

Formation ages and places of hydrated chondrite parent bodies

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When and where hydrated chondrite parent bodies accreted is a key question to understand the origin of chondritic water and the dynamical processes of the solar system evolution.

The ages of aqueous alteration can be inferred from ^{53}Mn - ^{53}Cr systematics of aqueously-formed secondary minerals (carbonates in CM, CI, and CR chondrites and fayalite in CV, CO, and ordinary chondrites). The ^{53}Mn - ^{53}Cr ages of carbonates and fayalite in carbonaceous chondrites (CCs) are similar, 3.5 – 5 Myr after the birth of the solar system (represented by CAIs in CVs) (Fujiya et al., 2013; Doyle et al., submitted). Fayalite in ordinary chondrites (OCs) seems to have formed slightly earlier (~1 Myr) than that in CCs. These observations are consistent with ^{26}Al - ^{26}Mg ages of chondrules (e.g., Kita and Ushikubo, 2012). The ^{53}Mn - ^{53}Cr ages of aqueous alteration indicate that water activity on CC and OC parent bodies started almost contemporaneously, and that the dominant heat source for aqueous alteration is the decay energy of ^{26}Al (half-life: 0.7 Myr). Based on these ages, numerical simulations of the thermal history of CC and OC parent bodies suggest that they accreted 2.5 – 4 Myr after CAIs (Sugiura and Fujiya, 2014).

The formation places of chondrite parent bodies in the protoplanetary disk are more difficult to be inferred. The estimated water (ice) to rock mass ratios of CCs and OCs (<0.6) (e.g., Clayton and Mayeda, 1999), significantly lower than the solar value (1.2) (Lodders, 2003), suggest that CC and OC parent bodies accreted near from the snow line. The inferred D/H ratios and O isotopic compositions of water in CCs are likely to be significantly different from those of primordial (molecular cloud) water and/or most Oort-cloud comets measured so far, indicating that they must have recorded various degrees of isotopic re-equilibration between primordial water and nebular gas (Alexander et al., 2012; Krot et al., 2013). Model predictions on the location of the snow line 2.5 – 4 Myr after CAIs (e.g., Ciesla and Cuzzi; 2006) and both the temporal and the spatial distribution of H and O isotopic ratios of water in the protoplanetary disk (e.g., Yang et al., 2011, 2013) suggest that hydrated chondrite parent bodies accreted in the main asteroid belt. I am currently trying to constrain the contribution of C from cometary inorganic ice to the C inventory in CCs. Carbon isotopic ratios of carbonates in Murchison (CM) suggest that C reservoirs in primitive aqueous fluids were highly enriched in ^{13}C with $\delta^{13}\text{C} > 70$ ‰, which provides no evidence for C contribution from cometary ice (Fujiya et al., submitted).

Keywords: Hydrated chondrites, Formation ages, Formation places, Compositions of stable isotopes

Aqueously altered clasts in the NWA1232 CO3 carbonaceous chondrite

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CO parent bodies are generally believed to have escaped significant degree of brecciation. However, Northwest Africa 1232 (NWA1232) CO3 chondrite is a breccia consisting of three distinct lithologies (A, B, and C) that have experienced different degrees of thermal metamorphism [1,2]. Recently, we found that lithology A, which is the host lithology of NWA1232, contains numerous small clasts (100-1800 μm in diameter) of different metamorphic grades [3,4]. Most of the clasts show little evidence of thermal metamorphism and contain significant amounts of hydrous minerals. These characteristics have not been known in other CO3 chondrites and potentially provide new insights into the formation of CO3 chondrites. Here, we report the results of detailed mineralogical and petrographic study of the hydrous clasts in NWA 1232. We used SEM-EDS, TEM (STEM)-EDS, EPMA-WDS, and SR-XRD.

Small clasts occur abundantly throughout lithology A. Among them, hydrous clasts are most abundant and comprise ~2 vol.% of lithology A. Each of the clasts typically consists of one chondrule surrounded by a fine grained matrix, exhibiting the appearance of a chondrule with a rim. Some clasts contain multiple chondrules and CAIs embedded in a matrix.

Most chondrules in the clasts are Mg-rich Type I, and their olivine phenocrysts have homogeneous low Fe compositions (Fa_1) and show almost no Fe-Mg zoning; these are similar to those in CO3.0 chondrites. Enstatite phenocrysts and mesostasis were partially replaced by fine grains (10-20 nm) of phyllosilicates and an Fe-Si-Mg-rich amorphous material. The amorphous material contains small amounts of fine olivine grains (<100 nm). High-resolution TEM observations reveal that the most abundant phyllosilicate exhibits ~0.7 nm basal spacing; thus, it is serpentine. Minor amounts of smectite with 1.0-1.1 nm basal spacing, were also observed. These phyllosilicates are compositionally indistinguishable from the amorphous material.

The rim-like matrix surrounding the chondrules mainly consists of an Fe-Si-Mg rich amorphous material, and contains relatively coarse grains (1-2 μm) of magnetite, forsteritic olivine, enstatite, fine grains (100-500 nm) of olivine, troilite, and finer grains (10-20 nm) of serpentine. The amorphous material is compositionally indistinguishable from the serpentine and mineralogically and compositionally similar to that within the chondrules.

Our results suggest that the clasts have experienced weak aqueous alteration in the meteorite parent body. However, the evidence of aqueous alteration was not observed in the outside of the clasts. These suggest that the clasts were formed and transported from a region in the parent body that was different from the region where the meteorite was finally lithified, and subsequently they were incorporated into lithology A. We note that the mineralogical and compositional characteristics of chondrules and matrix in the clasts are similar to those in the primitive CO3.0 chondrite ALHA77307 [5].

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Keywords: CO chondrite, clast, brecciation, aqueous alteration, TEM, SR-XRD

Hydrothermal alteration experiments of MgO-SiO₂ amorphous silicates

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Hydrothermal alteration is one of the important processes for solid evolution in the early solar system. So far, many experiments have focused on reaction between aqueous fluids and meteorite itself or crystalline silicates, such as olivine and pyroxene (e.g., [1, 2]). Reaction with amorphous silicates is also important because interstellar dust as original building blocks of planetesimals is mostly amorphous [3] and primitive amorphous silicates are present in cometary dust as GEMS (e.g., [4]) and some carbonaceous chondrites (e.g., [5]). Hydrothermal alteration experiments with amorphous silicates were also carried out using natural GEMS [6] and synthetic materials with CI composition [7]. However, any experiments using amorphous silicates in a simple system have not been made. In this study, hydrothermal alteration experiments of amorphous silicates in the system MgO-SiO₂-H₂O were carried out to understand basic features of hydrous layer silicate formation.

Nanoparticles of amorphous silicates ~10 nm in size with nearly enstatite and forsterite compositions (E and F: Mg/Si = 1.15 and 2.02) were synthesized by the induction thermal plasma method as starting materials. Intermediate compositions were also prepared by mixing E and F (ME, M and MF: Mg/Si=1.25, 1.5 and 1.75). Three types of experiments were made. (1) Synthetic experiments: Starting materials and pure water (water/rock ratio: W/R=5.0) were sealed and heated at 50, 100, 150 and 200°C or held at room temperature for 0 to 1344 hrs. The run products were dried in vacuum and examined by powder X-ray diffraction (XRD) and SEM/EDX. (2) In-site experiments: Starting materials mixed with pure water (W/R=5.0) were exposed in water-saturated atmosphere at 60°C and repeatedly measured by in-situ powder XRD (total duration: 9-292.5 hrs.). The final run products were dried in vacuum and measured again by powder XRD. (3) Swelling experiments: some dried run products were immersed in ethylene glycol, glycerol or water and measured by powder XRD to check swelling.

Layer silicates formed in all the run products even at room temperature. Brucite and sometimes magnesite were formed in runs using F. XRD reflection peaks of the layer silicates are weak and broad particularly for the (001) peak suggesting that they are thin and have low crystallinity. The (001) lattice spacing and the chemical compositions of the run products together with the results of the swelling experiments indicate that the layer silicates are disordered mixed layer minerals of stevensite (Mg-smectite) or vermiculite, serpentine and talc. The (001) peak becomes sharper to some extent and shift to higher 2θ (smaller lattice spacing) with increasing run duration or temperature except for runs using E. The crystallinity and degree of mixing order also changed by drying process. The experimental results can be explained by that aqueous solutions of high degree of supersaturation with respect to layer silicate minerals formed by instantaneous dissolution of metastable amorphous silicate nanoparticles with high reactivity, and disordered mixed layer minerals metastably formed from these solutions. The presence of disordered mixed layer minerals in some carbonaceous chondrites, such as in CI [8], indicates that they formed from amorphous silicates. It should also be noted that we could not discuss hydrothermal conditions based on the present features of layer silicate minerals in meteorites, as they could be different from those during hydrothermal alteration.

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Keywords: hydrous layer silicate, hydrous alteration, carbonaceous chondrites, induction thermal plasma method, disordered mixed layer minerals

Hydrothermal and heating experiments: Implications for formation process of nepheline in carbonaceous chondrite

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Nepheline (NaAlSiO_4) and sodalite ($\text{Na}_4\text{Al}_3\text{Si}_3\text{O}_{12}\text{Cl}$) are widely distributed in chondrules and matrices of CO/CV chondrites (Kimura et al. 2014; Matsumoto et al. 2014). Because of the low condensation temperatures of such Na-rich minerals, these are likely not primary phases condensed from the solar nebula, but are secondary phases altered from melilite or plagioclase. According to previous petrographic studies (e.g. Russell et al., 1998; Tomeoka and Itoh, 2004), nepheline and sodalite are considered to form by hydrothermal processes on their parent body. Because of the high volatility and high mobility, Na element is potentially a powerful indicator reflecting aqueous environments of the chondrite parent bodies. In this study, we performed hydrothermal alteration experiments to understand the formation process of Na-rich minerals in chondrite parent bodies.

We used i) synthetic melilite (gehlenite, $\text{Ca}_2\text{Al}_2\text{SiO}_7$) + SiO_2 , and ii) natural plagioclase (An_{48}) as starting materials. Four reaction solutions of 1N (mol/l)-NaOH (pH 0), H_2O (pH 7), 0.1N-NaOH (pH 13) and 1N-NaOH (pH 14) were reacted with three different water/rock ratios (10, 100 and 1000). Na^+ concentration in all solutions was maintained at 1 mol/l by addition of NaCl. Hydrothermal alteration experiments were performed with a PTFE reaction vessel loaded into steel autoclave and were carried out at temperature at 200 °C and pressure of 1.5 MPa for run duration time of 168 hours. The recovered samples were analyzed by XRD, SEM, TEM, and TG-DTA.

In the experiments using melilite+ SiO_2 with W/R = 100 and pH 7-14 solutions, hydrogrossular ($\text{Ca}_3\text{Al}_2[\text{SiO}_4]_{1.6}[\text{O}_4\text{H}_4]_{1.3}$) formed as a secondary phase with low SiO_2 contents, while analcime ($\text{NaAlSi}_2\text{O}_6\text{H}_2\text{O}$), nepheline-hydrate ($\text{NaAlSiO}_4\text{H}_2\text{O}$), basic-cancrinite ($\text{Na}_8\text{Al}_6\text{Si}_6\text{O}_{24}[\text{OH}]_2 \bullet 2\text{H}_2\text{O}$) and tobermorite ($\text{Ca}_5\text{Si}_6\text{O}_{16}[\text{OH}]_2 \bullet 4\text{H}_2\text{O}$) formed with high SiO_2 contents. Although no secondary phase was observed at high W/R (=1000), analcime and basic-cancrinite occurred at low W/R (=10). Under the pH 0 solution, melilite was dissolved and no secondary phases formed. In the experiments using plagioclase with W/R = 100 and pH 0-7 solutions, no secondary phases formed. On the other hand, under pH 13-14 solutions, analcime and nepheline-hydrate formed with W/R = 10 and 100.

A TG-DTA of nepheline-hydrate at heating rate 5 °C/min in air showed that the dehydration started at ~100 °C and finished at ~800 °C. From the DTA and XRD data of this condition, nepheline-hydrate is considered to be transformed into nepheline at ~800 °C. From the TG-DTAs of nepheline-hydrate with different heating rates, we estimated that nepheline-hydrate could alter to nepheline for at least ~ 10^3 years at 400 °C. A TG-DTA of analcime at heating rate 2 °C/min in air showed that dehydration started at ~120 °C and finished at ~500 °C. The XRD data showed analcime is transformed into nepheline and amorphous material below 1000 °C, although we failed to estimate the required time of the transformation because of the DTA curves of analcime were too complicated.

The present results show melilite and plagioclase are easily altered into hydrous minerals like nepheline-hydrate and analcime in Na^+ -rich high alkali solutions. Because the experimental condition demonstrated in the present study seems to be comparable to actual carbonaceous chondrite parent body, we infer that nepheline in carbonaceous chondrite formed initially as intermediate products like nepheline-hydrate and analcime during aqueous alteration, and these hydrous minerals were subsequently dehydrated to nepheline.

Keywords: nepheline, melilite, aqueous alteration, hydrothermal experiments, dehydration heating experiments, carbonaceous chondrite

Microtextures in the MAC 88107 carbonaceous chondrite: Is this meteorite really a primary accretionary rock?

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MAC 88107 is an ungrouped carbonaceous chondrite having intermediate properties between the CM and CO groups [1]. Carbonaceous chondrites generally consist of chondrules and inclusions embedded in a fine-grained matrix, which constitutes >30 vol.% of the meteorites. In contrast, MAC 88107 virtually has no matrix and consists mainly of chondrules and inclusions surrounded by rims. The rims are thought to have formed by accretion of dust onto the surface of chondrules and inclusions while floating in the solar nebula [2]. Thus, meteorites like MAC 88107 are thought to be “primary accretionary rocks”, which formed directly from the solar nebular [2]. However, recent studies of chondrules/rims in the Mokoia CV chondrite show abundant evidence for rim formation within the meteorite parent body [3,4]. Based on the model proposed by [3, 4], the rims are remnants of former matrix adhering to chondrules and inclusions.

To reevaluate the formation processes of MAC 88107, we performed detailed observation and analyses of this meteorite using SEM-EDS, STEM-EDS, and SR-XRD.

The rims in MAC 88107 mainly consist of extremely fine-grained material (<1 μ m) with small fragments (1-10 μ m) of forsterite, enstatite, and magnetite-fayalite-hedenbergite aggregates. Our STEM observations and SR-XRD measurements reveal that the extremely fine-grained material consists mostly of an amorphous material with minor amounts of phyllosilicate, olivine, pyroxene, pentlandite, and magnetite.

Many rims in MAC 88107 contain characteristic veins (~10 μ m in width) consisting of coarse grains (1-15 μ m) of magnetite, fayalite, and hedenbergite. Krot et al. (2000) [1] suggested that these veins were formed during fluid-assisted oxidation of metal-sulfide nodules in chondrule peripheries. The oxidation was associated with a volume increase, induced high stresses in adjacent rims, and eventually broke to form fractures, which were subsequently filled with growing magnetite. However this model cannot explain the presence of veins in rims around CAIs which have no opaque nodules. If the veins were originated from cracks formed by brecciation of the present meteorite lithology, there should have been veins penetrating multiple chondrules/rims. However such vein is absent. These facts suggest that the veins were formed from cracks that resulted from brecciation of a precursor lithology of the chondrules/rims.

Characteristic layers, 5-30 μ m in width, occur between neighboring rims. These layers consist of relatively coarse-grained magnetite, fayalite, and hedenbergite. Krot et al. (2000) [1] termed them “inter-rim layers”. The mineralogy and texture of inter-rim layers resemble to those of veins in the rims.

We found several clasts which contain multiple chondrules in MAC 88107. These clasts are 150-500 μ m in diameter and have round shapes. They contact with adjacent chondrules or rims directly or through inter-rim layers. The chondrules in the clasts have no rims. The matrices of the clasts have mineralogy and texture similar to the chondrule rims in the outside of the clasts. These observations suggest that the chondrules/rims are actually clasts that resulted from brecciation of a precursor lithology and subsequent transportation and abrasion.

From these results, we suggest that MAC 88107 was formed through the following processes: (1) formation of chondrule/rim clasts by brecciation of a precursor lithology, (2) transportation and abrasion, and (3) accumulation and lithification of those clasts.

References:

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Keywords: MAC 88107, Primary accretionary rock, Carbonaceous chondrite, Brecciation, Aqueous alteration, Chondrule rims

Estimation of the equilibrium form of olivine from negative crystals in an equilibrated chondrite

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Olivine is one of the most common minerals both in the solar system and the interstellar and circumstellar environments. Adsorption of hydrogen atoms and water molecules onto the olivine surfaces may play a significant role in the formation of hydrogen molecules in the interstellar medium and the origin of water in the earth [2,3]. The crystal orientations of the olivine surfaces may affect adsorption efficiency and reaction rates, and thus it is important to understand crystal forms of olivine. Although the equilibrium form of olivine has been investigated based on the surface energies of olivine obtained by Ab initio calculations [1], it has not been confirmed by observations of naturally occurred olivine.

Many voids with facets (negative crystals) several μm in size have been identified within olivine grains in HAYABUSA samples and equilibrated chondrites [4]. These negative crystals appear to be the equilibrium form because equilibrated chondrites have experienced thermal metamorphism at $\sim 800^\circ\text{C}$. In this study, we analyze the three-dimensional shapes of negative crystals in an olivine grain to obtain the equilibrium form of olivine using micro-sampling with focused ion beam (FIB) and micro-tomography [5]. The results are compared with the estimated equilibrium form by the Ab initio calculations.

A thin section of Tuxtuac meteorite (LL5) was used in this study. A negative-crystal-rich region of an olivine grain was identified by an optical microscope. The $30 \times 30 \times 60 \mu\text{m}^3$ sized micro-sample was prepared by an FIB technique (FEI Quanta 200 3DS). X-ray microtomography was performed at the SPring-8 BL47XU beamline at (7 keV, 70.5 nm/voxel). Negative crystals were extracted three-dimensionally by binarization of the CT images. The orientation of the host olivine crystal was determined with an FE-SEM/EBSD (JEOL 7001F/HKL CHANNEL5) and face angles of the negative crystals were measured using the 3D-CT images. Crystal plane indices of negative crystals were determined from the obtained crystal orientation and face angles.

We identified seven negative crystals 2-4 μm in size. They aligned in the same plane within the olivine crystal suggesting that they are a healed crack as well as those in the HAYABUSA samples [5]. All negative crystals have developed facets of the same crystal orientations and have similar morphology, and some edges were rounded, which indicate that the morphology of the negative crystals almost represents the equilibrium form of olivine. The (100), (001), and (011) facets were observed and (100) facet was most well-developed. The other facets seem to be (001), (102), and (343) planes although these facets were small with respect to the spatial resolution of the micro-tomography.

Forsterite surface energies determined by Ab initio calculation increase in order of (010), (120), (001), (101), (111), (021), and (110) [1]. The observed (010), (120), and (001) planes have low surface energies while the surface energy of the (100) plane is high [6] and those of (011), (102), and (343) are probably high as well. The possible interpretations of unexpected facet planes with high energies are (1) changing surface energy due to adsorption of molecules onto the crystal surface, (2) temperature dependence of the surface free energy, and (3) kinetic effect. If surface energies change due to adsorption of molecules, anisotropy of the surface structures would play an important role for surface stability. Evaporation rate of forsterite along the a-axis in hydrogen gas at less than 1400°C is smaller than those along the b- and c-axes [7], which is consistent with this study.

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Keywords: equilibrium form, negative crystal, equilibrated chondrite, olivine, Ab initio, surface energy

An experimental study on the formation of the type I chondrules

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Introduction: Chondrules have been reproduced at 1 bar under the IW-0.5 oxygen pressure, where FeO-rich, type II chondrules are produced. Although type I chondrules have been experimentally examined by Cohen and Hewins (2004), the charge was quenched and dissimilar to natural chondrules. In the present study, chondrules were reproduced under the condition of the IW-3 to -5 with the various cooling rates mainly of 100 oC/h, where FeO-poor, type I chondrules are produced.

Experiments: Three kinds of sintered pellets of homogenized powders with chondritic compositions were prepared as starting materials of the present experiments; (1) Allende CV3.2, (2) NWA 1465 (a carbonaceous chondrite), and (3) powder mixtures including iron, olivine, enstatite, anorthite, and diopside. The peak temperatures of the runs 1200-1550 oC and the cooling rates 80-10000 oC/h. Two cases were considered in the use of silica powder as Si-rich gas source and in the absent of the silica powder. The furnace was low pressure type controlling below the atmospheric pressure under the reduced condition, whose pressure was controlled to be mainly 100 Pa. The capsule made of alumina has an orifice (1 mm) on the top and the charge is held using the Mo-wire of 0.2 mm in diameter in the capsule. Silica was put on the bottom of the capsule for its use. The vapor pressure from the charge and silica in the capsule is about 1 Pa at ~1450 oC of the peak temperature. When the furnace total pressure is controlled to be 100 Pa using the hydrogen gas, the hydrogen gas enters into the capsule balancing the outside of the capsule. The oxygen pressure is IW-3 in the case of the existence of silica powder, and IW-4 in the absence of the silica powder at 1450 oC. The starting materials (1) and (2) correspond to the metallic iron poor starting materials and the runs of 53 times were carried out (23 times with Si-rich gas and 30 times without Si-rich gas). The starting material (3) corresponds to the metallic iron rich starting materials and the runs of 21 times were carried out (11 times with Si-rich gas and 10 times without Si-rich gas). The polished sections were made from the run produces, and observed and analyzed using an electron probe micro analyzer (EPMA, JXA-8200).

Results and discussion: Poikilitically enclosed rounded forsteritic olivines in porphyritic enstatites formed from metallic iron poor starting materials at the peak temperature of 1450 oC and the cooling rate of 100 oC/h, which is similar to type I chondrules. The rounded olivines are relict phase, dusty olivines and forsterites, common in chondrules. The texture of the runs with Si-rich gas source is similar to the type I chondrules. Although kamacite droplets enclosed in olivines and pyroxenes are usually abundant in type I chondrules, they are not common in the run products. The iron content of the run products decreased. Since the volume of the absorption of iron into the Mo wire is much less than the volume of compositional change, the significant of the iron oxide component directly evaporated but not the reduction into metallic iron.

Metallic iron melt tends to aggregate into a clump at the peak temperature more than 1500 oC from metallic iron rich starting materials. The metallic iron was dispersed in the charge due to the low melting degree at the peak temperature less than 1500 oC, but the iron was deficient on the charge surface, suggesting the evaporation of iron. The textures are not similar to chondrules.

Considering the present experimental results, we may prefer the metal-deficient precursors for the type I chondrules rather than metal-bearing precursors. However, the origin of kamacite droplets enclosed in Mg-rich olivine and pyroxenes in type I chondrules is still unclear. They may be originated from the injection during the chondrule melting or the iron vapor was saturated during the chondrule melting.

Reference: Cohen B. A. and Hewins R. H. 2004. GCA, 68, 1677-1689.

Keywords: chondrule, type I, chondrule formation, reproduction, experimental study

Cosmic-ray exposure age and heliocentric distance of the parent body of the rumuruti chondrite PRE 95410

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We measured concentrations and isotopic ratios of noble gases in the rumuruti (R) chondrite Mt. Prestrud (PRE) 95410. This meteorite contains high concentrations of solar and cosmogenic noble gases, from which solar gas implantation rate (concentrations of solar noble gases implanted per unit time) can be estimated. By comparing the solar gas implantation rates between PRE 95410 and lunar regolith samples, the parent-body heliocentric distance of the meteorite can be obtained, as solar wind flux is inversely related to the square of heliocentric distance. Based on the exposure model of solar noble gases and galactic cosmic rays, we calculated the exposure age on its parent body (15.4 ± 5.2 Ma), exposure age in space after ejection from the parent body (9.5 ± 1.3 Ma), and heliocentric distance of the parent body (1.3 ± 0.2 AU). The calculated exposure age in space is consistent with the peak of space exposure age distribution of other R chondrites. The derived heliocentric distance suggests the location of parent body when constituents of the PRE 95410 meteorite were exposed to the solar wind. From the previous studies of mineralogy and chemistry, R chondrites might have formed between the regions where ordinary and carbonaceous chondrites formed (2-4 AU). Hence the heliocentric distance of the PRE 95410 parent body studied in this work is not consistent with the formation region. This may imply that the parent body of the PRE 95410 migrated from the R chondrite formation region to the inner area where irradiated by solar wind before the ejection of the meteorite (9.5 ± 1.3 Ma). Kr isotopic ratios show excesses of ⁸⁰Kr and ⁸²Kr produced by neutron capture reaction on Br during space exposure. The minimum radius of the PRE 95410 meteoroid was calculated as 53 cm from the abundances of neutron-induced Kr.

Keywords: Rumuruti chondrite, noble gas analysis, cosmic-ray exposure age, heliocentric distance

Nucleosynthetic Strontium Isotope Anomalies in Carbonaceous Chondrites

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We present precise Sr isotopic compositions in samples from sequential acid leaching experiments for three carbonaceous chondrites, Allende, Murchison, and Tagish Lake, together with those in the bulk aliquots of these meteorites. The chondritic acid leachates and residues were characterized by Sr isotope anomalies with variable $\mu^{84}\text{Sr}$ values (10^6 relative deviation from a standard material) ranging from +120 to -4700 ppm, documenting multiple nucleosynthetic sources within a single meteorite. In addition, the $\mu^{84}\text{Sr}$ patterns across leaching samples for individual chondrites differed from one another. The highest $\mu^{84}\text{Sr}$ values were observed for leaching Step 3 (HCl+H₂O, 75 °C) for Allende and Murchison likely because of the incorporation of calcium and aluminum-rich inclusions (CAIs). In contrast, extremely low $\mu^{84}\text{Sr}$ values were observed in the later fractions (Steps 6 and 7) for Murchison and Tagish Lake, suggesting the existence of s-process-enriched presolar SiC grains derived from AGB stars.

A $\mu^{84}\text{Sr}$ - $\epsilon^{54}\text{Cr}$ diagram was prepared with the CAIs and bulk aliquots of carbonaceous chondrites and other meteorites (non-carbonaceous) that were plotted separately; however, they still formed a global positive correlation. CAIs presented the highest $\mu^{84}\text{Sr}$ and $\epsilon^{54}\text{Cr}$ values, whereas carbonaceous chondrites and noncarbonaceous meteorites had intermediate and the lowest $\mu^{84}\text{Sr}$ and $\epsilon^{54}\text{Cr}$ values, respectively. The positive trend was interpreted as resulting from global thermal processing in which sublimation of high $\mu^{84}\text{Sr}$ and $\epsilon^{54}\text{Cr}$ carriers generated the excess $\mu^{84}\text{Sr}$ and $\epsilon^{54}\text{Cr}$ signatures in CAIs, while noncarbonaceous planetesimals accreted from materials that underwent significant thermal processing and thus had relatively low $\mu^{84}\text{Sr}$ and $\epsilon^{54}\text{Cr}$ values. Apart from the global trend, the carbonaceous chondrites and noncarbonaceous meteorites both exhibited intrinsic variations that highlight an isotopic dichotomy similar to that observed in other isotope combinations (e.g., $\epsilon^{54}\text{Cr}$ - $\epsilon^{50}\text{Ti}$, $\epsilon^{54}\text{Cr}$ - $\Delta^{17}\text{O}$). A plausible scenario for creation of the intrinsic variations involves local thermal processing (e.g., flash heating for chondrule formation) caused by additional selective destruction of presolar grains different than that caused by global thermal processing. The existence of such a global positive trend and local variations for two meteorite groups suggests a complicated dynamic history for the dust grains with respect to thermal processing, material transportation, and mixing in the protoplanetary disk prior to planetesimal formation.

Keywords: Sr isotopes, presolar grains, acid leaching, chondrites, nucleosynthesis, thermal processing

High precision neodymium isotopic analysis of chondrites with complete sample digestion

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A variety of isotope anomalies have been discovered in bulk chondrites and differentiated meteorites (e.g., Cr, Mo [1, 2]). These results point to the existence of planetary-scale isotope heterogeneities for refractory heavy elements, which are most likely due to the heterogeneous distribution of presolar grains (e.g., SiC, graphite) in the protosolar nebula before the onset of planetesimal formation.

High precision Nd isotope analyses in meteorites have been the center of interest in recent cosmochemistry community. One of the most remarkable results is that chondrites possess $^{142}\text{Nd}/^{144}\text{Nd}$ ratios ~ 20 ppm lower than those in terrestrial rocks [3]. The anomaly was interpreted to be caused by the Sm-Nd fractionation via early differentiation of the terrestrial mantle. On the other hand, variations in stable Nd isotopes (e.g., $^{148,150}\text{Nd}/^{144}\text{Nd}$) have been documented in chondrites [4]. Although the authors concluded that the observed variation was due to incomplete digestion of presolar grain-bearing samples, the existence of Nd isotope anomalies in bulk aliquots of chondrites remains unclear unless high precision Nd isotope data with complete sample digestion become available.

In this study, we revisit high precision Nd isotope analysis of chondrites coupled with a new sample digestion technique that confirms complete dissolution of acid resistant presolar grains. We also develop a modified dynamic multicollection method using TIMS to improve the analytical reproducibilities.

We investigated two carbonaceous chondrites (Murchison, CM2; Allende, CV3), five ordinary chondrites (Kesen, H4; Chergach, H5; Saratov, L4; Hamlet, LL4; St. Severin, LL6). The ordinary and Rumuruti chondrites with a petrologic grade greater than 3 were dissolved by a conventional acid digestion method using $\text{HNO}_3 + \text{HF} + \text{HClO}_4$ [5]. For carbonaceous chondrites, each sample was digested using a high-pressure digestion system (DAB-2, Berghof) with $\text{HF} + \text{HNO}_3 + \text{H}_2\text{SO}_4$ to completely dissolve acid resistant presolar grains [6].

The Nd isotope compositions were measured by TIMS (Triton-plus, Tokyo Tech). In previous studies, Nd isotope compositions of bulk meteorites have been commonly measured in the “static-multicollection” mode, which may be affected by the time-related deterioration of Faraday cups [7]. In contrast, the “multi-static” [8] or “dynamic-multicollection” methods can reduce the effect of cup deterioration by acquiring Nd isotopes with multiple lines of different cup configurations within a single analytical cycle. In this study, we developed a modified “dynamic-multicollection” method.

In contrast to the static mode, the dynamic method achieved improved reproducibilities as follows; $^{142}\text{Nd}/^{144}\text{Nd}$: 2.8 ppm, $^{148}\text{Nd}/^{144}\text{Nd}$: 4.5 ppm, and $^{150}\text{Nd}/^{144}\text{Nd}$: 9.2 ppm. It should be noted that improvements of reproducibilities are evident for $^{148}\text{Nd}/^{144}\text{Nd}$ and $^{150}\text{Nd}/^{144}\text{Nd}$ ratios even compared to those obtained in the multi-static method (6 ppm and 19 ppm, respectively) conducted in [8].

All samples have $\mu^{142}\text{Nd}$ values 20 – 30 ppm lower than the terrestrial value. In contrast, all but one sample (Allende) have $\mu^{148}\text{Nd}$ values indistinguishable from the terrestrial value. Likewise, $\mu^{150}\text{Nd}$ values in chondrites are generally within the range of the terrestrial component. Although the data points are limited, this study suggests that stable Nd isotopes were homogeneously distributed in the protosolar nebula, at least for carbonaceous, ordinary, and Rumuruti chondrites.

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Keywords: isotopic heterogeneity, isotopic anomaly, chondrite, neodymium, TIMS, presolar grain

Chemical composition of a precursor material of an Allende F(UN) CAI estimated from a mass dependent fractionation

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Calcium-aluminum-rich inclusions (CAIs) are considered to be the oldest solid materials in the solar system [e.g., 1, 2]. Some CAIs show mineralogy, texture, and isotopic signature that have experienced melting, evaporation and recrystallization [e.g., 3, 4]. These CAIs have lost primordial chemical compositions caused by evaporative loss of less refractory elements such as magnesium and silicon from the melt. Hence, in order to understand the origin and evolution of such 'igneous' and isotopically fractionated CAIs, estimations of primordial compositions of these CAIs are required.

Estimations of primordial compositions of 'normal' igneous CAIs have been conducted by determination of mass dependent isotopic fractionation degrees in Mg and Si [5, 6], but those of igneous CAIs with *Fractionation* and *Unknown Nuclear* effects (FUN [e.g., 7]) have been poorly carried out. FUN CAIs are amongst solar system materials with extreme mass dependent isotopic fractionations in Mg, O, and Si. The origin of FUN CAIs is still not well understood, but they must have information about an earliest stage of the solar system evolution. Here we report chemical and Si isotopic compositions of a CAI from Allende, called AL1B-F. AL1B-F is a forsterite-bearing CAI which shows large mass dependent isotopic fractionations in oxygen and magnesium [8]. These fractionation signatures indicate that AL1B-F is related to FUN CAIs.

FEG-EPMA (JEOL JXA-8530F, the Univ. of Tokyo) was used for petrologic studies. Silicon isotopic compositions have been measured by the NanoSIMS installed at Atmosphere and Ocean Research Institute, the Univ. of Tokyo.

AL1B-F is composed of two parts, a forsterite-rich core and a spinel-rich mantle. Among these two parts are filled with abundant secondary minerals (e.g., sodalite and nepheline). Silicon isotopes of forsterites and Al-Ti-rich pyroxenes in AL1B-F show a large mass dependent isotopic fractionation of up to ~22 ‰/amu.

In order to estimate the primordial composition of AL1B-F, we combined Si and previously measured Mg and O isotopic data [8]. If we assume that mass dependent isotopic fractionations in AL1B-F were resulted from simple one-stage evaporation event, ~80% of Mg and ~75% of Si must have been lost (evaporated) from the molten precursor of AL1B-F based on the experimentally determined isotopic fractionation factors [9]. Because of the presence of abundant secondary minerals, it is not possible to precisely determine the bulk chemical composition of the present AL1B-F. If we assume, however, that secondary minerals in AL1B-F are alteration products of primary melilites with Ak mole% of, e.g., ~89, which is a composition of melilites in the Vigarano forsterite-bearing FUN CAI 1623-5 [10], the estimated precursor composition for AL1B-F would be ~34 wt% MgO and ~50 wt% SiO₂. The result suggests that the precursor of AL1B-F also have a Mg- and Si-rich composition like C1, 1623-5, and CMS-1 FUN CAIs [9, 11].

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Keywords: FUN CAI, mass dependent isotopic fractionation, silicon isotopes, ion microprobe

Oxygen isotopic distribution of Type B1 CAI from the Vigarano

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Ca-Al-rich inclusions (CAIs) in meteorites are the oldest objects in the Solar System. Heterogeneous oxygen isotope distributions among inter- and intra-crystals have been observed for CAIs (e.g., Yurimoto et al., 1998). The heterogeneous oxygen isotope distributions of CAI minerals are interpreted to be the result of thermal processes in the solar nebular with different oxygen isotopic compositions, which are multiple melting, condensation, and solid-state diffusion processes, as well as aqueous and thermal metamorphism/alteration on the parent body (Yurimoto et al., 2008). However, most efficient processes to form oxygen isotopic compositions were different at each mineral and each CAI. In this study, we have conducted systematic oxygen isotope measurements for a large area in a CAI and compared with the petrography, in order to understand the processes to form the heterogeneous oxygen isotope distributions.

A type B1 CAI from the Vigarano was examined. FE-SEM-EDS system (JEOL JSM-7000F; Oxford X-Max 150) was used for petrographic observations. Oxygen isotopic compositions were measured using SIMS of Hokkaido University (Cameca ims-1280HR). Secondary ions ($^{16}\text{O}^-$, $^{17}\text{O}^-$, and $^{18}\text{O}^-$) were measured simultaneously in a multicollection mode. We selected three regions with sizes of 1 x 1 mm and measured oxygen isotopes of 40 x 40 points every 25 micrometers. Measurement time for each spot was 55 seconds including 10 seconds of ion counting time, pre-sputtering, and stage moving.

The CAI is 6.5 x 3 mm and has an irregular and a wrinkled surface. The CAI has a core-mantle structure. The core consists of spinel, melilite, anorthite, and Al-Ti-rich augite, while the mantle is composed mainly of melilite. The Wark-Lovering rim surrounds the CAI. The CAI is a fragment and an entire shape is unclear. A bulk chemical composition corresponds to those for typical Type B1 CAIs (Grossman, 1975). Crystallization sequences from a melt of the CAI composition were spinel, melilite, anorthite, and fassaite (Stolper, 1982).

The oxygen isotopic compositions of minerals in the CAI are distributed along the CCAM line in a three oxygen isotope diagram. Spinel shows a ^{16}O -rich composition ($\delta^{18}\text{O} \sim -45$ permil), while melilite shows a ^{16}O -poor composition ($\delta^{18}\text{O} \sim 11$ permil). Anorthite has bimodal distributions of oxygen isotopic compositions. Fassaite shows mainly a ^{16}O -rich composition and less spots having a ^{16}O -poor composition.

The oxygen isotopic compositions of mineral measured are not readily explained by crystallization sequences from the melt, indicating that the heterogeneous oxygen isotope distributions are the results of either multiple re-melting events or any other processes except for the melting event.

Keywords: CAI, meteorite, oxygen isotope, SIMS

Al-Mg chronology and oxygen isotope distributions of multiple melting for a Type C CAI from the Allende

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Ca-Al-rich inclusions (CAIs) in meteorites have undergone multiple melting processes in the early solar nebula, which indicates that transient heating events repeatedly occurred in the early Solar System (e.g., Yurimoto et al., 1998). A relative chronometer with Al-Mg systematics for the CAIs could be applied to determine the time interval of heating events. To determine the time interval of heating events for the CAIs, detailed histories of multiple melting events in the CAIs should be well understood. We have carried out a coordinated study of detailed petrographic observations and in-situ oxygen and Al-Mg isotope measurements for a Type C CAI, EK1-04-2, from Allende CV3. Precise petrographic observations and oxygen isotopic measurements were performed to understand the individual melting processes that occurred in the CAI. Investigation of the Al-Mg systematics was performed for minerals formed by different melting and recrystallization processes to determine the age differences of individual CAI melting events.

The CAI consists mainly of spinel, anorthite, olivine, and pyroxene, and has a core and mantle structure. Petrography of the core suggests that the crystallization sequences of the core minerals are spinel, anorthite, olivine, and pyroxene. The mantle has the same mineral assemblages as the core, and shows incomplete melting and solidification textures.

Oxygen isotopic compositions of the minerals are distributed along with a carbonaceous chondrite anhydrous mineral (CCAM) line ($\delta^{18}\text{O} = -44$ to $+9$ ‰), which indicates to preserve a chemical disequilibrium status in the CAI. Spinel shows ¹⁶O-rich signature ($\delta^{18}\text{O} \sim -43$ ‰), while anorthite shows ¹⁶O-poor signature ($\delta^{18}\text{O} \sim +8$ ‰). Olivine and pyroxene in the core have the same oxygen isotopic composition ($\delta^{18}\text{O} \sim -15$ ‰), which indicates their equilibrium. Olivine and pyroxene in the mantle have variable oxygen isotopic compositions and are slightly depleted in ¹⁶O ($\delta^{18}\text{O} = -13$ to -4 ‰) compared with the same minerals in the core. The ²⁶Al-Mg systematics is consistent with the disequilibrium status observed according to the petrography and oxygen isotopes. Spinel is plotted on a line of $(^{26}\text{Al}/^{27}\text{Al})_0 = (3.5 \pm 0.2) \times 10^{-5}$, anorthite is $(-1 \pm 5) \times 10^{-7}$, and olivine and pyroxene in the core are $(-1 \pm 7) \times 10^{-6}$. Plots of olivine and pyroxene in the mantle are scattered below the isochron of these minerals in the core.

The coordinated study of the oxygen and magnesium isotopes and the petrography indicates that the EK1-04-2 Type C CAI underwent multiple heating events after a precursor CAI formation. The precursor CAI was formed at 0.43 Myr after the formation of the Solar System defined by canonical CAI formation. At least 1.6 Myr after the precursor CAI formation, the CAI was partially melted and the partial melting melt exchanged oxygen isotopes with surrounding ¹⁶O-poor nebular gas. ¹⁶O-poor olivine and pyroxene in the core were recrystallized from the partial melting melt. Subsequently, Al-rich chondrules accreted on the CAI, and the CAI experienced partial melting again and recrystallized to form the mantle. The oxygen and magnesium isotopes in anorthite were redistributed during thermal metamorphism in the Allende parent body. Our study reveals that the CAI had been retained in the solar nebula for at least 1.6 Myr and underwent multiple melting events in the nebula, and oxygen and ²⁶Al-Mg systematics has been partially disturbed depending on crystal sizes by metamorphism on the parent body.

Mixing of exogenic impactor materials on the surface of asteroids: Disruption, penetration, and consolidation

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Dawn observation found dark materials on the surface of the hypothesized HED meteorites' parent body, asteroid 4 Vesta. The dark materials are interpreted to be fragments of carbonaceous chondrite impactors, while HED meteorites were known to include carbonaceous chondrite clasts. Meteorite fall named Almahata Sitta, originally was an asteroid 2008 TC₃, is a polymict ureilite with various foreign materials such as enstatite chondrites, ordinary chondrites, one carbonaceous chondrite, etc. Dust particles returned from comet 81P/Wild 2, a Jupiter-Family Comet (JFC) contained materials condensed at high temperature formed near the Sun. It shows that materials of different temperature histories were mixed into the comet at some point during its formation and evolution. Mixing process of materials of different origins occurred ubiquitously or repeatedly on small bodies.

We performed impact experiments of simulated small bodies as targets using various impactors including rocks, metals, and porous sintered materials in order to examine the degree of disruption, penetration, and consolidation of impactors in relation with the material properties and impact velocity for the purpose of understanding the mixing process of materials due to collision. We found that (1) degree of impactor's disruption can be described by the ratio of initial pressure to dynamic strength of the impactor, (2) penetration depth of silicate impactor is only up to 100 times of its size even when it collides with an icy body of 90% porosity, however, (3) impactor with large porosity can survive with larger fraction and can penetrate deeper, probably because local microscopic collapse of pores inhibits growth of overall fracture, and (4) when impacted on fine regolith at high velocity, mutual consolidation of impactor fragments and also with regolith particles occurs owing to temperature increase due to compaction of pores in regolith.

We will summarize these results in relation with porosity, size of impacting body, and impact velocity and will discuss them in comparison with observational evidences, especially, of asteroids.

Keywords: asteroids, collision, laboratory experiments, meteorites, regolith

Experimental confirmation of ringwoodite crystallization from shock-induced melts

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Many high-pressure phases have been identified in meteorites that experienced heavy collisions. The presence of high-pressure phase may provide an estimate of pressure condition. However, the dynamic formation conditions may not be equal to those at static pressures and there is no firm experimental report to indicate the ringwoodite formation at dynamic pressures, although there are Hugoniot data and trials to synthesize ringwoodite by shock compressions. We tried to confirm the ringwoodite formation by hypervelocity impacts from two powdered mixtures of biotite and cristobalite (sample A) and phlogopite and cristobalite (sample B) for Fe-rich and Mg-rich ringwoodites, respectively. When we used stainless steel containers for recovery, the container had reacted with the biotite melt to form chromite spinels. No spinel phase was observed in sample B. When we used copper containers for sample A, X-ray diffraction data on the recovered samples indicated a spinel phase ($a = 0.8257$ nm). Because the lattice constant is greater than that of Fe₂SiO₄ (ahrensite) and significantly less than those of magnesioferrite and magnetite, the composition can be a Fe-rich ringwoodite. However, detailed scanning electron microscopy indicated no obvious crystals on the polished surface where there were many spherical voids. Finally the Raman spectroscopy investigations detected spectra similar to Fe-rich ringwoodite in the voids. We will try to investigate the spinel phase using analytical transmission electron microscopy.

The present experimental results confirm the formation of ringwoodite from shock-induced melts. Further studies need to provide Mg-rich ringwoodite formation and the minimum dynamic pressures required to the formation. If such experiments are extended to the other high-pressure phases present in meteorites, the shock pressure estimation will be more powerful and helpful than the present.

Keywords: ringwoodite, shock-induced melt, crystallization, recovery shots

Formation of metastable lingunite

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Lingunite (hollandite-type NaAlSi₃O₈) has been frequently found in shocked meteorites with other high-pressure minerals (Liu and El Goresy, 2007). According to the laser-heated diamond anvil cell (LHDAC) experiment by Liu (1978), following the decomposition of albite (NaAlSi₃O₈) into jadeite (NaAlSi₂O₆) plus silica (SiO₂) at 2-3 GPa, these phases recombine to form lingunite in the range of pressure between 21 and 24 GPa, and then it decomposes again into calcium ferrite-type NaAlSiO₄ plus stishovite at pressures above 24 GPa. Similarly, Tutti (2007) observed lingunite as a minor phase at 21-23 GPa and 2273K using LHDAC. In contrast to these LHDAC studies, high-pressure experiments using multi-anvil type (MA) apparatus revealed that the maximum solubility of NaAlSi₃O₈ component in hollandite structure is limited to ~50 mol% at 14-25 GPa and 1073-2673K (Yagi et al., 1994, Liu, 2006) and NaAlSi₃O₈ lingunite is not stable at least up to 2273K (Akaogi et al., 2010). This contradiction has not been solved yet, which makes it difficult to understand the shock conditions for the presence of lingunite in shocked meteorites.

To investigate the lingunite puzzle, we focused on the formation process of lingunite by conducting time-series experiments. We performed high-pressure experiments at 18-27 GPa and 1073-2023K using both LHDAC and MA apparatus. Powders of natural albite, oligoclase and labradorite are used as starting materials. Existing phases were identified by X-ray diffraction method.

The quenching experiments using MA apparatus revealed that lingunite does not form in 5 min, but forms in 60 min as a single phase from oligoclase at 20 GPa and 1473K. In situ X-ray diffraction study indicated that oligoclase becomes amorphous with increasing pressure and temperature. At 22 GPa and 1473K, lingunite first crystallizes from the complete amorphous oligoclase in 100 sec, and it decomposes into stishovite and CAS phase in 60 min. These results suggest that lingunite forms as a metastable phase by solid-state reaction after the amorphization of oligoclase, which might have also occurred with maskelynite in shocked chondritic meteorites (Tomioka et al., 2000). In contrast, lingunite was not observed when albite and labradorite were used as starting materials. The amorphization pressure increases with increasing albite component. The pressure condition for complete amorphization of albite is higher than that for the lingunite formation. No lingunite observed from the albite sample in this study implies that the complete amorphization is required for the metastable formation of lingunite by solid-state reaction. In the case of labradorite, lingunite was not formed even after the complete amorphization. This is consistent with the observation that lingunite with labradorite composition in martian shocked meteorites crystallized not by solid-state reaction but from plagioclase melt (e.g., El Goresy et al., 2013).

Shock-metamorphosed zircons from the Jack Hills metaconglomerate in the Narryer Gneiss Complex, Western Australia

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An intense flux of extraterrestrial bodies into inner solar system during ca. 3.8-4.1, called as Late Heavy Bombardment (LHB), has been hypothesized originally from studies of the Moon. Extraterrestrial impacts by meteoritic bodies and comets on the early Earth play a significant role for the initial state of crustal, atmospheric and biological evolution. Considering the planetary size, the Earth should have suffered approximately 20 times the impact flux compared to the Moon. Ancient terrestrial evidence of impact in the early Earth is, however, scarcely preserved due to surface erosion, sedimentary burial and tectonic destruction. To date, the oldest impact structure on the Earth is the 2.02 Ga Vredefort Dome, South Africa, and another oldest evidence of impact is 3.47-3.24 Ga spherule layers in the Barberton Greenstone Belt, South Africa. The impact chronology from these spherule layers suggest that the impact flux was significantly higher 3.5 Ga than today, but there are no terrestrial evidence of impact prior 3.5 Ga.

Geological information during Hadean era (before 4.0 Ga) can be deduced from detrital zircons as old as 4.4 Ga preserved in metasedimentary rocks at Jack Hills in the Narryer Gneiss Complex, Western Australia. Previous studies have reported that the Jack Hills metasedimentary rocks contain detrital zircons with ages continuously spanning from ca. 3.0 up to 4.4 Ga, but evidence of impact, such as shock-metamorphosed minerals, have not been confirmed. In the study we first report shock-metamorphosed detrital zircons from the Jack Hills metaconglomerate, in the Narryer Gneiss complex, Western Australia. A total of 8993 detrital zircons were investigated for the surface and internal structure using a scanning electron microscope and optical microscope with/without acid treatment, and four types of shock-metamorphosed zircons were currently identified; (1) curvi-planar (non-planar) feature (n = 6), (2) multiple sets of planar feature (n = 7), (3) partly granular (polycrystalline) texture (n = 2), and (4) fully granular texture (n = 10). Of these four, multiple sets of planar feature are proved for diagnostic evidence for impact origin, and now observed as annealed (decorated) planar feature, probably due to post-impact thermal heating or regional metamorphic overprint. Coarse polycrystalline zircon represents several micro-meter sized crystallites in a glassy $ZrSiO_4$ matrix that may resulted from shock-induced amorphization and subsequent recrystallization. This grain shows abundant micro-vesicles and tiny $ThSiO_4$ phase suggesting incipient melting and degassing.

Shock-metamorphosed zircons are often utilized for impact-dating due to their partly or completely Pb-loss (age resetting). Therefore, impact age determinations on shock-metamorphosed detrital zircons from the Jack Hills metaconglomerate would provide significant clues not only for the deciphering the impact history on the early Earth but also for the verifying LHB hypothesis.

Shock P-T history of Martian meteorites as revealed by electron microscopy of "brown" olivine

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Introduction: Most Martian meteorites are heavily shocked and exhibit various shock features. Among these shock features, olivine darkening ("brown" olivine) is unique in Martian meteorites. Detailed observation of brown olivine by TEM reveals that olivine darkening is due to the presence of iron nano-particles (Fe metal and/or magnetite) ranging 5-20 nm in size in olivine. Although nano-particles are considered to be formed by heavy shock metamorphism, their formation processes and conditions are not well understood and there is a possibility to obtain new constraints on shock events on Mars. In this study, we observed and compared Martian meteorites containing brown olivine with those without brown olivine and heavily-shocked chondrite for revealing their darkening processes and conditions and seeking singularities of planetary shock events on Mars.

Sample: Eight shergottites (NWA 1950, LAR 06319, LEW 88516, Y984028, NWA 1068, RBT 04261, LAR 12095 and Tissint) and one L chondrite (NWA 4719) were observed in this study. Four shergottites (NWA 1950, LAR 06319, LEW 88516, Y984028) contain darkened olivine. Olivine grains in other samples are almost colorless although they are certainly heavily shocked.

Results and Discussion: Observation of brown olivine by optical microscopy reveals heterogeneous coloration on the scale of tens of μm . In three shergottites (LAR 06319, LEW 88516, Y984028) olivine around shock melts is recrystallized and not darkened.

The brown areas look brighter in BSE images and have fewer cracks and low crystallinity by EBSD compared to the colorless areas. Observation at high magnifications reveals that some brighter areas of NWA 1950 are composed of abundant lenticular areas with submicron-sized Fe particles around them. Shergottites with brown olivine contain no high pressure minerals although they show similar lamellar textures to olivine with high pressure polymorphs.

In contrast, shergottites without brown olivine contain high pressure minerals and also partially darkened areas in olivine adjacent to shock melt veins. These darkened areas show similar microstructures to those of brown olivine, and therefore their formation processes seem to be identical and olivine darkening (formation of iron nano-particles) requires high temperature because darkened areas are present only near shock melts in these meteorites without brown olivine. It is conceivable that the heterogeneity of olivine darkening corresponds to temperature difference. Since temperature heterogeneity is vanished within about <1 sec, iron nano-particles need to be formed in such a short time. The absence of Si-rich phases means that mechanism of nano-particle formation is similar to the initial process found in olivine reduction experiments whose rate is limited by atomic diffusion rates. Atomic diffusion in olivine is too slow to reflect temperature heterogeneity to their color distribution in such a short time, and therefore rapid diffusion, for example transformation to high pressure minerals, is needed. Lenticular areas in brown olivine with low crystallinity may have been a high pressure mineral and, if that is the case, it seems easy to reflect temperature heterogeneity to their coloration. Thus, Martian meteorites with brown olivine have undergone such a high pressure-temperature condition in a short time (~a few ms) on which most olivine in the meteorites is transformed to high pressure polymorphs. Temperature increase throughout meteorite induces a slow cooling of shock melt and it seems also related to recrystallization of olivine and back-transformation of high pressure minerals around shock melt.

This condition may be induced by an extremely strong shock event and meteorites with brown olivine have undergone such stronger shock than those containing high pressure minerals and colorless olivine. Thus, the collisional events ejecting rocks from Mars may be extremely strong compared to those affecting the other meteorites.

Keywords: Mars, Martian meteorite, High pressure mineral, shock, brown olivine

Pressure dependence of cathodoluminescence spectra of shocked quartz.

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Impact cratering is a ubiquitous process on both terrestrial planets and small bodies in the solar system. Study on impact craters on the Earth provides a unique opportunity to constrain planetary-scale impact phenomena. In particular, reconstruction of distribution and shock pressure recorded on the shock-metamorphosed minerals would provide vital information on partition of the impact energy and cratering mechanism on Earth.

Quartz, which is one of the most abundant and widely distributed rock-forming minerals on the Earth, has been conventionally used to evaluate shock pressure on the minerals. However, the precious shock estimations using quartz, such as PDFs, are rather qualitative, hence it is required for more detailed evaluation of shock pressure to develop new advanced method. Recently, cathodoluminescence (CL), emissions of photon from materials in ultraviolet to infrared wavelength regions, especially CL of quartz is expected to be used as shock barometer because of change of luminescent properties with shock metamorphism, but systematic study focused on pressure dependence of CL spectra have never been reported. In this study, we conducted a series of systematic shock recovery experiments of impact on quartz crystals and analyzed recovered samples to evaluate the CL spectral features and their dependence on shock pressure.

The shock recovery experiments were performed on natural and synthesis quartz crystals in the National Institute for Material Science (NIMS) using a one stage propellant gun. The velocity range of flyer plate was 0.5 to 1.8 km/s, which produces peak shock pressure from 5 to 40 GPa. Recovered samples were polished to make thin sections, and then are coated by carbon for CL analysis. CL measurements of grain by grain were conducted at Okayama University of Science using a SEM-CL (SEM combined with a grating monochromator) instrument.

Based on CL measurements of over a hundred shock-recovered quartz grains, we found drastic change in spectral pattern with an increase in shock pressure. The starting material of synthetic and natural quartz has only one broad emission peak around 630 nm, but another broad peak around 450-460 nm (blue emission) appears from the shock-recovered samples at 10 to 20 GPa. At higher pressure, the CL intensity of blue emission of shocked quartz increases drastically up to 100 times as large as that of the starting materials. On the other hand, CL emission intensity around 630 nm changes less than 3 times in spite of the pressure increase.

CL spectra of quartz with β -form (e.g., quartz from Goroku, Sendai) show extremely intense blue emission and unique luminescent properties depending on temperature (e.g. activation energy) similar to quartz from terrestrial impact craters, suggesting probably relationship of the blue CL with defects associated with Daufine twins formed as a result of α - β transition. On the other hand, Daufine twins are also confirmed in the experimentally shocked quartz under TEM observation. Therefore, the rise of blue emission intensity in our measurements suggests generation and density increase of defects related to Daufine twins with ascending shock pressure. Quantitative evaluation of this relationship presumably enables us to construct a new quantitative pressure barometer or thermometer for shocked quartz grains.

Keywords: shock metamorphism, shocked quartz, cathodoluminescence

High-pressure polymorphs in Gujba CB type carbonaceous chondrite.

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One of the most unambiguous evidences for shock metamorphism is a dense polymorph, high-pressure polymorph in and around the shock-melt veins and/or melt-pockets of shocked meteorites. Now, the existences of high-pressure polymorphs have been reported from ordinary chondrite, enstatite chondrite, ureilite, eucrite, iron meteorite, lunar meteorite, Apollo sample and Martian meteorite (e.g., Ohtani et al., 2004; Miyahara et al., 2014). On the other hand, it is widely accepted that carbonaceous chondrites were less shocked than ordinary chondrites, implying that high-pressure polymorphs would not be included in carbonaceous chondrites. However, Hollister et al. (2014) identified ringwoodite and stishovite from Khatyrka CV type carbonaceous chondrite. Shock metamorphism in carbonaceous chondrites may be overlooked or underestimated. CB type carbonaceous chondrite is a unique grouplet because it consists mainly of metallic Fe-Ni and chondritic fragment (Weisberg et al., 2001). Although the origin of CB type carbonaceous chondrite has been under debated, some previous studies propose that it may be formed through a planetesimal collision (Weisberg et al., 2010). In this study, we investigated a high-pressure polymorph in CB type carbonaceous chondrite, Gujba to clarify its shock metamorphism history and origin using FEG-SEM, EPMA and laser Raman spectroscopy.

We prepared a polished Gujba sample for this study. Gujba studied here consists of metal and chondritic fragment. Shock-melt veins occur widely between the chondritic fragments and metals. The major constituent minerals of the chondritic fragments are low-Ca pyroxene (Fs₁₋₅, En₈₉₋₉₈, Wo₀₋₆), Mg-rich olivine (Fa₁₋₁₈, Fo₈₂₋₉₉) and Ca pyroxene (Fs₁₋₁₃, En₄₁₋₆₆, Wo₃₂₋₅₇) based on EPMA analysis. Many mineral fragments and fine-grained chondritic fragments are entrained in the shock-melt veins. We confirmed the existences of many kinds of high-pressure polymorphs from such fragments and chondritic fragments adjacent to the shock-melt veins. Raman spectroscopy analyses indicate that olivine entrained in the shock-melt veins transform into wadsleyite. A small amount of ringwoodite is accompanied with some wadsleyite. Low-Ca pyroxene is replaced with akimotoite, majorite or probably bridgmanite. Minor Fe-rich olivine (Fa₃₉₋₄₀) and albitic feldspar (Ab₈₂An₁₃Or₅) are entrained in the shock-melt veins. Lamellar ringwoodite occurs in the Fe-rich olivine. The albitic feldspar is replaced with jadeite, lingunite or maskelynite. We also clarified the distributions of these high-pressure polymorphs in Gujba studied here. High-pressure polymorphs occur ubiquitously in and around the shock-melt veins, indicating that the parent-body of Gujba was heavily shocked. The metals, which consist of kamacite and minor FeS, show evidence for melting. Alternatively, we can also infer that Gujba is a part of shock-met veins induced by a collision.

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Keywords: carbonaceous chondrite, high-pressure polymorph, shock metamorphism

Symmetry of majorite garnet in shocked chondrites revisited: A TEM study

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Majorite is a garnet-structured mineral with a composition on the join $(\text{Mg,Fe})\text{SiO}_3$ - $(\text{Mg,Fe})_3\text{Al}_2\text{Si}_3\text{O}_{12}$, which contains Si in the octahedral sites. Al-free majorite samples synthesized by Kawai-type multianvil apparatus are known to have a tetragonal symmetry (space group $I4_1/a$) [1]. However, all of natural Al-free majorite samples found in shocked chondrites have been reported to be cubic ($Ia-3d$) [2]. Single-crystal X-ray study of a synthetic MgSiO_3 majorite clarified the tetragonal distortion of majorite is caused by Mg-Si ordering in the octahedral sites [3]. Subsequent microstructural observations of synthetic Al-free majorite by TEM concluded that the tetragonal phase is formed from the cubic phase through the cation ordering even upon rapid quenching (10^3 °C/sec) and the symmetry reduction phase derives modulated and twinning structures [4]. On the contrary, cubic $(\text{Mg,Fe})\text{SiO}_3$ majorite in shocked chondrites is inferred to have been preserved due to higher cooling rate than that in high-pressure synthesis where the cation ordering is inhibited.

Due to very small tetragonal distortion from the cubic phase ($c/a = 0.99$) and very weak extra reflections for the tetragonal phase, it is difficult to judge the symmetry of small amount of natural majorite samples by powder X-ray diffractometry. For the symmetry analyses of submicron-sized grains, single-crystal electron diffraction is a suitable method, since the intensities of weak reflections to diagnose the tetragonal symmetry are enhanced by the effect of dynamical diffraction. In this study, we revisit symmetries of majorite grains in shocked ordinary chondrites (Tenham; L6, Y-75100; H6) and also synthetic $(\text{Mg,Fe})\text{SiO}_3$ majorite by TEM. The intensity of $\{101\}$ reflections, which appears only for the $I4_1/a$ tetragonal phase, is under investigation by selected area electron diffraction.

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Keywords: majorite, shock metamorphism, high-pressure phase transition, TEM

Basin forming event on Vesta: Petrologic evidence from a diogenite, NWA 5480

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The HED (Howardite, Eucrite, Diogenite) meteorites are the largest group of achondrites, and are derived from the regolith and crustal lithologies of asteroid 4-Vesta, which is the only surviving differentiated protoplanet in the Solar System. Diogenites are considered to have been derived from intrusions from within the crust or from a deep global layer. Some of them display high levels of platinum group element (PGE) concentrations that have been attributed to impact events on the parent body.

NWA 5480 is an unusual olivine diogenite. It has a heterogeneous crystalline texture. Irregular and subrounded clasts of dunite and fragments of olivine and chromite are set in an igneous matrix mainly composed of Low-Ca pyroxene. In some cases, large dunite clasts are intersected by pyroxene matrix. Flow textures are observed near some clasts. Olivine, chromite, and pyroxene show minor chemical zoning, implying relatively rapid cooling compared to typical diogenites. NWA 5480 contains a significant amount of PGE (CI x ~0.001 for Ir) with chondritic relative proportions. All these line of evidence support that NWA 5480 is an impact melt breccia from a target composed of olivine and pyroxene-rich lithologies.

Upon heating of olivine diogenites, low-Ca pyroxene is the earliest phase to melt, and olivine and chromite are the last at >~1600 C. The irregular and rounded shape resulted from resorption, and pyroxene veins and flow textures formed by violent mixing during impact melting. The pyroxene matrix crystallized from impact melts. Cooling rates estimated from the shape of Ca zoning of profiles near the rims in olivine fragments could be several tens of C/year, corresponding to burial depths less than a few km in impact melt sheet. An impact crater with diameter >a few hundreds km would be needed to produce impact melt sheet >a few km in thickness. Thus, we suggest that NWA 5480 was derived from impact melts from a very large crater of Vesta. The bulk chemical compositions indicate that the target was an area where olivine and orthopyroxene-rich rocks are largely exposed. However, there are no such areas on Vesta except minor olivine spots. Thus, it is likely that NWA 5480 sampled a part of large impact melt sheet (>a few km thick) formed by melting of deep crustal materials rich in olivine and orthopyroxene. One of the best candidates is the Rheasilvia basin (~500 km diameter), where orthopyroxene-rich materials were observed in the crater floor.

Keywords: diogenite, Vesta, impact melt breccia, impact crater

^{147}Sm - ^{143}Nd and ^{146}Sm - ^{142}Nd chronology of a basaltic eucrite, NWA 7188

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Eucrites are achondritic meteorites originating from the Vesta's crust. They can be petrographically classified into basaltic and cumulate eucrites. Determination of precise ages for eucrites will constrain the period of igneous activity and the following thermal metamorphism of Vesta and may further provide insights into its differentiation and thermal history. We investigate the long-lived ^{147}Sm - ^{143}Nd ($T_{1/2} = 1.06 \times 10^{11}$ yr) and the short-lived ^{146}Sm - ^{142}Nd ($T_{1/2} = 6.8 \times 10^7$ yr [1]) systematics of a basaltic eucrite, NWA 7188 and compare the results with the ages obtained in previous chronological studies on cumulate and basaltic eucrites. To obtain highly precise age data, we developed the techniques for determining Nd and Sm concentrations and Nd isotope compositions in meteorite samples.

NWA 7188 was crushed and sieved into four sizes; G1) 500 — 1700 μm , G2) 250 — 500 μm , G3) 106 — 250 μm , and G4) ≤ 106 μm . G3 and G4 were separated into pyroxene and plagioclase grains by handpicking. We determined the ^{147}Sm - ^{143}Nd and ^{146}Sm - ^{142}Nd ages of NWA 7188 using G1, G3-px, G4-px, G3-pl, and G4-pl. These were dissolved using a mixture of concentrated pure acids (HClO_4 , HF, and HNO_3). After the sample digestion, $\sim 10\%$ of the solution was removed and mixed with ^{149}Sm - and ^{145}Nd -enriched spikes to determine the Sm and Nd concentrations by ID-ICP-MS (X-series II, Thermo). The remainder of the sample solution was used for highly precise Nd isotope analysis by TIMS (TRITON plus) with the dynamic multicollection mode. Nd was separated by a three-step column chemistry procedure; 1) major elements were removed by passing through a cation exchange resin, 2) Ce was removed using the LN resin (Eichrom) by oxidizing Ce^{3+} into Ce^{4+} using KBrO_3 [2] and 3) Nd was separated from Sm using the LN resin. We achieved $\text{Ce}/\text{Nd} = \sim 3 \times 10^{-5}$ and $\text{Sm}/\text{Nd} = \sim 4 \times 10^{-5}$ with $\geq 91\%$ Nd recovery.

The ^{147}Sm - ^{143}Nd mineral isochron of NWA 7188 yields an age of 4203 ± 970 Ma. In contrast, we obtained a much older ^{146}Sm - ^{142}Nd mineral isochron age of $4549 \pm {}^{28}_{40}$ Ma when an initial solar system ratio of $^{146}\text{Sm}/^{144}\text{Sm} = 0.0094$ at 4568 Ma was applied [1]. It is presumed that thermal metamorphism on the Vesta has some effects on the ^{147}Sm - ^{143}Nd age while the ^{146}Sm - ^{142}Nd age represents the timing of the last Sm-Nd isotopic closure. Therefore, we use the ^{146}Sm - ^{142}Nd age of NWA 7188 in the following discussion.

The ^{146}Sm - ^{142}Nd age ($4549 \pm {}^{28}_{40}$ Ma) for NWA 7188 is consistent with the ^{147}Sm - ^{143}Nd age for cumulate eucrites (4546 ± 8 Ma [3]) within analytical uncertainties. This suggests that the parent body processes associated with the last Sm-Nd isotopic closure were contemporaneous for basaltic and cumulate eucrites. Likewise, the ^{146}Sm - ^{142}Nd age of NWA 7188 is not resolvable from the metamorphic age of Agoult [4]. According to the ^{53}Mn - ^{53}Cr systematics [5], the last global Mn/Cr fractionation in the mantle of the Vesta occurred at 4564.8 ± 0.9 Ma, the timing when basaltic magmas have formed in the mantle. This implies that basaltic eucrites quenched rapidly on the surface of eucrite parent body, but thermal metamorphism may have affected both Sm-Nd and U-Pb systematics. No apparent age difference between basaltic and cumulate eucrites implies that both types of eucrites might have experienced similar cooling history as opposed to their petrographic distinction, or more likely that the time difference is too subtle to be resolved by the ^{146}Sm - ^{142}Nd system.

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Keywords: basaltic eucrite, Sm-Nd chronology, NWA 7188

Olivine petrofabric and chemical study of Divnoe ungrouped primitive achondrite

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Brachinite is a small group of achondrites mostly composed of olivine. Currently ~30 meteorites belong to brachinite, however, what are called “brachinite-like” but classified as “ungrouped” exist (e.g., Day et al. 2012; Keil 2014). Such “brachinite-like” meteorites have a similar rock texture, oxygen isotopic compositions or trace elements abundance to brachinite but do not perfectly match, and so they are classified as ungrouped. It is important to study these meteorites to understand the formation process of brachinite in general and their parental differentiated asteroid(s). Divnoe is one of such “brachinite-like” meteorite discovered in Russia in 1981 (Graham 1983; Petaev et al. 1994) and recently some meteorites similar to Divnoe have been found from hot and cold deserts (e.g., RBT 04239 and Tafassasset). These findings may establish a new achondrite group (“Divnoeite”) including some of above “brachinite-like” meteorites (Gardner et al. 2007). In this study we analyzed Divnoe from mineralogical and chemical points of view to better understand its petrogenesis.

Divnoe is a medium- to coarse-grained achondrite (~0.5-2.0 mm in grain size) dominated by olivine (Fo₇₂₋₈₀) and apparent shape preferred orientation (SPO) of olivine grains is observed by optical microscopic observation. It is known that dislocation-creep controlled flow of upper mantle materials develops crystallographic preferred orientation (CPO) of olivine and seismic anisotropy in the Earth (Ohuchi 2013). Recently, Tkalcec et al. (2013) studied lattice preferred orientation (LPO) of olivine in diogenite and proposed solid-state plastic deformation in the dynamic interior of 4Vesta. Our EBSD (Electron Back Scatter Diffraction) analysis of Divnoe olivine grains shows that they are preferentially aligned along [001], which is parallel to elongation of olivine grains. The dimensions of [100] and [010] are randomly oriented. This observation indicates that CPO of Divnoe olivine was achieved when olivine grains accumulated in magma rather than plastic deformation after solidification in the interior of the parent body, which is consistent with previous study (Ando et al. 2003). Therefore, Divnoe is likely an olivine cumulate rock. However, we found that its bulk chemical composition has high abundance of siderophile elements (Ir, Pt: ~0.5 x CI), which is unusual for an olivine cumulate. Such high abundance of siderophile elements may be derived from addition of an iron meteorite component by impact. It is required to analyze other Divnoe-like meteorites to see whether (1) olivine SPO is present or not and (2) such chemical signature (e.g., enrichment of siderophiles) is similarly seen.

Crystallization and cooling conditions for the diogenite formation in the turbulent magma ocean of the asteroid 4 Vesta

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The asteroid 4 Vesta has been completely differentiated to core and mantle despite its small size, of which surface materials are howardite-eucrite-diogenite (HED) meteorites that we know the detailed petrology, and therefor, is a good target for understanding differentiation of terrestrial planets. A new differentiation model for crust formation was developed by taking magma ocean fluid dynamics, chemical equilibrium, presence of ²⁶Al, and cooling into consideration with special care to crystal separation. The role of crystal size, thickness of the conductive lib, and fO₂ are evaluated as parameters. Large crystals (1cm) settle and form a km-thick cumulate layer of orthopyroxene with Mg# of 0.70-0.90 in 20 thousand years, which almost agrees with the Mg# of diogenites, whereas thinner layers are formed if the grain size is smaller.

Keywords: magma ocean, Asteroid 4 Vesta, HED meteorites

A mineralogical and chemical study of primitive achondrite NWA 6704

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Introduction: Primitive achondrites provide unique insights into the early stage of growth and differentiation of planetesimals. Northwest Africa (NWA) 6704 is an ungrouped primitive achondrite found in Algeria in 2010. This meteorite is composed mainly of low-Ca pyroxene, olivine, feldspar, chromite, awaruite, heazlewoodite, pentlandite, and whitlockite. The U-Pb dating of this meteorite shows the crystallization age of 4563.75 ± 0.41 Ma (1). To better understand the formation process of this primitive meteorite, we carried out a mineralogical and chemical study of NWA 6704 using SEM-EDS, EPMA and LA-ICP-MS.

Results & Discussion: Seven polished thin sections (TS-1⁷; 10×18mm; 10×20mm; 9×10mm; 10×12mm; 10×11mm; 15×28mm; 19×28mm) have been investigated. The texture is best characterized by the existence of many orthopyroxene (Opx) megacrysts up to 1.56 cm in length ($\text{Fs}_{40-42}\text{En}_{53-57}\text{Wo}_{3-4}$). Olivine (Fa_{50-53}) typically occur as interstices associated with feldspar and have subhedral shapes, but locally occur as the vermicular olivine within the Opx. The vermicular olivine share the same optical extinction position under crossed-nicols and occurs only in one place near the center of each Opx crystal if present. Feldspar ($\text{Ab}_{91-93}\text{An}_{5-6}\text{Or}_{2-30}$) occurs in the Opx and the interstices as anhedral cusped grains with extremely low dihedral angles and sharing the same optical extinction position over up to 1cm, which implies that it's pseudomorph of a melt-filled interstitial pore. Other phases include chromite ($\text{Cr}\# = 0.90-0.96$) and awaruite (78-81 wt% Ni) also occur associated with feldspar. We estimated $f\text{O}_2$ of FMQ -2.6 using the Oliv-Opx-Spl oxygen geobarometer (4).

The texture is properly represented by aggregate of large Opx hollow (skeletal) megacrysts with finer interstices. Other phases such as olivine are present in the hollow cores, and some of them are isolated from the interstices. The contiguity of feldspar in the Opx megacrysts with or without olivine in three dimensions is clearly demonstrated by the fact that the same optical extinction position are shared by feldspar isolated in the megacrysts and those present in the interstices. The distribution of the Opx hollow crystal and the vermicular olivine are highly heterogeneous. In TS-1 and TS-6, Opx <6.2 mm across are dominant and more than five of them include vermicular olivine; in TS-2⁵ and TS-7, Opx megacrysts up to 1.56cm across are common and only a few of them contain vermicular olivine. The vermicular olivine may be a decomposition product of the precursor pyroxene through the abrupt heating (>1400 °C). There is a negative correlation between size of Opx and the number of vermicular-olivine bearing Opx. Given this fact, a new scenario can be derived: this vermicular olivine and its host acted as nuclei for crystallization of hollow Opx crystals, and the number of nuclei limits the size of Opx. Thus, more nuclei in a given area result in smaller grain size as seen in TS-1,6. The abnormally large size of Opx and its hollow morphology indicates that the initial crystallization occurred under rapid cooling. The SEM images, however, show that some pigeonite contain sub-micrometer-size augite ($\text{Fs}_{17}\text{En}_{45}\text{Wo}_{39}$) exsolution lamellae. We estimated equilibrium temperatures of 1050 °C using two-pyroxene geothermometry (3) and of 773 °C using Oliv-Spl FeMg₋₁ exchange thermometry (4). The cooling rate between 1100-950 °C was estimated to be ~0.02 °C/hr from the thickness and wavelength of multiple exsolution lamellae (5), indicating significant decrease in cooling rate at a later crystallization stage. The inferred thermal history suggests high internal temperature of the parent planetesimal due to ²⁶Al decay, which was abruptly heated by impact followed by rapid cooling up to the ambient temperature to sustain slower cooling.

References: (1) Iizuka et al. (2013), (2) Warren et al. (2012), (3) Lindsley & Andersen (1983), (4) Ballhaus et al. (1991), (5) Jackson (1961)

Keywords: Primitive achondrite, NWA6704

Development on the post-ionization SNMS and application for the isotopic measurement of Presolar SiC grains

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Isotopic composition of the individual presolar grains in primitive meteorites provide us the information on nucleosynthesis in Asymptotic Giant Branch stars (AGB stars) or supernova explosion that had occurred prior to the formation of the Solar system. So far, isotopic measurement of individual presolar grains often has been carried out by using Secondary Ion Mass Spectrometry (SIMS). However, since secondary ion yield of SIMS is less than 1 %, precise analysis of minor elements in individual grains has been difficult. That is, improvement of sensitivity is highly desired for better understanding of the nucleosynthesis.

Here, we report on the development on the post-ionization Sputtered Neutral Mass Spectrometry (SNMS) and an application for the isotopic measurement of presolar silicon carbide grains (SiCs). In this analytical system, the neutrals particles sputtered by Ga ion beam of which diameter is 40nm~2 μm was post-ionized by femt-second laser, and separated by the multi-turn time-of-flight mass spectrometer 'MULTUM' depending on their masses. Last year, we has achieved the about 1000 times higher secondary ion yield for Pb signals with high mass resolution (Nakabayashi et al. 2014).

Based on the preliminary Si isotope analysis using SNMS, we confirmed that current SNMS system shows good reproducibility of terrestrial Si isotopic ratios and figured out that the instrumental mass fractionation is -150~200 ‰/amu. Moreover, we successfully separate ²⁸Si (=27.977 amu) and ²⁹Si (=28.976 amu) peaks from interference peaks such as N₂ (=28.006 amu), CO (=27.995 amu) and ²⁸SiH (=28.985 amu) by using MULTUM. Finally, we also confirm the heavy isotopic anomaly of Si isotopes ($\delta^{30}\text{Si}/^{28}\text{Si}=100\sim 200\text{‰}$, $\delta^{29}\text{Si}/^{28}\text{Si}=130\sim 200\text{‰}$) of presolar SiC candidates extracted from Murchison meteorite, which are consistent with those of previous studies.

Keywords: SIMS, presolar grain, in-situ analysis, isotope anomaly, nuclear synthesis, meteorite

Development of precise in-situ U-Th-Pb dating of phosphate using LA-MC-ICP-MS

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Phosphates in meteorites have high concentration of U and Th (<10 ug/g) and therefore, time-resolved history for planetesimal formation has been constrained based on U-Pb systematics using phosphates in chondrites^{1);2)}. To establish a reliable chronological scenario, in-situ dating with high spatial resolution is important because these target material could possibly experience the gain or loss of Pb through the thermal effect or shock metamorphism by impacts among meteorites³⁾ and interaction with water. To clarify the time interval for the formation of the chondritic parent bodies, required time resolution is a few million years at least¹⁾. However, it is difficult to achieve the enough precision to discuss the planetesimal history using in-situ dating technique. Laser ablation method (LA) is flexible sampling technique in aspects that the spatial resolution can be readily changed and the various sampling protocols are possible. Recently, the highly-precise age determination has been reported by multiple collector ICP mass spectrometry couple with LA sample introducing technique (LA-MC-ICP-MS)⁴⁾. In this study, the development of precise in-situ U-Th-Pb dating method of phosphate using LA-MC-ICP-MS is carried out for the purpose of establishment of the time-resolved story for the thermal and accretion history of the chondritic parent bodies.

Problematic points which limit the analytical precision in in-situ U-Th-Pb age determination of phosphate using laser ablation ICP mass spectrometry (LA-ICP-MS) are as followed: (1)error associated with large counting statistics derived from deficit of amount of sampling, (2)absence of age-homogeneous standard reference material of phosphate desired for precise measurement, (3)heterogeneity of Pb isotopic composition in standard reference material caused by non-radiogenic Pb which might be initially distributed in the material, (4)matrix effect associated with the difference of volatility between measured elements, (5) the isobaric interference on ²⁰⁴Pb from ²⁰⁴Hg. To overcome these problems, in this study, we tested the following approaches listed below: (a)enhancement of the total sensitivity through the improvement of interface region of ICP mass spectrometer and the integration of target material using multiple-spot ablation method⁵⁾, (b)evaluation of the age homogeneity of Madagascar apatite⁶⁾, (c)evaluation of the common-Pb correction for the standard reference material based on ²⁰⁷Pb method, (d)reduction of the matrix effect by utilizing the femtosecond laser ablation system and development of the method to evaluate the system closure using three isotope diagram for Pb, (e)test of isochron method based on normalization by ²⁰⁸Pb. To identify the suitable phase for analysis, in addition, imaging mass spectrometry for U, Th and Pb isotopes in large area (cm x cm order) by LA-ICP-MS is also tested, and the possibility of age determination using laser ablation in liquid method⁷⁾ is estimated as well.

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Keywords: Parent body, Phosphate, U-Th-Pb geochronology, LA-ICP-MS

Hydrogen diffusion experiment of apatite crystal.

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Recently many studies focused on the origin of water in the solar system and Earth with Chondrite, differentiated meteorite, Moon and Mars (e.g., Greenwood et al., 2008; Greenwood et al., 2011; Alexander et al., 2012). Especially H₂O and hydrogen isotopic compositions of apatite in the Moon and ordinary chondrites has been discussed about the origin of water in the magma of the Moon and at the Earth orbit material (Greenwood et al., 2011; Yanai et al., 2014). However, it is still controversial about the origin of water in the apatite grain since it is unclear about the investigation of H-diffusivity in the apatite during thermal metamorphism in the magma and parent body. In this study, we performed H-diffusion experiments using fluorapatite.

Some slices from a durango apatite grain were used in the H-diffusion experiment. These slices polished with diamond are with mirror surface and then were annealed under D₂O/O₂ gas at 400oC-700oC for several hours. H-diffusion coefficients in the samples were determined using depth profiles of D concentration of the samples obtained by secondary ion mass spectrometry (SIMS) (Cameca ims-4f-E7 at Kyoto university and Cameca ims-4f at NIMS). The water content of these slices of a apatite grain were measured and minor impurities of apatite were also measured using SIMS.

In this talk we will report H-diffusion coefficient in the apatite using H-diffusion experiment and discuss about investigation of H-diffusivity in the apatite.

Keywords: Hydrogen, apatite, diffusion coefficient, early solar system, SIMS

Relationship between oxygen diffusivity and Ca/Ti ratio in perovskite

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<**Introduction**> Calcium-aluminum-rich inclusions (CAIs) in carbonaceous chondrites are composed of refractory minerals, such as mellilite, spinel, and perovskite and believed to be the first solid in the early solar system. Previous studies reported heterogeneous oxygen (O)-isotopic compositions among the CAI minerals (Clayton et al. 1973). The O-isotopic compositions are considered to be a marker of O-isotopic composition in the solar nebula (Yurimoto et al. 1998; Itoh and Yurimoto 2003; Park et al. 2012). However, it is unclear that the heterogeneous O-isotopic composition is resulted from whether environment where the mineral crystalized or diffusion during heating event after the crystallization. To discuss this issue, investigation of O-diffusivity of minerals is necessary.

Perovskite crystals show several order of larger O-diffusion coefficients than other CAI minerals (Gautason and Muehlenbachs 1993; Ryerson and McKeegan 1994; Sakaguchi and Haneda 1996). Thus, their O-isotopic compositions can provide us an important key to understand O-diffusivity of CAI minerals and thermal histories of CAIs. Diffusivity of solids is influenced by impurity in the sample (e.g. Sakaguchi et al. 2010). In this study, we performed O-diffusion experiments perovskite with different Ca/Ti ratio to understand O-diffusivity in perovskite.

<**Experimental**> Polycrystalline perovskite samples were prepared by a conventional sintering technique. High-purity reagent-grade powders of CaCO₃ and TiO₂ were used as the starting materials. These powders were mixed as Ca/Ti=0.098-1.002 by ball milling with PSZ balls. The powders were pressed as a sheet and were sintered at 1350 degC for 2 h in the atmosphere. Then, perovskite samples with different Ca/Ti ratio were obtained. Different of Ca/Ti ratio of the samples from initial value due to a contamination of ZrO₂ from PSZ balls was calibrated within the error of ~0.001. The samples were polished by a chemical mechanical polishing and then were annealed under ¹⁸O₂ gas at 750-1000 degC for 20 min~several hours. O-diffusion coefficients in the samples were determined using depth profiles of ¹⁸O concentration of the samples obtained by secondary ion mass spectrometry (SIMS) (Cameca ims-4f). Distributions of ¹⁸O ions in cross section of the samples were observed using high spatial-resolution ion image obtained by NanoSIMS (Cameca NanoSIMS 50).

<**Results and discussion**>Ca-rich sample and Ti-rich sample showed different O-diffusivity. In Ti-rich sample, two contributions to the O-diffusion mechanism are observed in depth profiles of ¹⁸O concentration obtained from Ti-rich perovskite samples. One starts near surface (diffusion I) and the other starts from about a few 100 nm depth (diffusion II) of the sample. In Ca-rich perovskite samples, only one diffusion mechanism was observed. O-diffusion coefficients (D) in Ca-rich samples were larger than that of Ti-rich samples, in both diffusion I and II. D value of 950 degC in the Ti-rich sample (Ca/Ti=0.098) was 7x10⁻¹³cm²/s (diffusion I) and 1x10⁻¹¹cm²/s (diffusion II), whereas the value of a Ca-rich sample (Ca/Ti=1.002) was 8x10⁻¹⁰cm²/s.

High resolution ¹⁸O concentration image of Ti-rich samples showed decrease of ¹⁸O concentration around the grain boundaries, indicating formation of blocking layer for O-diffusion. Such blocking layer has also been reported in BaTiO₃ with perovskite structure (Watanabe et al. 2014).

This study revealed difference of O-diffusivity in perovskite by slight change of Ca/Ti ratio and larger O-diffusivity of Ca-rich perovskite than Ti-rich perovskite. Larger O-diffusion coefficients of perovskite than that of other CAI minerals suggested that perovskite record the final thermal process, which the CAI experienced. This study suggests that shorter timescale required for change of O-isotopic composition for Ca-rich perovskite than previously considered. We will also discuss defect chemistry and mechanism of the O-diffusion in this presentation.

Keywords: perovskite, oxygen diffusivity

Metamorphism of a type B CAI from the reduced CV3 chondrite Efremovka.

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Calcium-aluminium-rich inclusions (CAIs) are among the first rocks to form in the solar nebula and are abundant in CV chondrites [1,2]. Most CAIs have been altered and the understanding of metamorphic effects is essential for interpreting the metamorphic conditions of parent bodies and formation conditions in the solar nebula. Especially, the CV3 chondrite Allende has undergone relatively strong metamorphism, and previous studies have addressed effects of metamorphism on Allende CAIs [3,4]. On the other hand, the CV3 chondrite Efremovka has undergone less metamorphism than Allende [5]. Therefore study of Efremovka is useful for understanding more primitive conditions. In this study, we describe secondary minerals, their textures and modal abundances in a type B CAI from Efremovka (CGI-10), using elemental mapping, quantitative EPMA, cathodoluminescence, and Raman spectroscopy. We compare secondary features of CGI-10 with Allende CAIs, particularly with the Allende type B CAI 4022 (see [3]).

CGI-10 is a type B CAI with primary spinel, anorthite and fassaite, all with 16O-rich compositions, and melilite that is 16O-poor [6]. The main secondary minerals in CGI-10 are sodalite, nepheline, Fe-bearing spinel and an elongate secondary mineral (ESM) with composition $\text{CaAl}_2\text{Si}_2\text{O}_8$. These secondary minerals are concentrated in FeO-alkali-rich domains near the CAI rim. Similar minerals and textures are observed in Allende CAIs [3]; however, FeO-alkali-rich domains are more abundant in Allende CAIs than in CGI-10 (approx. 1.5 mode% in CGI-10 vs. 8% in CAI 4022) and ESM is more abundant in the FeO-alkali-rich domains of Allende CAIs than in CGI-10 (trace in CGI-10 vs. 2.5 mode% in 4022). Furthermore, the main ESM in Allende appears to be dmisteinbergite [7,8], whereas the main ESM in Efremovka CGI-10 appears to be anorthite, based on the similarity of its cathodoluminescence (CL) to the CL of primary anorthite. Dmisteinbergite occurs in coal heaps in Chelyabinsk, Russia that underwent pyrometamorphism at low pressure (Earth surface), over a short time (10 to 15 years), at temperatures ranging up to 1200C and in the presence of gases of varying compositions and variable $f(\text{O}_2)$ [9]. Some crystallization in the coal heaps apparently occurred during annealing while the heaps cooled from peak temperatures [9]. Although peak metamorphic temperatures were lower in Allende than in the Chelyabinsk coal heaps, it is possible that Allende shared some similarities in metamorphic conditions (low pressure, variable gas compositions, short heating & cooling times) that favored the formation of dmisteinbergite instead of anorthite.

In contrast with Allende CAIs [4], no grossular-rich veins were identified in CGI-10, and the mode of secondary minerals in CGI-10 (~2%) is much less than in Allende type B CAIs (~30%), consistent with the interpretation that Allende was metamorphosed at higher temperature than Efremovka [5]. On the other hand, the concentration of Na₂O in CGI-10 is similar to that of a similar Allende CAI (both approx. 0.3 wt%). The similarity in Na₂O-abundances suggests that infiltration of Na into CAIs during metamorphism is not due completely to metamorphic temperature, but is related to other factors (for example, composition and abundance of fluid available during metamorphism). In any case, CGI-10 has undergone some metamorphic recrystallization. Thus, when Efremovka CAIs are used to interpret conditions from the solar nebula, metamorphic effects should also be considered.

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Keywords: Ca-Al-rich inclusions, CV3 chondrites, metamorphism

Hydro thermal experiments of Allende CV3 chondrite under reducing condition

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Introduction

Of a range of small asteroidal bodies, aqueous alteration is one of the most important processes affecting early cosmic materials including carbonaceous chondrites. Aqueous fluids in the chondrite parent bodies modified the primary mineralogical characteristics formed in the solar nebula, and resulted in formation of a variety of secondary phases, including phyllosilicates, magnetite and carbonates. CI and CM chondrites are widely recognized as extensively aqueous-altered meteorites. Other carbonaceous chondrites of types 3 (CR, CO, and CV) and some unequilibrated ordinary chondrites have also clear evidences of various degrees of aqueous alteration. The diversity in alteration assemblages among various chondrites likely reflect the aqueous environment (e.g., temperature, dissolved ion, water/rock ratio, fO_2 , etc.) of the parent bodies. Although many mineralogical, isotopic and theoretical studies has been made to unravel the complex effects of aqueous alteration in the carbonaceous chondrites, re-productive experimental approaches using chondritic materials were not really examined. Here, in order to elucidate the actual behavior of chondrite toward aqueous fluid, we report hydrothermal alteration experiments of Allende meteorites.

Methods

We use Allende CV3 carbonaceous chondrites as starting materials. We cut out block-shaped Allende meteorites (2.5 mm × 2.5 mm × 6 mm) by diamond blades, and never crushed ones in order to examine micro-textures before and after aqueous alteration experiments. All blocks were preliminary observed by an SEM, and blocks containing extremely large chondrules, dark inclusions and fusion clasts were not used as starting materials. Hydrothermal alteration experiments were performed with PTFE double-vessels (1 ml and 25 ml) loaded into a steel autoclave. To maintain a reducing ambient during experiments, H_2 gas was generated in an outer vessel (25 ml) by reaction with HCl solution and magnesium metal, while the starting materials (Allende block and reaction solution) were set in a small (1ml) inner vessel with screw-on lid. All hydro-thermal experiments were carried out at 200 degree C and pressure of liquid-vapor saturation (~15 bar) for 168 hours. Four different pH solutions (7.0, 8.5, 10.0, 14.0) were reacted with the Allende block with three different water/rock (W/R) ratios (0.5, 2.0, 8.0 vol./vol.), and total 12 runs were performed. After the recovered samples dried for 24 hours in an oven maintained at 60 degree C, these were analyzed by a scanning electron microscope (SEM) equipped with an energy-dispersive X-ray spectrometer (EDS), synchrotron X-ray diffraction (XRD), and a transmission electron microscope (TEM).

Results and Discussion

Most of recovered sample blocks retained their original shapes, and no brown or reddish coloration on their surface was developed. A variety of alteration phases (e.g., magnetite, anhydrite and calcite) have been precipitated on the surface, which are ubiquitously distributed regardless of substrates. SEM observations of the inner texture showed that, in all 12 runs, serpentine formed as a main secondary phase at interstitial space of olivine grains in the matrices. Smectite coexists with serpentine only in low W/R (0.5) and strong alkaline (pH 14) condition. A silica-rich amorphous material is observed in low W/R (0.5) and neutral solutions (pH 7) condition. From analyses of SR-XRD and image processing of SEM, these phyllosilicates are generally more abundant as pH value or W/R ratio increases. TEM observations show serpentine and smectite are topotaxially crystallized on the surface of olivine grains, with the crystallographic relation of (010)olivine parallel to (001)serpentine and (001)smectite. The present study indicate that i) W/R ratios affect the mineral assemblage and abundance of secondary phases and ii) hydrated mineral formation under a reducing ambient easily proceeds (even in neutral solution) compared with the previous studies.

Keywords: carbonaceous chondrite, hydrothermal experiment, Allende, phyllosilicate, serpentine

Characterization of aqueous alteration of the C-complex asteroids by reflectance spectra of carbonaceous chondrites

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C-complex asteroids are a key object to elucidate the evolution of volatile materials in the solar system, because they are enriched in volatiles and most abundant asteroids. Therefore, we need to investigate the mineralogy and the degree of aqueous alteration of the C-complex asteroids using reflectance spectroscopy. Each absorption band observed in reflectance spectra of C-complex asteroids should be characterized and assigned based on reflectance spectra of carbonaceous chondrites which are thought to derive from the C-complex asteroids (e.g. Beck et al. 2010, Takir et al. 2013). Therefore, we need to measure reflectance spectra of hydrated carbonaceous chondrites whose mineralogy and petrology are well characterized. In this study, we measured reflectance spectra of 16 CM carbonaceous chondrites under vacuum (2hPa) using a FT-IR (Fourier Transform Infrared) spectrometer and investigated relationships between features of reflectance spectra and mineralogical properties, especially between the degree of aqueous alteration and properties of $3\mu\text{m}$ and $0.7\mu\text{m}$ absorption bands that are related to hydrous minerals. All CM chondrite samples were characterized in detail by synchrotron X-ray diffraction and electron microscopy (Nakata et al. 2014).

There is a weak correlation between the $3\mu\text{m}$ -band absorption position and subtype of the meteorites that expresses the alteration degrees (Rubin et al. 2007). Samples with the absorption position at shorter wavelengths are more altered, while those with the absorption position at longer wavelengths are less altered. This trend is consistent with previous studies (Beck et al. 2010, Takir et al. 2013). However, samples with the absorption position at middle wavelengths show a wide range of alteration degrees. Therefore, it is the case that the absorption position at shortest or longest wavelengths corresponds to greatest or least alteration, respectively. In addition, Fe/Mg ratios of matrix phyllosilicates and the $3\mu\text{m}$ -band absorption position show a weak correlation. The absorption position shifts from long to short wavelengths with decreasing Fe contents.

On the other hand, $0.7\mu\text{m}$ -band depth shows no correlation to subtype and thus the $0.7\mu\text{m}$ -band properties seem to have no relation to the degrees of aqueous alteration. The $0.7\mu\text{m}$ -band depth also shows no correlation to Fe/Mg ratio of matrix phyllosilicates. Since the $0.7\mu\text{m}$ band is attributed to Fe^{3+} and Fe^{2+} charge transfer in phyllosilicates, the lack of the correlation suggests no clear relation between the alteration degrees and $\text{Fe}^{3+}/\text{Fe}^{2+}$ ratios of matrix phyllosilicates

In this study, we investigated relationships between $3\mu\text{m}$ and $0.7\mu\text{m}$ band properties and the mineralogy of CM carbonaceous chondrites, and we show that it is possible to characterize to some extent the degrees of aqueous alteration of C-complex asteroids using reflectance spectroscopy.

Keywords: aqueous alteration, C-complex asteroids, reflectance spectra

Experimental reproduction of textures of radial pyroxene chondrules by a gas-jet levitation

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Chondrules, igneous silicate spherules, formed by transient heating and rapid cooling, are ubiquitously contained in primitive chondrites. But, the formation conditions of chondrules in the protoplanetary disks are still unclear. In order to constrain the formation conditions of chondrules, a number of reproduction experiments in a electric furnace have been performed (e.g., Tsuchiyama et al., 1980). In these experiments, a melt droplet sample was hung on a platinum wire and thus the melt did contact with a platinum loop wire, possibly leading to heterogeneous nucleation near equilibrium temperatures. While, Tsukamoto et al. (2000) started a non-contact method, namely acoustic levitation method and microgravity levitation for the first time for chondrule synthesis, followed by Nagashima et al. (2006) who employed a gas jet levitation method. In both cases, space environment was simulated because crystallization of a melting silicate sphere occurs in a levitated condition.

In this study, we performed experiments to reproduce the textures of radial pyroxene chondrules using the gas jet levitation furnace used in Nagashima et al. (2006). Samples are 1-2mm spheres and chemical compositions of the samples are similar as of the natural radial pyroxene chondrules excluding iron. The sample was completely melted at about 1600-1800 °C with a 100W CO₂ laser and cooling rate after heating was about 10⁴-10⁶K / hr. We used an argon gas in order to levitate samples. After the experiment, the samples were analyzed by optical microscope and scanning electron microscope. Three dimensional images of the internal texture were also obtained after crystallization using X-ray computed tomography at SPring-8 (BL20B2).

The textures similar to natural radial pyroxene chondrules were reproduced at the cooling rates of about 10⁴K/hr. At faster cooling rates (10⁵-10⁶K/hr), samples became transparent glass without any crystals. These glass chondrules rarely exist in nature. The cooling rates (~10⁴K/hr) that successfully reproduced chondrules are slower than the calculated cooling rate (10⁶K/hr) of the melt whose temperature decreases by radiation alone in vacuum. When the nucleation center of the radial texture was observed by high magnification optical microscopy, there in most cases exists a tiny particle that is different from the radial pyroxene. This implies the importance of the heterogeneous nucleation from the tiny particles, which might be formed prior to the formation of the radial texture or impurities simply coming from dusts or the chemical reagent. These experimental data would lead to a discussion on the chondrule formation in the protoplanetary disk in the presence of gases that slowed down the cooling.

Keywords: chondrule, radial pyroxene chondrule, gas jet levitation

Chondrule shapes as indicators of shock deformation in reduced vs. oxidized CV chondrites

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CV3 chondrites are classified into oxidized and reduced subgroups based mostly on the speciation of Fe into silicates and oxides (oxidized, CV3ox) or Fe-Ni metal (reduced, CV3red)[1,2]. Both oxidized and reduced subgroups are type 3, indicating low metamorphic grade; however, the CV3ox chondrite Allende has undergone metamorphism at higher temperatures and has experienced a greater degree of recrystallization than the CV3red chondrites Leoville and Efremovka [3]. It has been proposed that the lower extent of recrystallization in Leoville and Efremovka is due to an early impact event that deformed the CV3 parent body, causing lower porosities [4] and expelling ice from the part of the parent body where Leoville and Efremovka came from [5].

To identify evidence of deformation in the CV3 chondrites, we determined modal abundances of object types (chondrules, matrix, CAIs, etc.) and shapes of chondrules in three thin sections of CV3red chondrites (two of Efremovka and one of Leoville) and in two thin sections of CV3ox chondrites (both of Allende). Mosaics of the thin sections were prepared using the following types of images: plane polarized light and reflected light from petrographic microscope; back-scattered electron (BSE) and elemental (Na, Mg, Al, Si, P, S, K, Ca, Ti, Fe among others) maps collected by EPMA (JEOL JXA-8900 at Waseda University). Grids were overlain on the mosaic images and object types were identified at each grid node; between 1200 and 1700 points were counted on each thin section. Lengths and widths of chondrules were determined for each thin section. Ratios of $(L-W)/L$, where L = chondrule length and W = chondrule width, were calculated as indicators of two-dimensional chondrule shapes. As described below, distinct lengths vs. widths were identified in the CV3red chondrites, but Allende chondrules were more equant. Orientations of the chondrule lengths in the CV3red chondrites were measured and plotted in rose diagrams to evaluate similarities in chondrule orientations.

Modes show that the CV3red chondrites have lower proportions of matrix than the two CV3ox thin sections (Leoville and Efremovka matrix modes are 22-32%, compared to 34-40 mode % matrix in the two Allende thin sections) (also see [6]). Ratios of $(L-W)/L$ tend to be near 0.1 in the chondrules from Allende, indicating circular shapes in the plane of the thin section. In contrast, $(L-W)/L$ ratios in the Leoville and Efremovka thin sections tend to be near 0.4-0.5, indicating more elongate shapes. Chondrules in one Efremovka thin section are slightly more equant than in the other two CV3red thin sections (most $[L-W]/L$ values 0.25-0.40 vs. 0.35-0.50), probably as a result of orientation of the thin section vs. fabric in Efremovka. Furthermore, rose diagrams of long axes of chondrules show that the chondrule long axes are clustered together along a common orientation in each thin section of Leoville and Efremovka.

Porous matrix would compress more easily than chondrules and CAIs, so the low abundances of matrix in the CV3red chondrites could be explained by compression during shock. Such compression of matrix is consistent with the low porosity of CV3red chondrites [4]. The elongation of chondrules along a common orientation also could result from shock deformation. Therefore, the modes, and chondrule shapes and orientations support the interpretation that CV3red chondrites were deformed by shock and that CV3ox chondrites were less affected by this impact event on the CV3 parent body [5].

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Keywords: CV chondrites, metamorphism, chondrules, shock deformation

Attempt to Extract and Identify Weak-magnetic and Ferro/Ferri-magnetic Grains Included in Chondrites by Magnetic Force.

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The effect of magnetic force caused by a field gradient has been commonly used to extract ferro (ferri)- magnetic materials from grain ensembles. The extraction was realized because the forces that operated on the spontaneous magnetizations of these materials were exceedingly large compared to terrestrial gravity. Chondrites are formed from ensemble of grains with different chemical composition, and the origin of grains is considered to be heterogeneous. At an initial stage of investigating this type of sample, it is desirable to extract and identify the material of individual particles included in the ensemble. Such method should be non- destructive and easily performed; it should be conducted by a well-established physical or chemical concept. In the case of analyzing a heterogeneous organic solution, the process of extraction and identification has been well established by introducing the technique of chromatography; such method has not been established as yet on a mixture of heterogeneous solid-grains.

A principle was proposed to identify the grain efficiently, which was driven by magnetic volume force[1][2]. When an ensemble of heterogeneous grains is released at a point located in a monotonously decreasing field, the grains are expected to be separated into multiple groups of ensembles as they translate toward an area of $B = 0$. Each ensemble is consisted by a single material, which is identified by comparing the measured χ with a list of published values.

Here we extend the above-mentioned method on paramagnetic silicate grains that compose the chondrites by extracting and identifying the individual grains by the difference of their Fe mol %. The concentration of Fe is considered as an indicator to estimate the extent of evaporation and condensation in the course of individual grain formation. The size of particles was about 100-800 μm . Microgravity was generated by a compact drop shaft system having length of the shaft was 1.8m; duration of microgravity time was about 0.5 second. The experimental apparatus was set inside a rectangle box which had a size of 30 x 30 x 20 cm. Interior of the box consisted of a vacuum chamber equipped with an electric actuator, a sample holder controller, a magnetic circuit, a battery, and a high-vision video camera. The compact setup was realized by introducing a magnetic-circuit that was composed of small NdFeB permanent magnets. Maximum field intensity of the circuit was 0.7 T. The result achieved here on sub-mm-sized diamagnetic grain provides a technical basis to extend the analysis on micron-sized grains that compose the chondrites. It may be applied to search new types of pre-solar grains that are not identified as yet.

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Keywords: translational motion, microgravity, magnetic separation, magnetic extraction, nondestructive identification

A Partial Melting Study of an Ordinary Chondrite Composition with Application to the Felsic Asteroidal Crust Formation

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Melting experiments of a synthesized, alkali-bearing, H-chondrite composition were conducted at ambient pressure with three distinct oxygen fugacity conditions (IW-1, IW and IW+2). Oxygen fugacity conditions significantly influence the compositions of partial melts. Partial melts at IW-1 are distinctly enriched in SiO₂ relative to those of IW and IW+2 melts. The silica-enriched, reduced (IW-1) melts are characterized by high alkali contents and have silica-oversaturated compositions. In contrast, the silica-depleted, oxidized (?IW) melts, which are also enriched in alkali contents, have distinctly silica-undersaturated compositions. These experimental results suggest that alkali-rich, felsic, asteroidal crusts as represented by paired achondrites Graves Nunataks 06128 and 06129 should originate from a low-degree, relatively reduced partial melt from a parent body having near-chondritic compositions. Based on recent chronological constraints and numerical considerations as well as our experimental results, we propose that such felsic magmatism should have occurred in a parent body that is smaller in size and commenced accreting later than those highly differentiated asteroids having basaltic crusts and metallic cores.

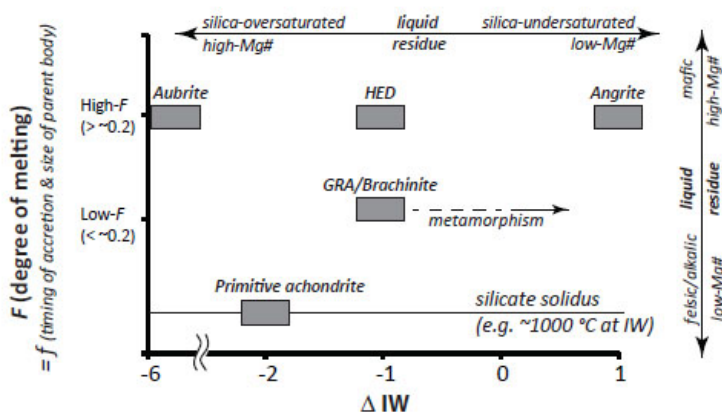
Figure caption:

Summary of magmatic conditions of achondrite suites in terms of degree of melting and fO₂ (relative to IW). See [1] for details.

Reference:

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Keywords: asteroid, crust, ordinary chondrite



High-pressure polymorphs in Type 7 and 5 ordinary chondrites

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Most ordinary chondrites experienced shock metamorphism on their parent-bodies. Many kinds of high-pressure polymorphs occur in shocked ordinary chondrites, and their impact pressure conditions have been estimated based on a phase equilibrium diagram. Ordinary chondrite is classified into petrologic Type 3 to Type 7 based on the degree of thermal metamorphism. The parent-body of ordinary chondrite is expected to consist of Type 7, Type 6, Type 5, Type 4 and Type 3 from inner to outer portions based on Onion shell model. Many previous studies have tried to estimate impact pressure conditions recorded in Type 6 ordinary chondrite using high-pressure polymorphs. However, other Type ordinary chondrite such as Type 5 and 7 have been hardly investigated so far. A systematic investigation using all petrologic Types is required to reveal an impact history on the parent-body of ordinary chondrite. In this study, we tried to estimate impact pressure conditions based on high-pressure polymorphs in Type 7 and Type 5 ordinary chondrites.

Asuka 880844 H7, Asuka 880933 LL7, Yamato 790120 H7, Yamato 790960 H7, Jilin H5 and Bassikounou H5 ordinary chondrites are obtained for this study. Preliminary optical microscopic observations reveal that all samples except for Yamato 790120 H7 and Yamato 790960 H7 include shock-melt veins. So, we focused our investigations on Asuka 880844 H7, Asuka 880933 LL7, Jilin H5 and Bassikounou H5 samples. Both Type 5 and Type 7 consist mainly of olivine, Low-Ca pyroxene, plagioclase, nickel-iron alloy and troilite. Although shock-melt veins occur in all samples, high-pressure polymorphs were not identified. We expected that feldspars in and around the shock-melt veins become amorphous (maskelynite). However, Raman analyses indicate that only a few feldspar become maskelynite. It is expected that high-pressure and -temperature conditions were achieved during a dynamic event, and high-pressure polymorphs would be formed in and around the shock-melt veins once in Type 7 at that time. However, the high-pressure polymorphs would be back-transformed into their low-pressure polymorphs again because high-temperature condition was retained even if after the dynamic event. Accordingly, it is difficult to estimate shock pressure conditions recorded in Type 7 based on high-pressure polymorphs. We used only H type ordinary chondrite as a Type 5 sample. It is inferred that the parent-body of H type ordinary chondrite may be smaller in size compared to L and LL Type. The duration of high-pressure condition during a dynamic event depends on the size of an asteroid. It is likely that the duration of high-pressure condition on H type ordinary chondrite parent-body is very short so that a high-pressure polymorph could not form, implying that few high-pressure polymorphs form in H5 ordinary chondrite

Keywords: ordinary chondrite, high-pressure polymorph, Type 7, Type 5

Effect of pressure loading path on PDFs orientation of planar deformation features (PDFs) in shocked quartz.

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Shocked quartz is the most widely known and convincing evidence of asteroid impact. It is characterized by presence of amorphous shock metamorphic microstructures, i.e., planar deformation features (PDFs). The PDFs are detectable by the optical microscope as thin straight parallel lamellae, spacing less than 10 μm , commonly orientated parallel to rational crystallographic planes of low Miller indices such as, (0001), {101n}, {112n}, {1122} planes. Previous shock recovery experiments show that the crystallographic orientations of PDFs (e.g., {1012}, {1013}, {1122} orientation) are formed at different shock pressure [Horz et al., 1968; Langenhorst & Deutch, 1994]. Therefore, the distribution and frequency of PDFs orientations of shocked quartz can be used to estimate average shock pressure, which is one of the most crucial parameters to constrain impact process and mechanism on the Earth and other solid bodies in the solar system.

The mechanism for PDFs formation is considered as local amorphization caused by the lattice collapse, on the shock front during shock wave passing through the quartz crystal [Goltrant et al., 1992]. The elastic instabilities in the shear modulus of specific planes occur with shock intensity. Therefore, they should be sensitive to the pressure loading path especially to the most intense first shock.

However, previous pressure calibration schemes are based on compilation of different shock experiments with different shock loading path (i.e., single shock method and reverberation method) [e.g., Stoffer & Langenhorst, 1994]. In this study, we therefore conducted a series of shock recovery experiments in order to clarify a characteristic features of PDFs for different pressure and different loading path.

The shock recovery experiments were conducted in the National Institute for Material Science (NIMS) with a one stage propellant gun. Start materials are natural and synthetic quartz crystals. The velocity range of flyer plate was 0.5 to 1.8 km/s, which produces peak shock pressure from 5 to 40 GPa for reverberation samples, and from 5 to 25 GPa for single shock samples, based on the impedance matching method. Recovered samples were mounted on epoxy resin. Orientations of PDFs were measured with 4-axis universal stage (U-stage) microscope.

In our experiments, shocked quartz grains show PDFs under the shock pressure over 10 GPa. At pressure above 30 GPa, grains are almost transformed to totally amorphous glass, but still remains their original crystal shapes (diaplectic glass). We will compare the characteristic features of PDFs orientation distribution for quartz grains shocked both by single shock and reverberation methods, and also discuss the sensitivity to the two different pressure loading paths, and revisit the previous pressure calibration schemes.

Keywords: shock metamorphism, shocked quartz, planar deformation features, shock recovery experiment

Traces of water in lunar meteorite

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Based on the Apollo mission, the lunar rocks from the Moon's surface to interior were considered to be anhydrous because of absence of hydrous minerals, lack of aqueous alteration effect evidence, and very low water contents below 1 ppb [1]. However, recent remote sensing observations, e.g. Chandrayaan-1, revealed that there exists molecular water and hydroxyl groups on the lunar surface by the refraction spectroscopy [2]. Furthermore, hydroxyl water was detected in volcanic glasses in lunar basalt at 46 ppm [3] and plagioclase in anorthosite at 5 to 6.4 ppm [4]. Previous study also founds hydroxyl-rich apatite (0.7 to 1.7 wt.%) in lunar meteorite NWA2977 [5]. These investigations had demonstrated that the water (or hydroxyl) contents on lunar surface are more abundant than expected previously, but there is still little evidence of water in the lunar interior. In this study, gabbroic and basaltic breccia lunar meteorites that have clasts interpreted as products of crystallization of late-stage magmatic residual liquids were studied using an electron microscope and spectroscopic analyses for the evaluation of their mineralogy and water contents.

Electron microscopic observation revealed that the lunar meteorites consist of gabbroic and basaltic clasts in their breccia matrix. The basaltic clasts contain quartz grains up to a several tens of micrometers in length. The quartz was crystallized above 40 MPa under a dry condition, which is corresponding to the depth below 10 km from the lunar surface [6]. It suggests that the lunar meteorites in the present study were originally formed in the lunar interior. As a result of synchrotron XRD analysis and Raman spectroscopy, the silica grains mainly consisting of moganite (monoclinic silica polymorphs) were observed in the breccia matrix, where the moganite content reaches >90 wt.% based on Raman spectroscopic methods [7-8]. These moganite grains were also present in shock veins, and they were surrounded by high-pressure polymorphs of silica, coesite and stishovite, implying that the moganite grains originally formed in the Moon, irrespective of terrestrial weathering nor back-transformation during an impact event. Terrestrial microcrystalline silica with >20 wt.% moganite content are known to be precipitated from alkaline fluid as evaporite [9]. Microcrystalline calcite also coexists with moganite in the lunar meteorites, and they possess traces of foaming in shock veins and close to fusion crusts with the melt glasses, implying their formations in the Moon. These facts suggest that a significant amount of water was present in the original magma in the lunar interior, where fluids probably assisted chemical/physical evolution of lunar inner materials. For quantitative estimation of water content in the magma, we will perform infrared spectroscopy for the constituent minerals such as olivine, pyroxene and feldspar in gabbroic and basaltic clasts of the lunar meteorites.

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Insight into the thermochemical state of the Jovian core from ab initio thermodynamic integration MD

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The thermochemical state of the Jovian rocky core still remains unclear. Although the rocky components of the cores in gas giants are likely to be MgO, SiO₂ and Fe (Guillot, 1999), reported melting condition are still limited up to ~500-1500 GPa (Boates and Bonev, 2013; Millot *et al.*, 2015; Bouchet *et al.*, 2013), which are far lower than the Jovian core P,T condition (4 TPa and 20 kK) estimated based on the equations of state of gas materials determined by ab initio calculations (Nettelmann *et al.*, 2012), primarily due to experimental difficulty. Thermochemical state (e.g., solid or liquid) of these materials might influence the tidal dissipation of planets and the mixing of the core and envelope. In order to clarify the phase equilibria, we have developed a new technique for calculating free energies of liquid and crystalline states based on the thermodynamic integration method (Frenkel and Smit, 2001) combined with the ab initio molecular dynamics method.

We obtained the following new findings: (1) calculated density of the MgO-SiO₂-Fe mixture is found highly consistent with the modeled Jovian core density, (2) although the melting temperatures of MgO and SiO₂ are higher than the modeled Jovian core temperature, the eutectic temperature is lower. These could serve for more detailed modeling of Jovian interior.

Keywords: ab initio calculation, thermodynamic integration method, Jovian core

Investigation of organic matter in the matrix of Allende meteorite using FIB and compact STXM

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Allende meteorite (Allende), which classed as carbonaceous meteorite (CV3), fell in Pueblito de Allende, Chihuahua State, Mexico, on February 8, 1969. Allende is composed of chondrule, Calcium-aluminium-rich inclusions (CAIs), Amoeboid olivine aggregates (AOAs), Fe/Ni metal, and matrix. Organic matters (OM) is contained in matrix phase, however, distribution of OM in the Allende at the micrometer-scale has not investigated yet.

In this study, we analyzed a focused ion beam (FIB) milled thin section (approximately 150 nm) of Allende transected a region of fine-grained matrix employing carbon X-ray absorption near-edge structure (C-XANES) spectroscopy using compact scanning transmission X-ray microscope (cSTXM), which is installed to BL-13A at Photon Factory, high energy acceleration research organization. The cSTXM analysis revealed that the thin section of matrix of Allende contains c-rich grains, the C-XANES spectra were obtained from eight of these grains of which are similar to Allende's insoluble organic matter (IOM) reported by Cody et al. (2008). Some of eight c-rich grains has exciton peak of the diamond (288.6-288.9 eV). Existence of this peak indicates the possibility that nano-diamonds were contained in these grains. In addition, one of the c-rich grains has possibility of contain of C-N bond (nitrile or heterocyclic imine). As to diffuse OM, distributing like vein, whose C-XANES were carboxylic rich OM compared to grains. Diffuse OM was investigated in CM and CI chondrites by C-XANES, whose compositions were mainly labile and low molecules OM and which was carboxylic rich OM was reported by Le Guillou et al. (2014). However, C-XANES of diffuse OM, was investigated in this study, was different from already reported. This difference caused from the difference of empirical temperature between CV, CI, and CM.

From these results, OM was existed in matrix phase as small grain and diffuse OM. And cSTXM measurement was revealed that speciation of OM. Grains have IOM-like feature, and possibility of containing nano-diamonds and C-N compounds. Diffuse OM is carboxylic-rich OM. As to the future work, existence of nano-diamonds and C-N compounds should be confirmed by TEM observation and N-XANES, respectively. In addition, metamorphosis of OM between OM and inorganic minerals when thermal denaturation should be investigate using cSTXM and Raman microscope.

Keywords: Allende meteorite, Scanning Transmission X-ray Microscope, Organic matter, Focused ion beam, Carbon XANES, Characterization