Suitability of chondrules for studying the magnetic field of the early solar system: An examination of synthetically produced dusty olivine

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Abstract

Chondritic meteorites are rare, yet incredibly valuable windows into the geophysical and geochemical environment of the early solar system. Dusty olivine grains containing exsolved nanometer scale iron nickel alloy inclusions are present in many chondritic meteorites and their remanent magnetization may give insight into the strength of the solar dynamo at the time of chondrule formation. Laboratory methods for determining the paleointensity of these rare materials must be optimized prior to conducting experiments on actual meteorite samples. To this end, we have used high temperature recrystallization techniques to produce synthetic dusty olivine samples with textures remarkably similar to those observed in chondritic meteorites.

The olivine grains used in these annealing experiments are from the 13 kya Haleyjabunga picritic basalt flow in Iceland and have compositions of Fo90, which closely resembles the olivine composition observed in chondritic meteorites. Samples were annealed at 1350 °C, 1315 °C and 1425 °C either under vacuum in the presence of graphite or under controlled oxygen fugacity using pure CO gas. The laboratory produced magnetic mineral assemblages in 4 different types of samples as well as the starting material have been characterized using low and high-temperature magnetic measurements, hysteresis loops, FORC diagrams, and scanning electron microscopy. The room temperature remanence properties of these materials have been explored using stepwise IRM and ARM acquisition and alternating field demagnetization. These synthesis techniques allow us to produce a wide range of iron nickel grain sizes with correspondingly large variations in coercivity (between 0 and 500 mT). High-temperature measurements of saturation magnetization show that all the samples reach their Curie temperatures at ~760 °C, consistent with kamacite, a low Ni high Fe metal alloy. Multiple experiments have shown that care must be taken to rigorously control the atmosphere in which the samples are heated and cooled in order to avoid forming trace amounts of magnetite on the surface of the samples. Future research will explore the feasibility of using modified Thellier protocols or the Shaw method to determine the paleointensity of laboratory induced thermoremanent magnetizations.
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Suitability of chondrules for studying the magnetic field of the early solar system: An examination of synthetically produced dusty olivine

1. Introduction
Chondrules are rounded, millimeter-sized objects comprising several different silicate phases (e.g., olivine & pyroxene) that formed during the earliest stages of our solar system’s evolution. They represent the first steps in the transition from protoplanetary disks of dust and gas to massive planet-sized objects. Despite their cosmological importance, researchers still do not agree on the mechanism and location of chondrule formation within the early solar system. Currently, the most popular theory for chondrule formation is due to heating by shock waves through an accretionary disk, although the source of the shock is unknown (Desch et al. 2005). A competing model suggests that chondrules may have formed very close to the young Sun, where magnetic reconnection events caused an instability in a region called the ‘X-point’, and were then flung to asteroidal distances by an ‘X-wind’ (Shu et al. 2000). Still another possible formation mechanism for chondrule is by impact of pre-formed planetesimals (Hutchinson et al. 2005).

The two main proposed formation sites for chondrules – either 0.1 AU from the Sun in the X-wind model or 3 AU from the Sun for the shock wave and impact models – correspond to locations with very different magnetic field strengths. Young stars produce strong fields. The equatorial field strength of a T-tauri star is estimated to be around 0.21 T (Shu et al. 2000). Objects that condense or crystallize within 0.1 AU of the young Sun are expected to acquire a very strong remanant magnetization. The magnetic field within the accretionary disk itself is unknown, but is thought to be orders of magnitude less than that of younger stars.

This study lays the foundation for a larger investigation of the remanent magnetization (RM) of chondrules, a property that may provide critical clues about their initial formation environment. The RM of meteorites records the intensity of the magnetic field present during their initial formation and subsequent alteration and metamorphism of the meteorite parent body. Multiple magnetization events can in many cases be unraveled during stepwise thermal or alternating-field demagnetization experiments, yielding important information about the magnetic and thermal conditions present during pre-and post-accretionary processes (Sugiura and Strangway 1988).
Previous estimates for the pre-accretionary magnetic field intensity are of the order 0.2-0.7 mT based on Thellier-Thellier studies (Lanoix et al. 1978). However, the heterogeneous nature of RM within a single meteorite may result in significant underestimation of the field intensity. Recently there have been significant improvements in rock magnetic techniques and instrumentation sensitivity, allowing for detailed characterization of mineral grains separated from chondritic meteorites. A key innovation in this area will be paleointensity studies at sub-chondrule length scales. Modern cryogenic magnetometers are sensitive enough to measure RM from individual silicate crystals, allowing the pre-accretionary remanence signal to be more effectively isolated. With the recent demonstration that ‘dusty’ olivine grains (i.e. olivine grains within chondrules containing inclusions of small Fe-Ni particles; Figure 1) have the potential to acquire and retain a primary component of thermoremanent magnetization (TRM) associated with chondrule formation (Uehara and Nakamura 2006), a comprehensive study of meteorite magnetism is now particularly timely.

Most traditional paleointensity studies begin with a pilot study on a subset of samples that characterizes their magneto-mineralogic assemblage and determines the temperature range over which they acquired their thermal remanence. This information is used to optimize the experimental protocol and determines which temperature steps will be used for thermal demagnetization and acquisition of partial thermoremanent magnetization (pTRM). Without such pilot studies, it is much more difficult to accurately calculate the paleointensity for a sample and to interpret its geological significance. Yet, the rarity of chondritic meteorite samples and the need to not alter their original mineral assemblage makes it impossible to conduct such pilot studies on chondrules.

To overcome this hurdle, and ultimately, to obtain reliable paleointensity measurements from chondrules, we have successfully produced a suite of synthetic dusty olivine samples and analyzed their magnetic carriers and properties. The distribution of Fe-Ni inclusions inside the synthetic dusty olivine grains is similar to the inclusion textures observed in natural dusty olivine grains in chondritic meteorites (Figure 1). The growth conditions for these synthetic materials can be carefully controlled in the lab to reproduce a number of different Fe-Ni inclusion textures commonly observed in olivine grains within chondrules. How olivine responds to alteration in terms of oxidation and reduction has been elucidated by Nitsan (1974, Figure 10). Additionally, the ambient magnetic field in the lab can be adjusted to learn more about how these inclusions...
acquire thermoremanent magnetization. In this study, we use synthetic materials as analogues for chondritic dusty olivines so that we may more accurately measure the paleointensity from extraterrestrial samples in the future.

2. Methods

2.1 Sample preparation

Samples were synthesized at the Department of Earth Sciences, University of Cambridge, UK. The starting material is a mixture of finely ground olivine (<75 μm) from the 13 ka Haleyjabunga picritic basalt flow in Iceland and high purity graphite powder. The olivine has an average composition of FeO90, which closely resembles the olivine composition observed in natural chondritic meteorites (Jones and Danielson, 1997). Samples were ground in an agate mortar under acetone for approximately 5 minutes and loaded into cylindrical capsules of electrolytic graphite, which were sealed using alumina-based cement (Zircar). It is important to note that all synthetic samples are polycrystalline with little to no observable mineral fabric.

Two different methods for reducing the olivine-graphite mixtures were explored. Sample 3 was sealed inside an evacuated quartz tube (fO2 was unknown) in an effort to minimize oxidation during heating and cooling in a muffle furnace. All other samples were placed in platinum crucibles and heated and cooled in a gas-mixing furnace under a stream of pure CO gas. Sample temperatures were monitored via a thermocouple located on the inside of the crucible. The thermal treatment details for all four synthetic samples reported in this study are shown in Table 1. All samples were cooled at a rate of 30°C per min until 750°C, near the Curie temperature of pure Fe, whereupon they were air or water quenched to room temperature (Table 1). Measured cooling curves of sample 3 and sample 4 are included at the end (Figure 14).

The thermal treatments for Sample 3 and 4 were based on the experimental design of Uehara and Nakamura (2006), who aimed to simulate chondrule formation through flash-heating in the presence of a magnetic field. The heating schedule for Sample 5 was designed to explore the effects of reduced peak temperatures and heating times on Fe-exsolution in olivine. Sample 6 was prepared using re-ground fragments of Sample 5 in order to investigate the effects of reheating on the olivine exsolution textures. By varying these parameters we hoped to come to a better understanding of the processes that lead to different exsolution textures in natural dusty olivine samples.
2.2 Measurements and Instrumentation

A broad range of rock magnetic analyses were conducted on the synthetic dusty olivine samples in order to identify their constituent magnetic phases and determine their magnetic acquisition and demagnetization behavior. All measurements were performed at the Institute for Rock Magnetism, University of Minnesota, Minneapolis, USA.

Samples’ magnetic remanence was measured using a 2G Enterprises superconducting rock magnetometer situated in a magnetically shielded room with a background field of <500 nT. Samples were demagnetized using alternating fields (AFs) up to 200 mT. Thermal demagnetization was not used in order to avoid oxidation of the Fe-Ni inclusions hosted by the olivine grains. Room temperature anhysteretic remanent magnetizations (ARMs) were imparted using a Schonstedt demagnetizer with a 0.1 mT DC field superimposed over a peak AF=200 mT (decay rate 0.005 mT/half-cycle). Room temperature isothermal remanent magnetizations (IRMs) were imparted in fields up to 1400 mT using an ASC pulse magnetizer.

Low-temperature magnetic properties were measured on a Quantum Design Magnetic Properties Measurement System (MPMS2) cryogenic susceptometer. In-phase (χ′) AC susceptibility was measured between 10 K and 300 K using a 238.7A/m(3 Oe) field and frequencies of 1, 3, 10, 30, 100, 300 and 1000 Hz in the starting materials. Samples 3, 4, 5 and 6 were cooled from 300 K to 20 K in a field of 2.5 T (“field cooling”, FC), and remanence was measured at 5 K intervals during warming back to 300 K. Samples were then cooled back to 20 K in zero field (“zero field cooling”, ZFC), given a 2.5 T SIRM, and remanence was again measured at 5K intervals during warming to 300K.

Room temperature hysteresis loops and first-order reversal curves (FORCs) were measured on a Princeton Measurements Corporation MicroMag Vibrating Sample Magnetometer and the measurements were processed using FORCinel v1.13 by Harrison & Feinberg (2008). Also, hysteresis loops were measured on Sample 3 and Sample 4 in continually streaming Helium every 10 degrees from 30°C to 770°C, with temperature varied at a rate of 10°C per minute. Saturation Magnetization (Ms) as a function of temperature were measured on selected samples between 30°C and 770°C.
3. Results

3.1 Electron Microscopy
Backscattered electron (BSE) images reveal two populations of Fe-Ni-Cr grains in all samples of experimentally reduced olivines (Figure 2). One population appears as submicron-sized inclusions in the interior of the olivine grains. These inclusions occur along (100) and (001) within the olivine grains, and give the silicate its distinctive dusty appearance. These same preferred orientations of metallic inclusions have been noted in previous studies of experimentally altered olivine (Boland and Duba 1981, 1986; Lemelle et al. 2000) as well as in meteoritic dusty olivine grains (Rambaldi and Wasson, 1982; Jones and Danielson, 1997). The second population of Fe-Ni-Cr grains occurs as coarse (5–15 μm), equidimensional crystals along olivine grains boundaries.

3.2 Remanence Acquisition and Demagnetization Behavior

3.2.1 “Natural” remanent magnetization and Demagnetization
The synthetic chondrule samples carry a strong “natural” remanent magnetization (NRM) on the order of $10^{-2} - 10^{-4}$ Am$^2$/kg that is resistant to full demagnetization by alternating fields. Significant amounts of NRM remain even after exposure to peak AF fields of 100–200 mT (Table 2). AF demagnetization at 100 mT removed 93.3% of the initial NRM of sample 3 and AF demagnetization at 200 mT removed 80.9%, 83.1% and 83.1% of the initial NRM of sample 4, sample 5 and sample 6, respectively. The median destructive fields (MDF) of samples 4, 5 and 6 are > 100 mT, while sample 3 shows an MDF of 28 mT. Less than 5% of sample 3’s magnetization remains after AF demagnetization at 67 mT.

Demagnetization steps for all the samples on vector component Zijderveld diagrams show approximate univectorial trajectories toward the origin and tight clusterings on equal area plots (Figure 4). These results indicate that samples acquired a thermoremanence on cooling with minimal amounts of sample movement during quenching.

3.2.2 Anhysteretic Remanent Magnetization and Demagnetization
Sample 5 and sample 6 show similar demagnetization spectra, MDFs, and ratios of remaining moment to the initial moment of ARM and NRM demagnetization (Figure 3c, d). In contrast, ARM and NRM demagnetization spectra for sample 3 and sample 4 are different (Figure 3a,b), suggesting that ARM is not a suitable analog for NRM for paleointensity studies (Table 3).
3.2.3 Isothermal Remanent Magnetization and Demagnetization

The IRM acquired by the synthetic samples is about 2 orders of magnitude greater than the initial NRM (Table 4). Sample 5 and sample 6 reached 50% of their magnetic saturation in fields ~125 mT and ~150 mT, and 95% of saturation in fields~325 mT and ~525 mT, respectively (Figure 5c,d). The acquisition curves for both samples plateau eventually by 1.2 T.

In contrast, the IRM acquisition curves for sample 3 and sample 4 did not reach saturation and the operations were aborted at 200 mT as a result of improper designed plan (Figure 5a, b), so it is hard to estimate at what point these two samples would become fully saturated.

The “R ratio” is the intersecting point between IRM acquisition and demagnetization and values <0.5 indicate a degree of particle interaction or multi-domain behavior (Cisowski, 1981). Sample 5 and sample 6 yielded R ratios of 0.33 and 0.4, respectively, which is in agreement with results from FORC measurements (section 3.4). R ratios were not determined for samples 3 and 4 because they were not fully saturated in the acquisition process.

3.3 Low temperature magnetic measurements

The results of the low-temperature FC/ZFC SIRM measurements in sample 3, 4, 5 and 6 and low-temperature AC susceptibility measurement in starting materials are displayed in Figure 6. In the starting materials, $1/\chi$ as a function of temperature is a straight line, indicating that the starting material is paramagnetic. In samples 4 and 5, the remanence was slightly higher for FC experiments ($M_{R, FC}$) than ZFC ($M_{R, ZFC}$). Plots of the first derivative of the data for samples 3, 4, and 5 consistently display peaks between 100K and 130K, which we interpret as evidence for the Verwey transition of magnetite. A significant difference $M_{R, FC}>M_{R, ZFC}$ below the Verwey transition typically indicates a predominance of stable single-domain (SSD) grains in samples containing pure magnetite (Moskowitz et al.1993.).

The low-temperature remanent magnetization of sample 6 displays no evidence of the Verwey transition. Both the FC and ZFC measurements decrease quasi-linearly with temperature from 20 K to 300 K. The first derivative of sample 6’s low temperature measurements does not indicate change in slope between 100 and 130K as observed in sample 3, 4 and 5. A thin film of pure iron exhibits low-temperature behavior similar to that of sample 6. The remanences decrease monotonically as the temperature progressively approaches room temperature.
3.4 First order reversal curves (FORCs)

FORC diagrams show significant differences in the distribution of coercivities for these synthetic samples. (Figure 7) The FORC distribution of starting material required an unusually high smoothing factor (SF= 12) because of the very weak ferromagnetic signal, and shows no distinguishable features other than a weak peak near $H_c = 5$ mT. In contrast, the FORC distributions of the synthetic samples extend noticeably further along the $H_c$ axis, indicating the presence of magnetic minerals produced during reduction of the olivine.

Sample 3 (Figure 7 B) shows a distinct coercivity peak centered along the $H_c$ axis at ~25 mT. The center of the coercivity distribution spreads ± 20mT in $H_u$ direction, which may be attributed to the particle interaction within the sample.

Samples 4, 5, and 6 (Figure 7 C, D, E) show at least two populations of magnetic grains. One population of grains shows coercivities ranging between 5 and 500 mT indicating a wide particle size distribution which plots directly along the $H_u = 0$ axis. This population consists of largely non-interacting, stable single domain grains. The coercivity of single domain grains of magnetite grains has a theoretical upper limit of ~300 mT (Dunlop and Özdemir, 1997), and consequently, the $H_c$ values >300 mT in the synthetic samples suggest a composition other than pure magnetite. We interpret this population to represent finely exsolved Fe-Ni metal within the olivine.

A second, magnetically softer population of grains is observed in samples 4, 5 and 6. This soft component is not immediately recognizable in the FORC diagram of sample 4. However, its presence is clear when the peak $H_c$ values in the FORC diagram (~125 mT) are compared with those in the marginal coercivity distribution (~30 mT), a coercivity distribution obtained by adding each vertical column of the FORC distribution. Close examination of the FORC diagram shows a diffuse region of strongly interacting, single domain grains between $H_c = 25$ and 75 mT similar to those observed in sample 3. This example of “magnetic unmixing” is one example of the power of FORC analyses in characterizing magnetic mineral assemblages.

The soft components in samples 5 and 6 are different from the soft component in sample 4. In samples 5 and 6, the soft components have $H_c$ values <25 mT with contours running roughly parallel to the vertical axis of the graph. Previous researchers have associated similar distributions with populations of pseudosingle- and multi-domain grains (Muxworthy & Dunlop, 2002; Loock
et al., 2008). It is difficult to ascribe a composition to the soft components in samples 4, 5, and 6 using only the FORC diagrams. A synthesis of the magnetic mineral assemblage is presented in the Discussion.

3.5 High temperature magnetic measurements

3.5.1 Saturation Magnetization as a function of temperature

Thermomagnetic data for both sample 3 and 4 were analyzed using the first derivative method (Figure 8a). Two Curie points are found for sample 3 at ~575°C (848K) and ~750°C (1023K), for sample 4 at ~585°C (858 K) and 770°C (1043 K). The higher Tc is indicative of Ni-poor Fe-Ni alloy, while the lower Tc corresponds to magnetite. More than half of the saturation magnetization in these specimens is carried by the high-Tc phase.

Thermomagnetic data from sample 5 and sample 6 are notably different from that of sample 3 and 4 (Figure 8b). The heating and cooling curves for sample 5 and sample 6 are non-reversible, indicating that the samples were mineralogically altered during heating.

Saturation magnetization (Ms) of sample 5 decreases smoothly up to ~300 °C, at which point Ms increases abruptly on further heating until a peak at ~370°C. Ms remains constant until ~400°C, where it begins to progressively decrease. Two minor inflection points are observed at ~575°C and ~725°C, respectively. The elevated Ms value at ~400°C is not present on cooling, although there is an inflection occurred at ~220°C. Room temperature saturation magnetization is slightly greater after thermal cycling than the original starting value. This suggests that a mineralogical change occurred, rather than a non-saturated moment in a high-anisotropy unblocking phase near 300°C.

The Ms(T) curve of sample 6 is similar to that of sample 5. However, the absence of an inflection point after 400°C during heating and the existence of an inflection point at ~575°C during cooling is notable.

3.5.2 Susceptibility as a function of temperature

In addition to the strong-field measurements described above, low-field susceptibilities were measured as a function of temperature on sample 6 and a subsample taken from the exterior of sample 5(Figure 9). The susceptibility as a function of Temperature curves broadly resemble the
Mₛ-T curves described above. There are several distinct features about these two diagrams: (1) the magnitude of “hump” formed above 300˚C decreases after each successive heating cycle and, (2) the final room temperature susceptibility after the second and third runs is greater than the initial starting value before heating.

4. Discussion

4.1 Mineral alteration and phase transformation during high temperature magnetic measurements

To explain these complicated thermomagnetic data, we tentatively propose the following explanation:

First, for the elevation of saturation magnetization between 300˚C and 400˚C, we tend neither to interpret that as evidence of oxidation of olivine, a reaction that may potentially form magnetite, nor reduction of olivine through which Fe could precipitate out (Table 5). We ran one independent thermomagnetic experiment on the starting material and found no notable increase of magnetization (Figure 8) during thermal treatment. If magnetite is generated as a result of oxidation of olivine, a similar “hump” should be observed within the same temperature range. Also, the lack of inflection between 575˚C ~ 585˚C on the heating curves of several independent thermomagnetic experiments on sample 6 not only confirms the absence of magnetite in sample 6, but also once again indicates that the enhancement of Mₛ should not be attributed to the oxidation of olivine (Figure 8). In addition, reduction of olivine does not seem to be plausible in elevating the Mₛ either, because reducing agents are required to reduce the olivine to precipitate Fe out and we did not find any evidence of graphite on the SEM images and there are literally no other reducing agents involved in the measurements. Most importantly, all previous experiments that could successfully generate Fe-Ni inclusions were conducted at high temperature (>1000˚C) (Uehara and Nakamura, 2006; Cohen et al.2004; Leroux et al.2003; Libourel, 1995; Connolly et al.1994; Boland and Duba, 1981).

Second, the enhancement of Mₛ should not be attributed to the oxidation of Fe-Ni alloys to magnetite (Fe₃O₄) or trevorite (NiFe₂O₄). Though both of these two minerals have Curie points near 585˚C, their saturation magnetizations are much lower than that of pure Fe or Fe-Ni alloy with low Ni content, which would otherwise lead to the decrease of Mₛ.
Therefore, the remaining mechanism that could cause this abrupt enhancement of \( M_s \) around 300 °C may be related to the intrinsic property of Fe-Ni. Here we interpret enhancement as a consequence of the transition of \( \gamma \) Fe-Ni, a paramagnetic phase to \( \alpha \) Fe-Ni, a ferromagnetic phase. XRD analysis done in University of Cambridge shows that a large proportion of Fe-Ni is quenched into the \( \gamma \) phase during rapid cooling simply because \( \gamma \) phase Fe-Ni is kinetically limited to be quenchable. This contrasts with \( \alpha \) Fe-Ni phase that is not found by rapid quenching, so it is possible to have a mixture of \( \alpha \) Fe-Ni phase and \( \gamma \) Fe-Ni phase in the sample before heating and the thermodynamically unstable \( \gamma \) Fe-Ni phase could be converted into \( \alpha \) Fe-Ni phase around 300 °C and this conversion subsequently leads to a sudden appearance of a magnetic signal.

In view of presence of Fe-Ni alloys with various Ni content as thermodynamically unstable phases that could be subject to complicated phase transformation below 500°C (Cacciamani et al. 2006; Yang et al. 1996; Zhang et al. 1994; Reuter et al. 1989; Lin and Chang, 1989), we feel that interpretation of the abrupt change of \( M_s \) near this temperature range is perhaps to be viewed with reservation. The exact mechanism of this is presently unresolved and probably will remain so until such a time as we understand it in detail.

Above 500°C for sample 5, considering that there are trace amounts of magnetite before heating, the presence of an inflection between 575°C~585°C on the heating curve could be explained as the ferrimagnetic-paramagnetic transition of native magnetite in sample 5. The coexistence of iron and magnetite above 585°C will cause an irreversible phase change leading to the formation of iron and wüstite while simultaneously consuming magnetite (Figure 11 Lindsley, 1991). The absence of inflection between 575°C~585°C characteristic of magnetite Curie point on cooling curve may be explained by this transformation. In additional to the phase diagram mentioned above, a phase diagram of Ni percentage in Fe versus temperature (Figure 12) should be taken into consideration as well to explain the rest of the observations. Based on this diagram, Ni poor Fe-Ni alloys above 500 °C are \( \alpha \)-phase and could transform to \( \gamma \)-phase at the curve separating \( \gamma \) phase zone from \( \alpha+\gamma \) phase zone. This phase transformation is Ni content dependent and always occurs at relatively higher temperature as \( \alpha \) phase transforms to \( \gamma \) phase on heating, whereas change of \( \gamma \) phase to \( \alpha \) phase will take place at a much lower temperature on cooling. This thermal hysteresis will become increasingly larger as Ni content in Fe progressively increases. According to the microprobe data obtained in University of Cambridge, Ni content in sample 5 is
determined to be less than 5 atm. %. When combining this result with the large thermal hysteresis observed on the $M_s$-T curves, we may explain the inflection~725°C on heating curve as a possible result of metal phase change instead of Curie point of some minerals.

For sample 6, since the starting material is re-ground of sample 5, Ni content should be further depleted after thermal treatment and this has been confirmed by the microprobe data (<3 atm. % Ni). Therefore, according to the equilibrium Fe-Ni phase diagram (Figure 12), the transformation of $\alpha$ phase to $\gamma$ phase on heating is expected to occur at higher temperature (>800°C) compared to that of sample 5 and subsequently displays a smaller thermal hysteresis on cooling than that of sample 6. This is in agreement with our observation and could possibly explain why there aren’t any inflections found between 400°C and 770°C during heating of sample 6.

Above 700°C, iron (especially fine-grained particles) will oxidize even in high-vacuum systems (Dunlop, 2001). Only heating in some regulated gas mixture will prevent this from happening. Given the fact that the sample is measured in helium and there are some Fe-Ni blebs devoid of protection from host olivine, oxidation is inevitable. This can explain why there is an inflection point between 575°C~585°C indicative of magnetite upon cooling of sample 6. The schematic diagrams on heating and cooling of sample 5 and sample 6 are concluded to display these complicated transition processes (Figure 13).

4.2 *Similarity of synthetic dusty olivines to naturally occurring chondrules*

Textural features of these synthetic samples are quite similar to those observed in dusty olivines from natural chondrites such as Semarkona (LL3.0), Bishunpur (LL3.1), Krymka (LL3.1), Chainpur (LL3.4), Ragland (LL3.4), Inman (L3.4), Murchison (CM2) (Rambaldi and Wasson, 1981 & 1982; Jones and Danielson, 1997; Leroux *et al.* 2003; Cohen and Hewins, 2004). Both the natural chondritic olivines and synthetic olivines contain submicron-sized metallic inclusions with crystallographic alignment along (100) and (001) alignment in the grains cores and larger (5-15 μm) metallic grains dispersed along the edge of host olivine. This phenomenon has been reported by Boland & Duba (1981) and Jones & Danielson (1997) and results from nucleation and metal growth in olivine during reduction.
In addition to the mineralogical similarities observed on the microscopic scale, these synthetic samples are comparable to natural chondrules in terms of their FORC distributions on the macroscopic scale. We compare our findings here with those of Acton et al. 2007.

The FORC distribution of sample 6 resembles that of chondrules from the Allende meteorite, a CV3 chondrite extensively studied by numerous rock magnetists. (Butler, 1972; Banerjee and Hargraves, 1972; Brecher and Arrhenius, 1974; Herndon et al. 1976; Sugiura, Lanoix and Strangway, 1979; Nagata and Funaki, 1983; Brenker and Krot, 2004; Weiss et al. 2009). Both sample 6 and Allende chondrules show a magnetically hard component with minimal magnetic interactions with coercivities beyond 300 mT along the $H_u = 0$ axis and a magnetically soft component running roughly parallel to the vertical axis of the graph. We attribute these components to the presence of single domain Fe-Ni inclusions and the multi-domain Fe-Ni blebs, respectively (Figure 7E). For comparison, FORC diagram of an Allende chondrule has been added to the figure.

Sample 5 also shares many similarities with the FORC distribution of Allende chondrules. However, it displays greater interactions at low coercivities (25-50 mT) than at higher coercivity range (>150 mT) (Figure 7D). Samples 3 and 4 do not resemble any previous published FORC distribution for any natural chondritic sample.

Though the remarkable similarities in terms of micro and macro structure between our synthetic samples and natural chondrules imply successful synthesis, one important difference exists: Our synthetic samples are only dominated by one type of Fe-Ni alloy (kamacite with <7 atm. % Ni content) while there are normally several other kinds of Fe-Ni alloys including taenite (>7 atm. % Ni content) and tetrataenite (normally~ 50 atm. % Ni content) (Butler R., 1972; Clarke R. & Scott E. 1980; Nagata T. & Funaki M., 1983; Morden S. & Collinson D., 1992) detectable in the natural chondrules. The presence of this suite of complicated magnetic mineralogy in the natural chondrules inevitably gives rise to the number of remanence carriers and therefore makes it hard to understand and interpret the manner as well as the history of remanence magnetization.

5. Conclusion
Our synthetic dusty olivine-bearing samples have mineral textures and rock magnetic properties similar to chondritic dusty olivines and these dusty olivine-bearing samples contain Fe-Ni
inclusions capable of recording magnetizations that could be stable over billions of years. By varying sample preparation methods, we could precisely control the concentration and composition of inclusions. Drop-quenching samples in water minimizes the production of magnetite.

6. Future Research
We intend to continue variation of synthesis parameters. (e.g., maximum temperatures, cooling rate, atmosphere control and quenching method) to produce a suite of synthetic dusty olivine samples for use in paleointensity experiments. (Prepared in known fields and heating & cooling sequences). We are hoping to use these synthetic samples to optimize our paleointensity protocols for easily-altered minerals such as Fe-Ni alloys. Ultimately, we hope to conduct experiments on single crystals of natural dusty olivine collected from minimally altered chondritic meteorites.

In order to achieve these goals, there are several issues that should be taken into consideration: First, the overall Ni/Fe ratio in synthetic samples is much lower than that of natural ones. Our samples are more like chondrules formed after recycling of their precursors containing higher Ni content and subsequently have undergone Ni depletion. Therefore, our samples may not represent the most primitive chondrules. Future experiments should use starting materials with higher Ni contents to better simulate natural ones.

Also, it’s essential to conduct detailed investigation of the unit cell and crystal structure of Fe-Ni within the samples prior to any high-temperature measurements (e.g. M_s-T, Thelier-Thelier and Shaw method). This effort will provide us perspective other than traditional rock magnetism methods into the suitability of samples for paleointensity research.

Finally, furnaces capable of attaining higher temperature (~800°C) are required to observe the Curie temperature of Fe in both high-field and low-field measurement. To prevent or minimize oxidation-reduction reactions that might occur during thermomagnetic experiments and future paleointensity experiments, the oxygen fugacity at any moment must be monitored by measurement, and a mixture of H₂/N₂, CO/CO₂ or H₂/CO₂ gasses flowed past the sample is preferred to maintain the silicate mineral within its stability field with respect to oxidation and reduction.
Tables

Table 1. Sample preparation details

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Starting Material</th>
<th>Furnace</th>
<th>Peak Temperature</th>
<th>Time at Peak Temp.</th>
<th>Quenching Environment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 3</td>
<td>Olivine/graphite</td>
<td>Muffler</td>
<td>1350˚C</td>
<td>10 min.</td>
<td>Evacuated quartz tube</td>
</tr>
<tr>
<td>Sample 4</td>
<td>Olivine/graphite</td>
<td>Gas-mixing</td>
<td>1350˚C</td>
<td>10 min.</td>
<td>Air</td>
</tr>
<tr>
<td>Sample 5</td>
<td>Olivine/graphite</td>
<td>Gas-mixing</td>
<td>1315˚C</td>
<td>3 min.</td>
<td>Air</td>
</tr>
<tr>
<td>Sample 6</td>
<td>Re-ground Sample 5</td>
<td>Gas-mixing</td>
<td>1425˚C</td>
<td>30 min.</td>
<td>Water</td>
</tr>
</tbody>
</table>

Table 2. Details of NRM demagnetization

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Mass(g)</th>
<th>Initial NRM(Am^2/kg)</th>
<th>Remaining NRM(Am^2/kg)</th>
<th>NRM_{AF_{max}}/NRM_0</th>
<th>MDF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 3</td>
<td>0.0048</td>
<td>4.931x10^{-3}</td>
<td>3.321x10^{-4} (100 mT)</td>
<td>6.7 %</td>
<td>28 mT</td>
</tr>
<tr>
<td>Sample 4</td>
<td>0.0146</td>
<td>1.110x10^{-2}</td>
<td>2.121x10^{-3} (200 mT)</td>
<td>19.1 %</td>
<td>105 mT</td>
</tr>
<tr>
<td>Sample 5</td>
<td>0.0266</td>
<td>4.605x10^{-4}</td>
<td>7.771x10^{-5} (200 mT)</td>
<td>16.9 %</td>
<td>100 mT</td>
</tr>
<tr>
<td>Sample 6</td>
<td>0.0941</td>
<td>2.250x10^{-3}</td>
<td>3.802x10^{-4} (200 mT)</td>
<td>16.9 %</td>
<td>110 mT</td>
</tr>
</tbody>
</table>
Table 3. Details of ARM demagnetization

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Mass(g)</th>
<th>Initial ARM(Am²/kg)</th>
<th>Remaining ARM(Am²/kg)</th>
<th>ARM_AFmax/ARM₀</th>
<th>MDF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 3</td>
<td>0.0048</td>
<td>4.408x10⁻³</td>
<td>1.208x10⁻³ (200 mT)</td>
<td>27.4 %</td>
<td>60 mT</td>
</tr>
<tr>
<td>Sample 4</td>
<td>0.0146</td>
<td>4.074x10⁻³</td>
<td>2.94x10⁻⁴ (200 mT)</td>
<td>7.2 %</td>
<td>125 mT</td>
</tr>
<tr>
<td>Sample 5</td>
<td>0.0266</td>
<td>4.624x10⁻⁴</td>
<td>6.951x10⁻⁵ (200 mT)</td>
<td>15.0 %</td>
<td>100 mT</td>
</tr>
<tr>
<td>Sample 6</td>
<td>0.0941</td>
<td>2.511x10⁻³</td>
<td>2.927x10⁻⁴ (200 mT)</td>
<td>11.7 %</td>
<td>110 mT</td>
</tr>
</tbody>
</table>

Table 4. Details of IRM demagnetization

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Mass(g)</th>
<th>Initial IRM(Am²/kg)</th>
<th>Remaining IRM(Am²/kg)</th>
<th>IRM_AFmax/IRM₀</th>
<th>MDF</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample 3</td>
<td>0.0048</td>
<td>2.318x10⁻¹</td>
<td>1.438x10⁻¹(200 mT)</td>
<td>0.6 %</td>
<td>28 mT</td>
</tr>
<tr>
<td>Sample 4</td>
<td>0.0146</td>
<td>1.673x10⁻¹</td>
<td>5.267x10⁻¹(200 mT)</td>
<td>3.2 %</td>
<td>75 mT</td>
</tr>
<tr>
<td>Sample 5</td>
<td>0.0266</td>
<td>4.222x10⁻²</td>
<td>4.741x10⁻²(200 mT)</td>
<td>11.2 %</td>
<td>65 mT</td>
</tr>
<tr>
<td>Sample 6</td>
<td>0.0941</td>
<td>1.266x10⁻¹</td>
<td>2.861x10⁻¹(200 mT)</td>
<td>22.6 %</td>
<td>85 mT</td>
</tr>
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</table>
Table 5. Equations of Isoactivity Curves of Olivine and Orthopyroxene Corresponding to Heterogeneous Oxidation-Reduction Equilibriums in the System MgO-Fe-SiO$_2$-O (Nitsan, 1974).

<table>
<thead>
<tr>
<th>3-Phase Field</th>
<th>Heterogeneous Reactions</th>
<th>Equation of Isoactivity Curve*</th>
</tr>
</thead>
<tbody>
<tr>
<td>OSM</td>
<td>$6\text{FeSi}_2\text{O}_4 + O_2 = 2\text{Fe}_3\text{O}_4 + 3\text{SiO}_2$</td>
<td>$\log f_{O_2} = -25738/T + 9.0 - 6 \log a_{Fe} + 2 \log a_{Mg}$ (1)</td>
</tr>
<tr>
<td>OPM</td>
<td>$6\text{FeSi}_2\text{O}_4 + 3O_2 = 2\text{Fe}_3\text{O}_4 + 3\text{FeSiO}_3$</td>
<td>$\log f_{O_2} = -22822/T + 7.08 + 6 \log a_{Fe} - 12 \log a_{Fe} + 2 \log a_{Mg}$ (2)</td>
</tr>
<tr>
<td>OSI</td>
<td>$\text{FeSi}_2\text{O}_4 = \text{Fe} + 2\text{SiO}_2 + \frac{1}{2}O_2$</td>
<td>$\log f_{O_2} = -26524/T + 5.54 + 2 \log a_{Fe}$ (3)</td>
</tr>
<tr>
<td>OPI</td>
<td>$2\text{FeSi}_2\text{O}_4 = \text{Fe} + 2\text{FeSiO}_3 + \frac{1}{2}O_2$</td>
<td>$\log f_{O_2} = -27496/T + 6.18 + 4 \log a_{Fe} - 2 \log a_{Fe}$ (4)</td>
</tr>
<tr>
<td>PSM</td>
<td>$3\text{FeSiO}_3 + \frac{1}{2}O_2 = 2\text{Fe}_3\text{O}_4 + 3\text{SiO}_2$</td>
<td>$\log f_{O_2} = -28654/T + 10.92 + 2 \log a_{Mg} - 6 \log a_{Fe}$ (5)</td>
</tr>
<tr>
<td>PSI</td>
<td>$\text{FeSiO}_3 = \text{Fe} + \text{SiO}_2 + \frac{1}{2}O_2$</td>
<td>$\log f_{O_2} = -25552/T + 4.90 + 2 \log a_{Fe}$ (6)</td>
</tr>
</tbody>
</table>

*Numbers in parentheses refer to appropriate equations.
Figure 1. (a) Dusty olivine from the Semarkona chondrite (Leroux et al. 2003). Bright spots are oriented inclusions of Fe-Ni alloy – potential carriers of primary remanent magnetization. (b) Synthetic dusty olivine produced in this study (Sample 6) by reduction of Fe-bearing olivine. Please note the difference in scale between the two images.
Figure 2. Backscattered electron (BSE) images of textures produced during olivine reduction, (a) sample 3, (b) sample 4 and (c) sample 6. Metallic phases appear as white, submicron inclusions within the olivine grains and as coarse (5 to 15 μm) crystals located at olivine grain boundaries. Scale bars are 10 μm for (a), 20 μm for (b) and 25 μm for (c).
Figure 3. Comparison between normalized ARM(blue), IRM(red) and NRM(green) demagnetization curves for sample 3 (a) sample 4 (b), sample 5 (c) and sample 6 (d).
Figure 4. Orthogonal vector diagrams of Alternating Field Demagnetization for sample 3 (upper left), sample 4 (upper right), sample 5 (lower left) and sample 6 (lower right) and equal area direction plots plotted by PaleoMag3.1 (Jones, 2002). Numbers correspond to demagnetization field intensities.
Figure 5. Normalized acquisition curves (blue) and AF demagnetization curves (red) of IRM for (a) sample 3 (b) sample 4 (c) sample 5 and (d) sample 6.
Figure 6.
Figure 6. Low-temperature AC susceptibility measurement in starting material and thermal decay of a field-cooled (FC) (solid) and zero-field-cooled (ZFC) (dashed) 2.5T LT-SIRM during warming to 300K. The first derivative of this remanence decay is shown in order to emphasize the presence or absence of the Verwey transition. (A) unreduced olivine, (B) sample 3, (C) sample 4, (D) sample 5, (E) sample 6, and (F) Fe-metal thin film.
Figure 7. FORC diagrams along with marginal coercivity distributions calculated by FORCinel (Harrison & Feinberg et al., 2008) (A) unreduced olivine (SF=12), (B) sample 3 (SF=5), (C) sample 4 (SF=6), (D) sample 5 (SF=10), and (E) sample 6 (SF=10). Please note the difference in both horizontal and vertical scales between samples. For comparison, FORC diagram of Allende chondrule (Acton et al. 2007) are included as well.
(a) Saturation of magnetization ($M_s$) for sample 3 and sample 4 obtained from hysteresis loops measurements from room temperature (30°C) to 770°C in an increment of 10°C where solid lines denote measurements and dash lines denote the first derivative, respectively.
(b) Induced magnetization measured in a 1.5T applied field during thermal cycling where solid and dash lines denote measurement and the 1st derivative, respectively. Both sample 5 and 6 were heated from 30°C to 770°C in an increment of 1°C during measurements, and subsequently cooled back to the onset temperature.
Figure 9. A three-run susceptibility as a function of temperature ($\chi$ vs $T$) measurement of subsample taken from exterior of sample 5 from room temperature (30°C) to 800°C and sample 6 from room temperature (20°C) to 700°C where solid lines denote heating and dash lines denote cooling, respectively. The first, second and the third run are labeled as black, red and blue curves.
Figure 10. The stability fields of olivines of various fayalite contents with respect to oxidation and reduction at 1 atm total pressure. Olivine of a given fayalite content is stable under $fO_2$-T conditions corresponding to the area enclosed by the light curve representing its composition (in mole percent fayalite). The univariant magnetite-wüsite (MW) and wüsite-iron (WI) equilibriums are included for comparison. The subsolidus heterogeneous reactions are listed in Table 5. (Nitsan, 1974).
Figure 11. A phase diagram for the join Fe-O at low pressures. Note breaks in the compositional scale and also the expanded scale near Fe. (Donald H. Lindsley, 1991)
Figure 12. Equilibrium Fe-Ni phase diagram displaying phase equilibrium for various Ni content in Fe versus temperature. (Cacciamani et al. 2006)
Figure 13. Schematic diagrams of heating and cooling curves for sample 5 and sample 6 during thermomagnetic measurements.
**Figure 14.** Initial cooling curves characterized by temperature as a function of duration for sample 3 and sample 4 during sample synthesis.
7. References


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