 10019 – Agee et al. 2015). Mineralogical composition coupled with oxygen isotopes and trace elemental composition of silicate and metal phases suggest several distinct source bodies for pallasites (Yang et al. 2010, Boesenberg et al. 2012; Ruzicka et al., 2017). Therefore, formation of pallasite-like material must have been relatively common on planetesimals in the early solar system. Furthermore, pallasites do not resemble crust, mantle or core material as sampled by other types of meteorites.

To add to the complexity of pallasite genesis is the bimodal texture of numerous samples, some are almost entirely composed of well-rounded and well-sorted olivine grains as opposed to specimens bearing exclusively fragments (Buseck 1977, Scott 1977). The nature of olivine fragments varies from broken polygonal crystals (e.g., Esquel), to submillimeter-sized often randomly oriented fragments (e.g., Huckitta). A microscopic analysis of fragments reveals that their edges are moderately to well-rounded suggesting some form of moderate reheating after brittle fracturing (Saiki et al. 2003). When pallasites present simultaneously rounded grains and fragments of olivine (mixed-type) those are not intermixed. Instead, rounded and fragmental olivine grains occupy discrete, confined portions.

Intuitively, the formation of rounded olivines is ascribed to prolonged annealing at significant depth inside a planetesimal, while highly fragmented ones would have formed by violent fracturing events. Two prevalent schools of thought (with some modifications) set the debate regarding the origin of pallasites, suggesting two formation mechanisms: (i) that pallasites represent the vicinity of the core-mantle boundary in planetesimals (CMB) in planetesimals (Ringwood 1961, Wasson and Choi 2003, Boesenberg et al. 2012, McKibbin et al. 2016) or (ii) a non-destructive impact introducing and mixing metal from the impactor with the target body olivine mantle (Mittlefehldt 1980, Scott and Taylor 1990, Tarduno et al., 2012, Solferino et al. 2015).

Some key arguments against formation in proximity of the CMB are: (i) The lack of iron meteorites representing the core of pallasite parent bodies (Eugster et al. 2006, Scott et al. 2009, Yang et al. 2010); (ii) REE concentration in phosphates, present as minor phases in numerous pallasites, indicating near surface crystallization (Hsu 2003); (iii) evidence that olivine and Fe-Ni are not from the same body (Hsu 2003); (iv) remanent magnetization recorded by Fe-Ni inclusions located inside olivine grains requiring trapping at relatively shallow depth to satisfy cooling-rate constraints (Tarduno et al. 2012, Bryson et al. 2015).

In the impact scenario the formation of rounded olivine pallasite-type material must occur via rounding of fragments followed by grain growth at a sufficient rate to yield large (i.e., up to 20 mm) rounded grains. This process takes place while olivine is surrounded by partially (or fully) molten Fe-Ni-S. This is confirmed by a number of textural evidences, inclusive of an increase in the surface area of olivine-olivine grain boundaries and formation of triple/quadruple junctions (Scott 1977, Saiki et al. 2003). The chief constraint is cooling of the interior of a pallasite source body that eventually will stop grain growth. Saiki et al. (2003) showed that rounding is a fast process at the scale of millimeter and submillimeter fragments. Solferino et al. (2015) performed annealing experiments on olivine plus Fe-S melt and demonstrated that coarsening of olivine fragments surrounded by Fe-S liquid is a viable mechanism to yield even the largest grains and to reproduce the characteristic textures observed in rounded olivine-bearing pallasites. However, it remained to be tested, whether the large fraction of S present in the experiments performed by Solferino et al. (2015), i.e., ~9.5 wt% of the total sample mass (i.e., olivine and Fe-S), and which falls outside of the range observed in natural samples (up to 6.4 wt% - Buseck 1977), enhanced the grain growth rate.

It must be noted that the silicate and metal proportions vary significantly among as well as within pallasite meteorites, often with very large metal crystals (tens of centimeters or more) separating one or more domains rich in olivine and/or troilite grains or fragments (e.g., Scott 1977). Nevertheless, those parts of samples where rounded olivine grains are prevalent, invariably show intermixing of olivine grains and metal crystals with minor amounts of troilite, and specific shapes of olivine-olivine grain boundaries. This type of texture can indeed be reproduced by experiments, if the starting material is carefully designed.

To add to the complexity of pallasite formation, the genesis of mixed-type pallasites remains hitherto unclear. Most studies on pallasites do not address this topic directly, though it was suggested that the genesis of mixed-type pallasites is related to dike-like intrusions of molten metal immediately following the impact (e.g., Yang et al. 2010) or later disturbance of the pallasites source region by multiple collisions (Fowler-Gerace et al. 2016).

This investigation aims to define experimentally the grain growth rate of olivine in a partially molten Fe-Ni-S matrix using a sulphur content compatible with observed pallasite values (Buseck 1977), the latter being the only and the most reasonable boundary condition for choosing a starting material. Therefore, the study will quantify the effect of sulphur content on olivine growth rate, by comparison with the results of Solferino et al. (2015), where an eutectic Fe-S composition (i.e., Fe = 70.5 wt% and S = 29.5 wt%) was used. The new composition of the Fe-Ni-S (with 10 wt% sulphur – Table 1) implies that during annealing solid Fe-Ni will be present in addition to olivine and Fe-S liquid, reproducing the three main phases observed in natural pallasites. Moreover, we discuss theoretical predictions for grain growth processes and supply a robust explanation of the mechanics of grain size increase in a three-phase system.

The newly computed olivine grain growth parameters will be employed in a 2D thermomechanical numerical model to find conditions satisfying all available constraints from natural samples. Additionally, these calculations will be used to test whether early mixing in the planetesimal mantle can explain the genesis of mixed-type pallasites.

2. Methods

2.1 Experimental setup

Two new series of experiments and one additional run to complete a previous sequence reported in Solferino et al. (2015) (third run series – see Table 1) were performed for this study. The starting materials were mixtures of natural olivine (San Carlos) and synthetic Fe-S powders (sulphur 99.999% purity in pieces of 1-2 mm size and iron 99.9% purity mesh 5 μm powder from CeramTM Inc.). The composition of the Fe-S employed in the first experimental series was 90 wt.% Fe and 10 wt.% S, while for the second set of experiments the Fe-S composition was varied from Fe:S = 70.5:29.5 wt.% (corresponding to the eutectic of the Fe-FeS binary system at 1 GPa – Brett and Bell 1969; Usselman 1975) to Fe:S = 95:5 wt.% (Table 1). One additional experiment used an olivine to Fe-S proportion of 68:32 vol.% with eutectic Fe-S composition. Fe-S powders and the final mixtures to be loaded in the sample charges were prepared following the procedure described in Solferino et al. (2015). The olivine powder was taken out of the same batch prepared for the experiments run by Solferino et al. (2015), with a mean grain size of 1.8(5) μm. Before use the olivine powder was kept for three days in a low vacuum furnace at 110 °C.

Graphite capsules (4.0 mm height x 2.0 mm diameter) were used to contain the olivine plus Fe-S powder. Before placing in the assembly the graphite capsules were fit into platinum capsules or wrapped into platinum foil. Annealing experiments (Table 1) were performed using a 14 mm bore hole end-loaded piston cylinder press with Talc-Pyrex sleeve and 36 mm long stepped graphite furnace. Crushable MgO cylinders were used to contain the capsule, while an Al_2O_3 cylinder contained the mullite thermocouple ceramic holder. The accuracy of B-type thermocouple (employed for all runs) was ±5 °C. A 0.6 mm thick dense alumina disk was placed between the thermocouple and the Pt-capsule. Pressure calibration was done using the fayalite + quartz = orthoferrosilite reaction (Bohlen et al. 1980) and the quartz-coesite polymorphic transition (Bose and Ganguly 1995). Shut down of the power resulted in quenching of the run charges at a rate of ~60 °C/s down to 200-250 °C. Run conditions, olivine and Fe-S volume proportion, and Fe-S composition for each run are reported in Table 1.

2.2 Compositional analysis

Run products were embedded in epoxy, polished, and coated with carbon for backscattered electron (BSE) imaging and wavelength dispersive spectrometry plus energy dispersive X-ray spectroscopy chemical characterization (WDS and EDS, respectively). Samples were characterized using three JEOL JXA-8200 electron microprobes at the laboratories of Universita' degli Studi di Milano (Italy), ETH Zurich (Switzerland), and Bayerisches Geoinstitut (Germany). Sample FS90S10-1 was analyzed by all three facilities for cross-reference (see Table 1).

Olivine analysis employed 15 kV, 20 nA and a focused beam with 1 μm width, whereas Fe-Ni and Fe-S were analyzed at 20 kV and 20 nA. To average the analyses of finely spaced Fe-S quench features (Fig. 1a) the beam was defocussed to 5-50 μm width. WDS standards employed for this study were forsterite, fayalite and liebenbergite (Ni_2SiO_4) for olivine analyses, and native iron, troilite (FeS), and bunsenite (NiO) were used for analysis of Fe-Ni and Fe-S. EDS spot analysis and X-ray elemental maps of whole capsules revealed that no intake of Fe (and/or Ni) into the Pt-foil occurred during experiments.

2.3 Digital Image Analysis

Two sets of measurements were operated after collection of BSE images utilizing the software ImageJ (public domain, Wayne Rasband, NIH, USA). The scope was to determine the fraction of sample surface area occupied by olivine (that we interpret as olivine volume fraction) and to quantify the size and shape of olivine grains. Prior to digital measurement BSE images were binarized and in the case of grain size and shape determination a further manual separation of olivine grains in direct contact was performed utilizing a photo editing program. Details on this procedure and achievement of statistical significance are described in Solferino et al. (2015). The largest uncertainty of olivine fraction determination is 4.8% (run OFS-15 - Table 1).

The circle equivalent diameter (CED) of the area of each object, $d_{2D} = 2(A/\pi)^{1/2}$ (where A is the area of the olivine grain) was taken as grain size, afterwards 2D apparent size was corrected for sectioning effect after Kong et al. (2005): $d_{3D} = d_{2D}/(\pi/4)$. The shape of each grain was quantified using the following parameters: Roundness $r = 4\pi/(MA)$ (where M is the semi-major axis of the object; Waddell 1932), and the ratio p of perimeter/CED.

3. Theory: Background on grain growth in multi-phase systems

Increase of the average grain size of a polycrystalline aggregate exposed to high temperature in presence of a secondary phase (often referred to as 'matrix') occurs by coarsening or normal grain growth (Hillert 1965) of grains in mutual contact (Ardell 1972). On the other hand, Ostwald ripening takes place for particles suspended in a fluid (Voorhees 1985). Coarsening in two-phase systems involves several cases, based on the following conditions: (i) whether the secondary phase is another solid rather than a liquid/fluid, (ii) whether the solubility of the main coarsening phase into the matrix is finite as opposed to the case where solubility is negligible (German 2010 and references therein). In general, solubility of the main phase into the surrounding matrix catalyzes coarsening. However, the amplitude of the dihedral angle at solid-solid-liquid interfaces (see Bulau et al. 1979 for a comprehensive discussion) and the fraction of secondary phase can counteract this grain growth rate enhancing effect (German 2010 and references therein).

The general form of the equation relating grain size increment with time is:

$$d^n = d_0^n + k_0 \exp[-E_a/(RT)]t \quad (1)$$

, where d is the grain size at time t, d_0 the starting grain size, n the growth exponent, k_0 the pre-exponential frequency factor for grain growth rate parameter (see German 2010 for further discussion of the physical meaning of k_0), E_a the activation energy of the process controlling coarsening, R the gas constant, and T the absolute temperature. The growth exponent n, generally assumes a value of 3 (Atkinson 1988). Yet, interaction with a competing coarsening secondary phase may reduce the grain growth rate, resulting in $n \geq 4$ (Guignard et al. 2012 and 2016), while grain growth is fastest when controlled by interfacial defect populations (n = 2 - German 2010). The latter case is appropriate for randomly oriented silicate minerals aggregates. Similarly, the presence of pores or a fluid/solid phase along the boundaries between grains of the major phase can effectively hinder its coarsening rate (Zener pinning – Smith 1948), resulting in $n > 3$. When two solid phases are present in the liquid, the interfacial energies of both solid phases with the liquid as well as surface energies have to be considered. The ratio of the area occupied by the liquid to the total grain boundary area must be evaluated for all interacting phases as well (Ahmed et al. 2013).

4. Results

4.1 Composition of main phases

Olivine composition is homogeneous in each experiment with no observable zonation at the individual grain scale. After annealing olivine becomes more Fe-rich with respect to the starting San Carlos olivine - except for run OFS-16, where fayalite content is the same as those of the starting material (within analytical error - Table 2). The NiO olivine content drops to zero (or nearly so, see run FS9010-3 - Table 2).

For run OFS-16 the eutectic composition of the Fe-FeS system at 1 GPa was used (Usselman 1975). Run temperature exceeded eutectic temperature, thus two phases were present: olivine and Fe-S melt. The resulting composition of olivine and quench product of Fe-S liquid is consistent with experiments by Solferino et al. (2015).

Experiments with Fe-S composition other than eutectic Fe-FeS yielded a third phase with composition of nearly pure iron and ~1 wt% Ni (indicated as solid Fe-Ni from now on - Table 2 and Fig. 1b,c,d). For these runs an analysis of the quench product of Fe-S melt was not possible due to the presence of quench immiscibility features that require collection of WDS analysis with a defocussed beam (>5 μm), while the largest Fe-S melt pools are a few micrometers across. In run FS9505 no obvious Fe-S melt pool was observed. Yet, X-ray elemental maps showed the presence of thin films and rounded features (< 2 μm across) enriched in sulphur and distributed across the sample (see Appendix A). A fourth phase with composition of Mg 26.66(33), Fe 43.34(40), and O 29.99(72) wt% was detected in the above-mentioned experiment.

4.2 Texture of run products

Neither the run time of the experiments nor the temperature affect the appearance of olivine grains in the experiments. Also the presence of solid Fe-Ni in various proportions does not seem to produce a noticeable difference compared to olivine plus Fe-S liquid runs (Fig. 1 and 2). Olivine grains triple junctions show 120° contact angle, while straight olivine-olivine grain boundaries are the prevalent textural feature.

Roundness of olivine grain is the same for all runs (within error), with an average value of ~0.7 (Table 3), corresponding to well rounded grains (Waddell 1932). The shape of olivine, expressed as the ratio of the grain perimeter to the CED, shows a marked sensitivity to the fraction of Fe-S liquid present during annealing (Fig. 2b). Specifically, when olivine and Fe-S liquid are the only two phases (Third run series – Table 1), grains become markedly more faceted (i.e., higher Perimeter/CED) when Fe-S < 15 vol%.

Fe-S liquid is located at triple junctions as well as larger pools between clusters of olivine grains in runs where no solid iron is present. The size of these pools is proportional to the olivine grain size. In OFS-16 (31.1 vol% FeS – Table 1) a few olivine grains appear to be completely surrounded by Fe-S. Whether the latter effect is due to 2D slicing, could not be ascertained. When solid iron-nickel is present in various proportions, Fe-S liquid occupies principally triple/quadruple olivine grains junctions, while most of the unconstrained intergranular gaps are filled by iron (Fig. 1c). A significant fraction of straight and mildly curved olivine boundaries seems to be in direct contact with iron, and occasionally pod-like-shaped iron occupies olivine triple junctions. Run FS8020, where the calculated Fe-S liquid/solid iron ratio is 14.4 (based on Usselman 1975), shows the presence of iron in the form of nuggets (high circularity features) or pod-like shapes (Fig. 1b), occasionally entirely surrounded by Fe-S liquid.

4.3 Olivine grain growth

Olivine grain growth rate was quantified for experiments with Fe/S = 90/10 and 20 vol% of Fe-S. Non-linear square fit of the 'first run series' data (Table 1) resulted in the following parameters: $n = 3.71(61)$, $k_0 = 3.23$ μm^{n}s^{-1}, $E_a = 101(42)$ kJ mol^{-1}. All three parameters were fit simultaneously to the non-linear square calculation and errors on temperature, initial grain size and average grain size measurements were considered. The maximum and minimum values of k_0 are 61.04 μm$^{4.32}$s^{-1} and 0.23 μm$^{3.10}$s^{-1}.

A comparison of the average grain size for run OFS-16 (Table 1) with experiments from Solferino et al. (2015) performed at the same temperature and run duration shows that there is an inverse correlation between grain growth rate and volume of Fe-S liquid. This justifies the choice of utilizing same olivine/Fe-S volume proportion runs only for quantification of grain growth parameters. Specifically, data were fit to the following logarithmic trend to an R^2 of 0.992: average olivine size = -8.359 ln(Fe-S vol%) + 40.708.

Finally, annealing of olivine and Fe-S mixtures in volume proportion of 80:20 with varying Fe/S ('second run series' - Table 1), yielded identical grain size for all Fe-S compositions except Fe/S=70.5/29.5 (OFS-9 – Table 3), where a significantly larger average olivine size was measured.

4.4 Normalized Grain Size Distributions (NGSD)

The frequency distribution for olivine grain size was normalized by dividing the frequency of each bin by the maximum frequency and dividing grain size of each bin by the mean grain size d_m. The procedure to define the width of bins (0.10 for this study) used to classify grain dimension followed Faul and Scott (2006) and is explained in detail in Solferino et al. (2015). Following this approach, it is possible to compare GSD of experiments with different average grain sizes.

NGSDs for the first run series are self-similar with more than 95% of grains $< 2.00\ d_m$, the peak of the distribution at 0.60 d_m (within 1σ), and maximum grain size $d_{max} < 2.50\ d_m$ (Fig. 3a; Table 3). This indicates that all annealing runs achieved steady-state grain growth, including those with the shortest duration of 40 hours.

For the second run series experiments the considerably larger average grain size observed for Fe/S=70.5/29.5 (see section 4.3 and Table 3) is accompanied by significant differences of GSD characteristics (Fig. 3c). The peak of OFS-9 distribution is located at 0.85 d_m, while for all other experiments it is 0.55 d_m. A sharp cut-off of the minimum grain size (d_{min}) is evident for all experiment, while d_{min} is ~0.2 d_m for a Fe/S ratio from 80/20 to 95/5, d_{min} is located at ~ 0.6 d_m for eutectic Fe-FeS composition. For the third run series the portion of NGSDs at $d > d_m$ is similar for all runs, with $d_{max} < 2.2\ d_m$. The position of the peak of the distribution shifts from ~0.6 d_m for Fe-S > 20 vol% to ~1.0 d_m for all other runs. The cut-off at small grain sizes is consistently at around 0.4 d_m for Fe-S ≤ 20 vol%, while it is at 0.3 and 0.2 d_m for Fe-S ranging from 32 to 40 vol%, respectively (Fig. 3b).

5. Discussion:

5.1 Phase and chemical equilibration

Attainment of chemical equilibration in the olivine + Fe-S system could be achieved by means of two reactions: $Mg_2SiO_4 + 6FeS + 4O_2 = Fe_2SiO_4 + 2MgFe_2O_4 + 3S_2$ leading to a fayalite content increase in olivine, and $Fe_2SiO_4 + 0.5S_2 = FeS + FeSiO_3 + 0.5O_2$ yielding more forsteritic olivine (Gaetani and Grove 1999). All but one experiment of this study produced a fayalite increment in the olivine composition. In run FS9505 an additional phase to olivine, solid iron, and Fe-S was observed. This phase has a composition similar to Mg-ferrite (see section 4.1), with the closest calculated formula (on the basis of mass fraction of Mg, Fe and O) being $Mg_2(Fe^{2+})_{0.5}Fe^{3+}O_4$. We hypothesize that the total Fe content of sample FS9505, larger than for all other runs, might have fostered growth of Mg-ferrite phase into larger, visible grains. Conversely, such a phase might not be visible in other experiments due to sub-micrometer size.

The most likely driver of chemical re-equilibration of olivine is oxygen fugacity. The graphite capsule only provides the highest fO_2 buffer (graphite-CO-CO_2 or CCO buffer - Holloway et al. 1992), while the actual redox conditions are determined by the sample material itself. Righter et al. (1990) defined an equation to relate fO_2 and forsterite content of olivine applicable to pallasite-like material, the latter being identical to the experimental charges of this study consisting of olivine with solid Fe-Ni and Fe-S. Using the aforementioned equation, experimental fO_2 was calculated as IW-0.69 to IW-0.53 (where IW is the Iron-Wüstite oxygen buffer – Table 2). These values are identical within 1σ, and they are 4 to 5 log units below CCO (depending on run temperature). This indicates that the presence of reduced Fe-S substantially decreases oxygen fugacity. A comparison with results from Solferino et al. (2015) shows that the mass fraction of Fe-S determines how reduced the sample charge is (cfr. OFS-4, -15, and -16 – Table 1 and 2), while the Fe/S ratio does not seem to have any relevant effect (cfr. OFS-9 with FS9505 – Table 1 and 2).

The disappearance of Ni from olivine and its presence in the solid iron phase indicates that nickel partitioned almost completely into Fe-S. If nickel partitioned solely into the solid iron phase, first order calculations indicate that the latter phase Ni content should be 0.8 to 1.4 wt%, depending on run temperature (i.e., solid Fe/ liquid Fe-S ratio) and Fe/S ratio of starting materials (based on Usselman 1975). Yet, a precise mass balance calculation cannot be performed, since it has been shown that at the experimental conditions nickel partitions into liquid Fe-S as well (Solferino et al. 2015). Nickel content can be measured in Fe-rich immiscibility features generated during quench of Fe-S liquid, provided that they are larger than 5-10 μm. This is not the case for this study, with features being sub-micrometric.

5.2 Grain growth of olivine in Fe-S liquid

A thorough discussion of the leading grain growth mechanism in olivine plus Fe-S liquid system was presented in Solferino et al. (2015). The additional experiment (OFS-16) performed for this study, confirmed the smooth trend of faster grain growth for smaller Fe-S liquid fraction (see Fig. 2a). Analysis of NGSDs plots (Fig. 3b) shows that for Fe-S ≥ 32 vol% the number of relatively small grains (i.e., $d < 0.5\ d_m$ – see section 4.4) preserved during annealing is significant. This is likely due to the presence of olivine grains almost completely surrounded by Fe-S liquid. For those grains coalescence is strongly inhibited due to reduced olivine-olivine grain boundary fractional area (German 2010). At the other end of the scale (10 vol% Fe-S) grain growth is fastest (Table 3). Observation of BSE images of experiment OFS-4 (see figure 1 in Solferino et al. 2015) shows that Fe-S liquid is located at triple/quadruple junctions only. In such a case, the olivine-olivine grain boundary area is maximized. The matrix phase represents no obstacle to grain boundary movement-controlled grain growth as it is not scattered along those grain boundaries. This is in accordance with the minimization of the system surface energy (German 2010, Ahmed et al. 2013) and is a consequence of the high dihedral angle displayed by Fe-S liquid in contact with olivine (e.g., Rose and Brenan 2001, Terasaki et al. 2005). An effect of grain-boundary movement-controlled grain growth is the development of more polygonal grains, i.e., more faceted grains, confirmed by the markedly higher value of Perimeter/CED of run OFS-4 (Table 3). Furthermore, the hypothesized threshold value for absence of Fe-S liquid along olivine-olivine grain boundaries, i.e., $f < 0.15$ (where f is non-dimensional and $0 \leq f \leq 1$), is in good agreement with Fe-S liquid interconnectivity determined via in-situ electrical impedance measurements (Bagdassarov et al. 2009).

The small grain growth exponent, n = 2.42(46), calculated for olivine:$Fe_{70.5}S_{29.5}$ = 80:20 vol% by Solferino et al. (2015) indicates that even for $f > 0.1$ Fe-S liquid is easily removed from olivine-olivine grain boundaries. This agrees with studies on textural equilibration of crystalline plus high dihedral angle liquid (Walte et al. 2007). The presence of Fe-S liquid decreases the growth rate with respect to pure phase olivine NGG (n = 2 Karato 1989), however it is not as effective as, for example, basalt melt (cfr.

Faul and Scott 2006). The latter conclusion is based on theoretical studies predicting that non-wetting liquids (Bulau et al 1979, von Bargen and Waff 1986) do not hinder the grain growth rate of the solid phase significantly, while wetting liquids do (German 2010 and references therein).

5.3 Grain growth of olivine in partially molten Fe-Ni-S

Analysis and interpretation of coarsening of olivine grains in the presence of solid Fe-Ni as well as Fe-S liquid is far more complex. A thorough discussion on the significance of the experimental data and scientific approach selected for the computation of olivine growth rate in partially molten Fe-Ni-S is presented in Appendix D, while here we focus on understanding the key process that governs coarsening. All experiments with partially molten Fe-Ni-S contained 20 vol% of Fe-Ni-S ($f = 0.2$). Since chemical equilibration of olivine and Fe-Ni-S takes place in the very first stages of annealing, together with rounding of olivine fragments (see section 5.1 and discussion in Solferino et al. 2015), no further chemical interaction between phases can occur. Therefore, the case-scenario with coarsening of a major phase, which is non-soluble in the matrix, is the best proxy for our experiments. Liquid Fe-S pools and olivine grains grow simultaneously in size. This is related to an increase of spacing between the growing grains (Yoshino et al. 2005, Solferino et al. 2015) and is no obstacle to olivine grain boundary movement. Above a certain fraction of solid Fe-Ni (i.e., $f \geq 0.1$ – Ahmed et al. 2013), its coarsening is the process limiting olivine growth. Under these conditions, the olivine growth exponent n has the same value of that of Fe-Ni (coupled grain growth - Guignard et al. 2016 and references therein). The scenario foresees the presence of pod-like-shaped solid Fe-Ni between olivine-olivine grain boundaries, where Zener pinning becomes effective. Normal grain growth of olivine can only proceed unhindered ($n = 3$ - German 2010), when solid Fe-Ni pods coalesce into larger ones that occupy exclusively triple/quadruple grain junctions, thus leaving the olivine-olivine boundaries (figure E1a, b). Although we did not quantify the coarsening of solid Fe-Ni in our experiments, observations by Saiki et al. (2003) and Guignard et al. (2012, 2016) for olivine plus Fe-Ni and forsterite plus nickel, respectively, can be used to infer the energetics of this process. Specifically, Guignard et al. (2016) calculated the activation energy for coarsening of solid nickel in forsterite:nickel = 80:20 volume proportion material annealed at 1340 °C to be 235 kJ/mol. E_a of forsterite grain growth was 400 kJ/mol, far in excess of 101(42) kJ/mol computed in this study. The latter value does not match any previously reported activation energy for olivine coarsening (for a tentative interpretation see Appendix E).

The forsterite (and nickel) grain growth rate in Guignard et al. (2016) was reduced as much as to yield a growth exponent n = 5. In comparison our calculated value n = 3.70(61) defines a faster coarsening, though comparable to the lower value indicated by Guignard et al. (2012). To our understanding the key to explain the inferred higher growth rate of Fe-Ni in our experiments is related to the presence of a third phase, here liquid Fe-S. This is a sensible hypothesis, considering that theory predicts that a liquid matrix phase (here Fe-S) with finite solubility in the solid phase (i.e., Fe-Ni) enhances the grain growth rate of the latter (German 2010 and references therein). In the 'olivine + Fe-Ni solid + Fe-S liquid' system, Fe-Ni growth is governed by diffusion along olivine-olivine grain boundaries and olivine triple junctions. This process is catalyzed with respect to the Fe-S liquid-absent system (e.g., Guignard et al. 2012, 2016) by thin films of Fe-S, which appear to be located between Fe-Ni and olivine (see Appendix A, figure A2b). Liquid Fe-S could be spread all along olivine-olivine grain boundaries (Gaetani and Grove, 1999) and it would act as a preferential medium for Fe and Ni atoms movement from Fe-Ni pod to pod enhancing the diffusion rate (further details in Appendix E and figure E1a,b).

5.4 Application to olivine grain growth in pallasites

A thorough comparison of experimental conditions with those in the pallasite formation environment was presented in Solferino et al. (2015). There it was shown that the application of experimental results from olivine plus Fe-S annealing to grain growth in rounded olivine-bearing pallasites is valid. Two questions remained open: Quantification of (i) the effect of solid Fe-Ni presence and (ii) a significantly smaller total sulphur content, 3.6 wt%, as opposed to 9.5 wt% of Solferino et al. (2015), on the olivine growth rate.

This is important in the light of effects of sulphur content (and by implication of Fe-S liquid fraction) on the kinetics of olivine coarsening (Ohtani 1983, Saiki et al. 2003).

The first run series experiments contained 3.6 wt% sulphur, within the range observed in pallasites, and 8.2 to 11.1 vol% solid Fe-Ni (calculated after Usselman (1975) for T = 1300 and 1100 °C, respectively – see supplementary material 'Table 1 Extended' for reference). The experimental solid iron-nickel fractions are smaller than those in natural pallasites, nevertheless these values are proximal or in excess of the fraction at which the matrix phase controls coarsening rate of the major phase (see section 3). Therefore, the experiments provide a valid analogue proxy for answering the two aforementioned questions.

A further indication that the new experiments are a better pallasite analogue with respect to those in Solferino et al. (2015) arises from the calculation of L_2 norms of NGSDs. NGSDs of the two most representative samples of rounded-olivine bearing pallasites (Brenham and Springwater), were compared to the NSGDs of the experiments with highest temperatures and longest run times (i.e., the most texturally matured) with (i) eutectic Fe-FeS (run OFS-10 of Solferino et al. 2015) and (ii) $Fe_{90}S_{10}$ (run FS9010-5 - Table 1). The resulting L_2 norm for the comparison between FS9010-5 and the two pallasites is 46.8% smaller than those for OFS-10 (see Appendix B for further details). Additionally, the L_2 norm of FS9010-5 is 34 to 52% smaller compared to those obtained for first order and second order surface diffusion controlled growth and Rayleigh NGSDs (see Faul and Scott (2006) and Solferino et al. (2015) for further details on theoretical grain size growth models and the definition of NGDS curves for theoretical grain growth mechanisms). This improved fit to pallasite data indicates that these might have originally formed with small amounts of sulphur present, in agreement with recent work suggesting efficient vapor loss from planetesimals (Hin et al. 2017).

6. Modelling

6.1. Numerical approach

A number of constraints must be considered for rounded olivine-bearing pallasites: (i) The depth of the intrusion of Fe-S into the target body mantle must allow cooling at a rate appropriate to form kamacite/taenite exsolution lamellae (Yang et al. 2010), (ii) temperatures must be beneath the Curie temperature while the core dynamo is active to allow for the recording of remanent magnetization (Tarduno et al. 2012, Bryson et al. 2015) and (iii) the olivine grains must be able to grow to the range observed (2-10 mm) in rounded olivine-bearing pallasites (Buseck 1977).

The thermal evolution of the planetesimal interior was studied using the 2D thermochemical code I2ELVIS (Gerya and Yuen 2003, 2007). The code uses a finite-difference fully-staggered grid and solves the equations for mass, momentum and energy conservation and includes depth-dependent gravity acceleration. The numerical model also accounts for radiogenic, shear and latent heat. The grid employed has 501 x 501 grid points, corresponding to a grid resolution of 1.5 km. The model setup is based on the 1D model of Tarduno et al. (2012) assuming a differentiated structure with an iron core and an olivine mantle. The starting temperature of the core and mantle is 1,327 °C and the model start time is 5 Myr after CAI formation. Thus, the starting condition assumes that the magma ocean stage ended while partial silicate melt is still present. The grain growth in the mantle is tracked on the Lagrangian markers and we assume an initial olivine grain size of 100 µm. For the olivine grains in the undisturbed mantle after and everywhere prior to the impact we use the grain growth rate for pure olivine (Karato 1989). This choice is justified by the lack of plagioclase and pyroxene in most pallasite samples and by the absence of any evidence of silicate melt presence during their formation. Once the pallasite-forming impact occurs the grain size in the impact region is reset to the starting value and the grain growth proceeds at the rate calculated in this study for olivine surrounded by partially molten Fe-S (see section 4.3). Since all grain growth parameters have error bars, we perform additional calculations using the combinations allowing for the fastest and slowest possible grain growth. For simplicity we consider for the impact zone a circular sector with 20° central angle extending across the depth of the entire mantle (Fig. 4a) and assume that the rheology is controlled by olivine everywhere in the mantle. For the density of the impact material we assume a mixture of 95% olivine and 5% Fe-S. Additionally, we consider that olivine coarsening becomes negligible once the temperature drops below the cotectic point for the sulphur-poor portion of the Fe-Ni-S system at ~950 °C (Kullerud 1963). For further details on the simulation method see Gerya and Yuen (2003, 2007) and Golabek et al. (2014). Applied physical parameters are those given in Golabek et al. (2014).

6.2 Numerical Model Results

We study multiple scenarios, considering planetesimal radii ranging from 100 to 300 km and pallasite-forming impacts occurring between 15 and 60 Myr after CAIs. Generally, deeply emplaced olivine + Fe-Ni-S evolves into rounded, texturally equilibrated olivine grains intermixed with Fe-Ni and Fe-S aggregates, while material close to the surface experiences very limited olivine grain growth and no textural maturation. Using a 200 km radius body as reference model the numerical results show that in case the pallasite-forming impact happens <50 Myr after CAIs convection in the partially molten mantle is still possible. This is a consequence of the low mantle viscosity (~10^{17} Pa s) caused by the presence of silicate melt. In this scenario olivine + Fe-Ni-S material originally emplaced at great depth can be mixed with undisturbed mantle olivine and with olivine-metal mixtures situated closer to the planetesimal surface (see Fig. 4a and supplementary movie). Further cooling of the pallasite source body eventually stops convection. This process might yield the formation of regions containing large, rounded olivine grains juxtaposed to smaller, fragmental olivines, i.e., mixed-type pallasites-like material. Finally, olivine-metal-sulphide mixtures originally located very close to the surface experience very limited olivine grain growth and no remixing into the mantle (see Fig. 4a).

At depths satisfying the natural pallasite cooling rate constraints (Yang et al. 2010), progressive cooling of the mantle, followed by crystallization of the Fe-Ni-S, stops olivine grain growth at around 50 Myr after CAI formation (Fig. 4b). Meanwhile the core remains molten for an additional >120 Myr. Based on the core-mantle boundary heat flux we can estimate the magnetic Reynolds number (see also Appendix C) to be ~2-3. Theoretically this value should be sufficient to drive a thermal dynamo on planetesimals

(Weiss et al. 2008), so it is viable that Fe-Ni inclusions in pallasites silicates could be remanently magnetized during this period of time (e.g., Tarduno et al. 2012). What is more, for early pallasite-forming collisions both the minimum and the maximum olivine grain size (see Fig. 4c) in the region satisfying constraints (i) and (ii) from above, is in agreement with observations from natural rounded olivine pallasites (Buseck 1977).

In smaller planetesimals with 100 km radius, mantle cooling is faster. Thus, both convection and olivine grain growth are limited. Also thermal dynamo activity ceases before the depth range satisfying the natural pallasite cooling rate constraints passes the Curie temperature. On the other hand, larger objects with 300 km radius experience slower cooling. This allows for extended grain growth and a long-lasting thermal dynamo, with magnetic Reynolds numbers around 7-10 while material at depths satisfying the natural pallasite cooling rate constraints passes the Curie temperature. Thus objects with radii ≥200 km seem to be viable pallasite parent bodies.

7. Final remarks and future directions

With this study employing both experimental work and numerical modelling it was possible to establish that:

- For $f > 0.1$ of non-silicate matrix phase containing a significant fraction of solid Fe-Ni, the presence of sulphur in the system accelerates olivine grain growth by means of catalyzing coarsening of the secondary phase, i.e., solid Fe-Ni.
- Coarsening of olivine fragments in partially molten Fe-Ni-S after a non-destructive impact on a planetesimal is a viable mechanism to form rounded olivine-bearing pallasites.
- Convection in the partially molten mantle of pallasite source bodies can yield mixed-type pallasite-like materials.

7.1 Model limitations and future directions:

It should be noted that our global models do not consider that a part of the Fe-Ni-S material could percolate from the pallasite-forming region to the core of the planetesimal (e.g., Bagdassarov et al. 2009b). Also we assume that the rheology of the pallasite forming region is dominated by olivine. To our knowledge the rheology of such material has never been studied and future laboratory measurements will be necessary to improve this aspect of the numerical models. Additionally, we assume a simple 2D circular sector geometry for the pallasite-forming impact zone. This can be improved in the future by using the output of advanced 3D impact models to obtain a more realistic starting condition.

With our models we were able to show that olivine plus Fe-Ni-S materials with very diverse olivine grain sizes (see Fig 4c) can be mixed before convection ceases. Based on currently available experimental data it is not possible to determine whether olivine fragments experiencing a limited grain growth from 100 to 110-150 μm (blue lines in Fig. 4c) become fragments with rounded edges or if they achieve a rounded grain shape. Thus, further experimentation on very short (~2-10 hours) annealing is necessary to verify this hypothesis.

Due to viscosity cut-offs in the numerical model the initial temperature is assumed to be below the liquidus of Fe-Ni-S, whereas trace elemental composition of the metal present in pallasites (e.g., Ir content of some Main Group pallasites – Yang et al. 2010 and references therein), indicates that metal phases crystallized starting from a fully molten status. Nevertheless, due to more efficient heat loss, we expect that temperatures in excess of Fe-Ni-S liquidus should not last for a very long time. Thus, it is likely that annealing of olivine in fully molten metal had only a limited effect on olivine grain growth.

Finally, the grid resolution of our global model does not allow us to study the microstructural evolution of pallasites. For this purpose, future small-scale models are necessary that employ as input high-resolution natural pallasite imagery and three-dimensional microstructural data produced from 3D micro-tomographies of experimental samples.

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Figure captions:

Figure 1: BSE images of run products (see table 1 for experimental conditions). (a) OFS-16: Quench immiscibility features (white) in Fe-S pool (light grey) and olivine (dark grey). (b) FS8020: Fe-Ni nuggets and pods (light grey) in contact with olivine grains (black) or surrounded by Fe-S (dark grey). (c) FS9010-1: Fe-Ni (light grey) occupying triple/quadruple junctions between olivine grains (black). (d) FS9505: Medium gray phase (white square) is pseudo-Mg-ferrite (see sections 4.1 and 5.1), Fe-Ni (light grey) and olivine (dark grey).

Figure 2: Grain size and shape plots. (a) Effect of Fe-S liquid volume fraction on average grain size for same temperature and duration of annealing (error bars 2σ). (b) Effect of Fe-S liquid volume fraction on grain shape for same temperature and duration of annealing (error bar 1σ). (c) Effect of varied Fe/S composition for a fixed 20 vol% Fe-S fraction on grain size (all runs same temperature and duration –

error bars 2σ). (d) Effect of varied Fe/S ratio for a fixed 20 vol% Fe-S fraction on grain shape (all runs same temperature and duration – error bars 1σ).

Figure 3: Normalized grain size distribution (NGSD) plots. (a) NGSDs for Fe/S = 90/10 and varied temperature and annealing time. (b) NGSDs for Fe-S volume fraction of 10 to 40 vol% (all runs same temperature and duration). (c) NGSDs for Fe/S ratio of 70/30 to 95/5 and a fixed Fe-S volume fraction of 20 vol% (all runs same temperature and duration).

Figure 4: (a) Magnification of global numerical model depicting the temporal evolution of grain size in the pallasite forming region for a body with r = 200 km. The pallasite forming impact occurs at t_{imp} = 15 Myr after CAIs and the fastest possible olivine grain growth in agreement with experimental data is used. The solid white line shows the surface of the body, while the white dashed lines depict the region, where cooling rates are in agreement with pallasite constraints. (b) Time vs. maximum olivine grain size at depth range in agreement with cooling rate constraints for models with r = 200 km assuming different times for the pallasite forming impact (solid lines), slowest (dotted lines) and fastest (dashed lines) grain growth in agreement with experimental data. An additional model with r = 300 km is shown in brown color. Black diamonds mark the time corresponding to the two snapshots displayed in (a), while the grey region marks typical olivine grain sizes found in pallasites (Buseck 1977). (c) Time vs. minimum (blue lines) and maximum (orange lines) olivine grain size at depth range in agreement with cooling rate constraints for models with r = 200 km and t_{imp} = 15 Myr after CAIs using line styles as in Fig. 4b.

Appendices A, B, C, D, E

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Appendixes:

Appendix A – Identification of Mg-Ferrite

In experiment FS9505 no recognizable Fe-S pool was detected. Moreover, the presence of a medium-grey phase, Mg-ferrite (Fig. 1d and section 5.1), occasionally located between olivine grain boundaries (Fig. A1), rendered identification of Fe-S via grey-scale thresholding with digital analysis of BSE images non-viable.

Calculations based on Usselman (1975) indicate that 6.5 vol% Fe-S should be present at experimental conditions. In order to ascertain the presence of Fe-S in the sample, we collected X-ray elemental maps with a JEOL-8200 electron microprobe at Dipartimento di Scienze della Terra Ardito Desio of University of Milan (Italy). Figure A2 illustrates that a phase was recognized containing Fe and S, but negligible Mg, Si. Thus, this phase is likely to be Fe-S.

Appendix B – Quantitative comparison between natural pallasites and experiments

Multiple slabs of Brenham and Springwater pallasites were photographed at National History Museum of London. Digital image analysis of those slabs was used to determine NGSDs. More than 1,200 olivine grain were analysed for each pallasite, far exceeding the few hundreds used to produce NGSDs in Solferino et al. (2015).

Brenham and Springwater NGSDs were compared graphically (Fig. B1b) and mathematically against grain size distributions for the longest and highest temperature experiments with $Fe_{70.5}S_{29.5}$ and $Fe_{90}S_{10}$, respectively.

The L_2 norm of experimental and theoretical distributions against each sample were calculated and the results are reported in table B1. The L_2 values for FS9010-5 is equal to 53.2% of OFS-10, 48.3, 65.6, 48.9, and 18.3% of first order surface reaction controlled, second order surface reaction controlled, Rayleigh, and Log-norm, respectively.

Table B1: Results L_2 of norm calculation.

NGSD	Brenham	Springwater	Sum
OFS-10	1.138	1.107	2.244
FS9010-5	0.691	0.504	1.195
2nd order	1.097	0.714	1.811
1st order	1.242	1.231	2.473
Rayleigh	1.235	1.209	2.444

Log-norm	2.978	3.551	6.529

Appendix C – Estimating potential thermal dynamo activity inside the pallasite parent body:

To determine the heat flux across the core-mantle boundary we define circular sectors each 1° wide. In each of these circular sectors we find the closest pair of silicate markers just above the core-mantle boundary. Using the thermal conductivity and the temperatures of both markers we compute the radial component of the conductive heat flux q'_{cond} in the lowermost mantle for each circular sector:

$$q'_{cond} = k_{mantle} \, (T'_{CMB} - T'_{mantle})/(D \cos \beta) \quad (1)$$

where T'_{CMB} and T'_{mantle} are the temperatures of the two markers (the first being located at the core-mantle boundary and the second inside the specific circular sector at shortest distance from the first marker), D is the distance between markers and β is the angle between both silicate markers as seen from the center of the planetesimal. Using the values obtained for all circular sectors, we determine the average conductive heat flux across the lowermost mantle q_{cond} and the average silicate temperature on the mantle side of the core-mantle boundary T_{CMB_mean}.

Using the radial velocity v_{rad} of the silicate marker closest to the core-mantle boundary, we can compute the advective heat flux in the lowermost mantle for each circular sector:

$$q'_{adv} = \rho_{mantle} \, c_{P\_mantle} \, v_{rad} (T'_{CMB} - T_{CMB\_mean}) \quad (2)$$

where ρ_{mantle} is the density of the mantle material at temperature T_{CMB_mean} (for density computation of solid and molten mantle material see Golabek et al. 2009, 2011) and c_{P_mantle} is the heat capacity of the mantle material. Using the information from all circular sectors, we can compute the average advective heat flux in the lowermost mantle q_{adv}.

The addition of conductive and advective heat flux returns the average combined mean heat flux through the lowermost silicate mantle q_{mantle}.

In the next step we compute the minimum conductive heat flux across the CMB required to drive thermal convection in the core (e.g. Stevenson, 2003):

$$q_{core} = k_{core} \, \alpha_{core} \, g_{core} \, T_{core\_mean}/c_{P\_core} \quad (3)$$

where k_{core} is the thermal conductivity, α_{core} is the thermal expansivity, T_{core_mean} is the average temperature and c_{P_core} is the heat capacity of the core material.

The gravity acceleration at the core-mantle boundary is computed as

$$g_{core} = 4/3\pi G\rho_{core}R_{core} \quad (4)$$

where G is the gravitational constant.

In case the core is still molten and $q_{mantle} > q_{core}$, we follow the method described in Aubert et al. (2009) to determine whether the thermal dynamo operates.

First we compute the magnetic Rayleigh number Ra_q:

$$Ra_q = g_{core}(q_{mantle}-q_{core})4\pi R_{core}^2(\alpha_{core}/c_{P\_core})/(4\pi\rho_{core}\Omega^3 R_{core}^4) \quad (5)$$

, where ρ_{core} is the density of the core, R_{core} is the radius of the core, $\Omega = 2\pi/\tau$ is the angular velocity and τ is the rotation period.

Now we compute the power per unit volume P making the assumption that no solid inner core exists:

$$P = 0.6Ra_q \quad (6)$$

Next we compute the Ekman number

$$Ek = \eta_{core}/(\rho_{core}\,\Omega R_{core}^2) \quad (7)$$

, where η_{core} is the viscosity of the core.

In the next step we determine the magnetic Prandtl number

$$Pr_{mag} = \eta_{core}/(\rho_{core}\,\mu_{diff}) \quad (8)$$

, where μ_{diff} is the magnetic diffusivity of molten iron.

Finally, we can compute the magnetic Reynolds number

$$Re_{mag} = 1.310P^{0.42}\,Pr_{mag}/Ek \quad (9)$$

For parameters used in the dynamo calculations see table below.

Parameter	***Symbol***	***Value***	***Units***
Parent body radius	R_P	100-300	km

Parent body core radius	R_{core}	50-150	km
Viscosity of liquid core	η_{core}	10^{-2}	Pa s
Heat capacity of silicate mantle	c_{P_mantle}	1000	J/(kg K)
Heat capacity of the core	c_{P_core}	1000	J/(kg K)
Thermal expansivity of the core	α_{core}	10^{-5}	1/K
Thermal conductivity of solid silicate mantle	k_{mantle}	3	W/(m K)
Thermal conductivity of the core	k_{core}	46	W/(m K)
Magnetic diffusivity of molten core	μ_{diff}	2	m^2/s
Assumed rotation period	τ	9.8	h

The following table provides additional information on the outcome of our numerical simulations. Indicated are (i) the depth range, where the cooling constraints (dT/dt = 2.5-9.0 K/Myr while T = 775-975 K) suggested by Yang et al. (2010) and Tarduno et al. (2012) is satisfied (inside the mantle of the target body) and (ii) the time window during which the aforementioned depth range passes the Curie temperature (~633 K) as given by Tarduno et al. (2012). This can be compared to the timing of core crystallization (assuming an eutectic Fe-FeS melting temperature taken from Chudinkovich and Boehler, 2007) and the magnetic Reynolds number Re_{mag} while the top of the region of interest passes the Curie temperature.

It has been suggested that Re_{mag} > 1-10 is necessary for a self-sufficient thermal dynamo in planetesimals (Weiss et al., 2008; Roberts, 2007). On this basis, only objects with radii of ≥200 km are reasonable pallasite parent bodies. However, it should be kept in mind that we assume here a purely thermal dynamo, while core crystallization might have extended the lifetime of the dynamo (Bryson et al., 2015).

Radius [km]	***Core radius [km]***	***Depth [km]***[a]	***Time [Myr]***[b]	***Core crystallization [Myr]***[c]	$\boldsymbol{Re_{mag}}$[d]
100	50	31.0 – 49.0	108 – 149	50	0
150	75	34.5 – 39.0	159 – 190	102	0
200	100	30.5 – 42.5	131 – 238	174	2 – 3
300	150	24.0 – 37.5	118 – 219	369	7 – 10

[a] Depth range inside the planetesimal, where the cooling constraints [dT/dt = 2.5-9.0 K/Myr while T = 775 – 975 K – Yang et al. (2010), Tarduno et al. (2012)] are satisfied.
[b] Time window mantle material at depth range of interest passes the Curie temperature (T ~ 633 K) – Tarduno et al. (2012)
[c] Time of core crystallization
[d] Magnetic Reynolds number during 2 Myr time window after mantle material at top of depth range of interest passed the Curie temperature (T ~ 633 K) – Tarduno et al. (2012)

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Appendix D: Grain growth of olivine in partially molten Fe-Ni-S matrix

The calculation of grain growth parameters for olivine in partially molten Fe-Ni-S is based on five new experiments. The fit to an exponential growth law in the form of eq. 1 (section 3) was performed by non-linear square minimization utilizing the add-on 'Solver' of the software MSExcel (supplementary material 2), simultaneously computing all three variables, n, E_a, and k_0 (the quality of the fit is within 5% of the data – Fig. D1). In order to produce meaningful growth parameters, it is necessary to consider experimental results obtained from short and very long experiments at various temperatures. This way it is possible to verify whether normal grain growth (NGG) is achieved in all experiments used for the calculations. To confirm textural equilibration and NGG after short annealing at low temperature, a sixth experiment (FS9010-6) was performed (Table 1). While for the five selected experiments the key characteristics indicating normal grain growth and steady-state coarsening are satisfied by the invariable position of the peak of the distribution (0.60 d_m within error) and absence of anomalously large grains (*i.e.*, no grains with $d > 2.50$ d_m and smoothly tapering tail for $d > 1.5$ d_m), run FS9010-6 NGSD shows a peak at 1.0 d_m, a secondary peak at 0.65 d_m and a few very large grains with $3.25 < d < 3.30$ (Fig. D2). What is more, while for the five selected runs more than 2/3 of grains have d < 1.00 d_m, only 50% of the olivine grains in experiment FS9010-6 have a diameter smaller than the average grain size, indicating either a non fully accomplished textural equilibration or a different dominant grain growth mechanism for that temperature and annealing of 50 hours. We infer that the secondary peak of FS9010-6 NGSD at 0.65 d_m would turn into the sole peak found for annealing > 90 h as seen in run FS9010-4 (Table 1). We speculate that annealing duration < 48 h would have a similar effect for T = 1300 °C. Therefore, we believe to have performed short annealing runs for the shortest feasible durations. The longest annealing was performed for 360 hours only being limited by the technical aspect of reliable temperature reading and survival of the furnace. While this might not be fully satisfactory, and those limitations could be overcome in other facilities, the quality of the fit for both temperatures suggests that departures from the calculated trend of grain growth for even longer annealing times is unlikely.

A review of the exiting database of experiments on olivine grain growth highlights that longest runs span up to 700 hours (Faul and Scott 2006), but > 95% of data is taken from runs with $t < 300$ hours. While the number of experiments used to calculate grain growth parameters might be smaller than for other studies (typically 6-12 runs), we conducted a statistical investigation to prove the suitability of the employed dataset, *i.e.*, provide a bracket of fastest and slowest olivine grain growth inside a potential pallasite parent body, where Fe-Ni-S alloy could not have existed in a fully molten state either at all or for a time sufficient to accomplish sensible olivine grain growth. For testing we added grain size data in the non-linear minimum square fit off by 20% from the theoretical value computed with the growth parameters reported in section 4.3. This drift with respect to calculated grain sizes is double then the average error on grain size measurement for each experiment. The results of this exercise indicate that adding further experiments would not perturb the calculated n and E_a beyond the maximum and minimum values computed beforehand (see section 4.3). Only when adding multiple fictive data and selecting a worst case scenario, the drift become significant (Test 7 of Table D1). Yet, a single experiment for 1,000 hours off the computed growth rate by 20% would affect the calculations of n and E_a more than the aforementioned combination of several additional experiments with $t < 360$ h (cf. Test 7 and Test 8 of Table D1). Technically, the conclusion is that all studies without experiments on timescale of thousands of hours could grossly over- or underestimate growth rate, independently of the total number of runs performed. This requirement is beyond the technical capability of most high-pressure apparatuses and/or availability of an instrument for several weeks without interruption.

In order to further investigate the quality of our experimental data, analysis of the verification of Zener relation shall be performed. This requires assessment of the grain growth rate of the competing coarsening phase, in this case, solid Fe-Ni. In comparison to experiments with a single matrix phase (e.g., solid Ni or molten Fe-S – Guignard et al. 2012, 2016, Solferino et al. 2015), the texture of the run products for olivine plus Fe-Ni and Fe-S is far more complex and individual BSE images do not allow to distinguish between quench product of Fe-S liquid and solid Fe-Ni to an extent sufficient to perform digital image analysis with an acceptable precision (attempts conducted for runs FS9010-2 and FS9010-5 yielded errors equal to 85 and 76% of the average grain size, respectively for repeated measurements and following the same procedure employed for olivine).

Therefore, we conclude that all considered, we have provided a far more realistic estimate of olivine growth rate with respect to the grain growth law computed in Solferino et al. (2015). Nevertheless, in order to comprehensively address the definition of grain growth of olivine in partially molten Fe-Ni-S, we suggest that further investigation should be carried on. It would be necessary to perform runs with extremely long annealing for $Fe_{90}S_{10}$ matrix composition, and to determine the grain growth parameters for other compositions of the Fe-Ni-S, completing the series initiated by FS8020 and FS9505 (Table 1). This is of further interest considering that our experiments show unique characteristics neither seen in subsolidus olivine+metal experiments nor in olivine+fully molten metal systems.

Table D1: Result of statistical analysis of the effects yield by introduction of fictive data to the non-linear least square calculation of olivine grain growth parameters.

	Offset of fictive data	**Fictive T (°C)**	**Fictive duration (h)**	***n***	***E*a (kJ mol^{-1})**
Test 1	Increase 20%	1300	96	4.311	118.7
Test 2	Reduction 20%	1300	96	3.258	71.6
Test 3	Increase 20%	1100	300	3.304	65.0
Test 4	Reduction 20%	1100	300	4.309	116.4
Test 5	Increase 20%	1300	250	3.491	107.6
Test 6	Reduction 20%	1300	250	4.199	91.2
Test 7	Worst case [a]			**5.302** [b]	**134.8**

Test 8	Increase 20%	1300	1000	**2.803**	**73.7**
Test 9	Reduction 20%	1300	1000	**5.306**	**135.2**

[a] worst case scenario attained by adding to least square calculation Test1, Test 4, and Test 6 data points

[b] bold font used for data outside of the min-max bracket

Reference values for n and E_a

$n = 3.703$ $n_{max} = 4.313$ $n_{min} = 3.093$

$E_a = 101.0$ kJ mol^{-1} $E_{a\,max} = 119.2$ kJ mol^{-1} $E_{a\,min} = 81.6$ kJ mol^{-1}

Appendix E: Energetics and dominant growth mechanism for olivine coarsening in Fe-Ni-S

The apparent anomalous E_a, i.e., 101.0 kJ mol^{-1}, computed for olivine grain growth in this study cannot be matched with any known value (e.g., E_a for atomic diffusion of Mg, Fe, Si, O through olivine lattice or along olivine grains surface), in that it is less than half of the smallest computed or calculated activation energy for any of the cited processes (e.g., Houlier et al. 1988, 1990, Gerald and Jaoul 1989).

A possibility remains that the computed activation energy is an apparent value. This scenario implies that different processes control olivine grain coarsening at the two explored temperatures. This is unlikely but not impossible, since the smaller fraction of Fe-S liquid at 1100 °C may imply that Fe-Ni coarsening at this temperature happens chiefly via grain boundary diffusion at Fe-Ni grains contacts, while at 1300 °C diffusion of Fe and Ni appears to be prevalently mediated by liquid Fe-S (section 5.3, figure E1a,b). Expanding the experimental dataset by investigating temperatures of 1150, 1200, 1250 and 1350 °C might help to answer this question, but it is beyond the purpose of this investigation.

In order to interpret the process leading to olivine grain size increase in Fe-Ni-S matrix we employed a mathematical fit (to eq. 1, section 3 and 4.3) and textural evidences (section 5.3). The shape of NGSD could also be used for this purpose, like done by Solferino et al. (2015) for the olivine + liquid Fe-S system. From a thorough perusal of theoretical NGSDs, all distributions relative to Ostwald ripening must be excluded, since this occurs only when the growing phase is dispersed in a matrix into which it has finite solubility, both conditions unsatisfied by our experiments. Several studies outlined the chief geometrical characters of NGSDs calculated for normal grain growth (NGG), where grains of the coarsening phase are largely in mutual contact, thus applicable to our system. The best match for all of the NGSDs of experiments used to compute olivine grain growth parameters (see Table 1 and Appendix D) is with theoretical distributions calculated for the coalescence of the primary solid due to grain boundary migration of a second phase (e.g., Takajo et al. 1984, Evans et al. 2001). Specifically, all of the aforementioned experimental NGSDs fit with the theoretical boundaries: $d_m < 1.0$ and $2.0 \leq d_{max} \leq 3.0$ (within error). This is also in good agreement with our interpretation of olivine coarsening in Fe-Ni-S matrix (see section 5.3), except for the additional role of FeS liquid, as illustrated by figure E1.

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Appendixes figure captions:

Figure A1: BSE image of FS9505. The dark grey phase is olivine, light grey is Fe-Ni and medium grey phase is pseudo-Mg-ferrite. Pseudo-Mg-ferrite is often located at olivine triple/quadruple junctions similarly to Fe-S liquid location observed in other runs.

Figure A2: BSE and X-ray elemental maps of an area of FS9505. (a) BSE image where no Fe-S is clearly evident. (b) Elemental map of Fe. (c) Elemental map of Mg. (d) Elemental map of S. (e) Elemental map of Si. (f) Color scale for all elemental maps. Elongated and pod-like features containing no Mg and Si, with an iron content intermediate between olivine and Fe-Ni and high sulfur concentration are Fe-S liquid pools or pockets. Pseudo-Mg-ferrite is easily distinguished from Fe-S, due its non-negligible Mg and Fe content, while Si and S are absent. It is interesting to notice that a lower Fe concentration halo (light green color – block b) seems to surround all Fe-Ni pods, perhaps indicating formation of thin films of Fe-S liquid between Fe-Ni and olivine grain boundaries. A different explanation could be an effect due to X-ray scattering on topographic discontinuities at olivine-Fe-Ni contacts generated during polishing.

Figure B1: (a) NGSDs for Brenham and Springwater pallasites and theoretical distributions for second order surface diffusion controlled (s.o.d.c.) grain growth, Rayleigh, and log-normal. (b) NGSDs for Brenham and Springwater pallasites, and OFS-10 (from Solferino et al. 2015) and FS9010-5 experiments.

Fig. C1: Time of core crystallization (red), time window material at top (dark blue) and bottom (light blue) of depth range satisfying cooling rate constraints passes Curie temperature for different planetesimal radii.

Figure D1: Comparison of data with calculated olivine grain growth using the parameters reported in section 4.3. The model fit with data to a deviation of less than 5%.

Figure D2: Normalized grain size distribution of four runs with $Fe_{90}S_{10}$ alloy. A noticeable double peak is evident for 50 h annealing at 1100 °C.

Figure E1: Olivine and Fe-Ni coarsening in 'olivine + Fe-Ni + Fe-S liquid' system. A: At the start of the process some Fe-Ni is trapped along olivine-olivine grain boundaries in the form of small pods, effectively hindering olivine grain boundary movement. B: Fe-Ni diffusion, mediated and catalysed by thin films of Fe-S liquid, drives all Fe-Ni into olivine-olivine-olivine triple junctions. C: Olivine grain boundaries that are not pinned by Fe-Ni any longer are free to move, promoting grain growth of the former phase. At the same time Fe-Ni diffusion from triple junction to triple junction takes place. D: Eventually a status of apparent equilibrium is reached, where a single mass of Fe-Ni is present at the triple junction between the three remaining olivine grains. It is evident that the grain size of olivine and Fe-Ni simultaneously increase during this process. Occasionally, after Fe-Ni removal from olivine-olivine-olivine triple junctions, small amounts of Fe-S liquid are left behind (see Fe-S in panel C), as observed in experimental samples (figure 1c and A2b).