Magnesium isotopic systematics of chondrules and CAIs from Allende, Murchison, Murray and Bjurbole. F.-Z. Teng[1,2], M. Wadhwa[1,†], P. E. Janney[1,†], L. Grossman[2], S. Simon[2], and N. Dauphas[2] [1]Department of Geology, The Field Museum, Chicago, IL 60605, [2]Department of the Geophysical Sciences, The University of Chicago, Chicago, IL 60637 (teng@geosci.uchicago.edu), [†]Present address: Center for Meteorite Studies, School of Earth and Space Exploration, Arizona State University, Tempe, AZ 85287.

Introduction: Several recent studies have demonstrated that high-precision Mg isotope systematics of chondrules and calcium-aluminum-rich inclusions (CAIs) can provide unique insights into the timing and conditions of the formation of these objects in the early history of the solar system [1-5]. In particular, these investigations suggest that the initial $^{26}Al/^{27}Al$ for the solar system may have been close to ~6 × 10^{-5} (rather than the canonical value of ~4.5 × 10^{-5} indicated by previous work [6]). Furthermore, these studies indicate that chondrule formation may have begun contemporaneously with CAI formation, and likely extended to a few My thereafter.

Most of these recent investigations have focused on chondrules and CAIs from the Allende meteorite, which belongs to the oxidized group of CV3 chondrites that have undergone complex secondary processing involving both low-temperature aqueous alteration and thermal metamorphism [7]. This could imply that the Al-Mg system in the Allende chondrules and CAIs was disturbed after their formation, thereby potentially complicating interpretation of the Mg isotope data.

To further investigate questions relating to the relative timing of chondrule and CAI formation and the processes affecting the mass-dependent Mg isotopic variations in these objects, we have undertaken a systematic study of Mg isotopic systematics of chondrules and CAIs from not only Allende but also two CM2 chondrites (Murchison and Murray) as well as an L4 ordinary chondrite (Bjurbole). Furthermore, since the CM2, oxidized CV3 and L4 chondrites have experienced different secondary alteration histories, comparison of the mass-dependent Mg isotopic variations in chondrules from these chondrites has the potential to inform about how aqueous and thermal alteration processes may have affected Mg isotopic systematics in these meteorites.

Samples: One CAI each from Allende (AC5-4) and Murchison (MC5-6) has been analyzed. AC5-4 is a spherical (275 μm diameter) compact Type A CAI. MC5-6 is a spinel-rich ellipsoid (480 μm × 350 μm) with pockets of melilite and fassaite. Textures of both indicate they crystallized from melts. Four chondrules from Allende, six from Murchison, three from Murray and four from Bjurbole were also studied. These chondrules show a range of textures (barred, porphyritic and radial). In addition, two olivine fragments from Murchison and two from Murray were also analyzed. Sizes of the chondrules from Allende, Murchison and Murray were in the range of ~550-900 μm, ~260-900 μm, and ~260-320 μm, respectively; those from Bjurbole were >1 mm. All objects from these chondrites were recovered by hand-picking after either gentle crushing in a boron carbide mortar (Bjurbole), or freeze-thaw disaggregation (Allende) followed by density separation (Murchison and Murray). Each CAI and chondrule sample was split, with one fraction mounted in a polished section, and the remainder reserved for bulk chemical and isotopic analyses.

Analytical Methods: Magnesium isotopic analyses were performed at the Isotope Geochemistry Laboratory of the Field Museum, following established procedures [8-10]. Samples were dissolved in a mixture of concentrated HF-HNO_3 (3:1) after washing and ultrasonication in Milli-Q® water. The solutions were evaporated to dryness the following day, refluxed with concentrated HNO_3 and heated until no precipitate remained, and then evaporated to dryness. The dried residue was then dissolved in 1 N HNO_3, in preparation for chromatographic separation and Al/Mg ratio analysis. Separation of Mg (with >99% yield) was achieved by multiple passes through cation exchange columns in a 1N HNO_3 medium. The total Mg procedural blank during this study was <1 ng and is negligible compared with the amount of Mg in samples (>1 μg Mg).

Purified Mg sample solutions were introduced to the Ar plasma using a Cetac Aridus® desolvating nebulizer fitted with a PFA spray chamber and 100 μL/min. micronebulizer. Samples were analyzed with a GV Isoprobe MC-ICP-MS, with ^{24}Mg, ^{25}Mg, ^{26}Mg and ^{27}Al measured simultaneously (using L3, Ax, H4 and H6 Faraday cups). A 200 ppb solution typically produced a ^{24}Mg signal of 3.5-4 × 10^{-11} A. To correct for instrumental mass bias, each sample analysis is bracketed by measurements of the DSM3 Mg isotope standard having similar signal intensity as the sample (within 10%). The mass-dependent fractionation in Mg isotopes in the sample is reported relative to the DSM3 standard in per mil units (or $\delta^{25}Mg$ and $\delta^{26}Mg$). To determine the non mass-dependent anomalies in the $^{26}Mg/^{24}Mg$ ratio due to the decay of ^{26}Al, i.e., $\Delta^{26}Mg^*$ for chondrules, the measured $^{26}Mg/^{24}Mg$ ratio was normalized to $^{25}Mg/^{24}Mg = 0.12663$ using the exponential law. For the two CAIs, the $\Delta^{26}Mg^*$ was calculated as suggested by Davis et al. [11] (i.e., using a slope of 0.514 on a plot of $\phi^{25}Mg$ vs. $\phi^{26}Mg$; see [11] for definitions of these notations). The external precision (typically for 3-4 repeat measurements) is ≤ ± 0.05‰ per amu (2SD) for mass-dependent fractionation ($\delta^{25}Mg$ and $\delta^{26}Mg$ values) and ≤ ± 0.05‰ (2SD) for the non mass-dependent $\Delta^{26}Mg^*$ values [8-10]. Al/Mg ratio measurements have so far been made for only the two CAIs.

Results and Discussion: All seventeen chondrules and four olivine grains analyzed thus far from Allende, Murchison, Murray, and Bjurbole define an array of mass-dependent variations in $\delta^{25}Mg$ and $\delta^{26}Mg$ (Fig. 1). No clearly resolvable excesses in radiogenic ^{26}Mg were detected in these samples. In contrast, the two CAIs, AC5-4 from Allende and MC5-6 from Murchison, fall to the right of the mass-dependent fractionation line shown in Fig. 1 and are characterized by $\Delta^{26}Mg^*$ values of 1.45 ± 0.11 ‰ and 0.83 ± 0.03 ‰, respectively (errors are 2SE). Model isochrons (assuming an initial $\Delta^{26}Mg^* = 0$) for AC5-4 and MC5-6 yield $^{26}Al/^{27}Al$ ratios of $(4.8 \pm 0.4) \times 10^{-5}$ and $(4.4 \pm 0.3) \times 10^{-5}$, respectively. Assuming an initial $\Delta^{26}Mg^*$ of -0.032 ± 0.004 [5] results in slightly (but not significantly) higher $^{26}Al/^{27}Al$ ratios of $(4.9 \pm 0.4) \times 10^{-5}$ and $(4.6 \pm 0.3) \times 10^{-5}$, respectively. In either case, the inferred $^{26}Al/^{27}Al$ ratios are lower than those suggested by some recent high-precision Mg isotope studies of (mostly igneous) CAIs [4,5], but are consistent with those reported by others for some unmelted CAIs and their components [12-14].

The total range in Mg mass-dependent fractionation for the chondrules analyzed here is 1.2‰ per amu, and is defined by Murchison chondrules that have $\delta^{25}Mg$ varying from –1.08 to +0.12‰. The two olivine grains from Murchison have $\delta^{25}Mg$ values (–0.30 and –0.22‰) that fall within this range. Chondrules and olivine grains from the other three meteorites define relatively narrower ranges (Fig. 1). The $\delta^{25}Mg$ values in Allende chondrules vary from –0.34 to 0.00‰ and fall within the range defined by previous studies [1-3]. Murray chondrules have $\delta^{25}Mg$ ranging from –0.35 to –0.16‰; the two olivines from this meteorite have $\delta^{25}Mg$ ~ –0.62 and –0.23‰. The four chondrules from the Bjurbole L4 chondrite show the narrowest ranges in $\delta^{25}Mg$ and $\delta^{26}Mg$, with average values of –0.36 ± 0.12‰ (2SD) and –0.68 ± 0.23‰ (2SD), respectively. Mg isotope compositions of three Bjurbole chondrules reported by [15] also fall within a relatively narrow range (which overlaps with that reported here). The relatively uniform Mg isotope compositions found for Bjurbole chondrules are consistent with equilibration during thermal metamorphism, as might be expected for a Type 4 ordinary chondrite.

The chondrule data in Fig. 1 define a slope of 0.502 ± 0.013 (2σ). This slope, which is defined primarily by Murchison chondrules, is consistent with the mass-dependent fractionation in these chondrules as having resulted from a kinetic fractionation process (expected slope for which is 0.510, compared to 0.520 for an equilibrium process). As suggested for Allende chondrules [4], the range of mass-dependent fractionation in chondrules of Murchison and Murray is likely to have resulted from mixing of chondrule precursor materials (which may have been affected by kinetic fractionation processes such as evaporation and recondensation) and secondary alteration processes.

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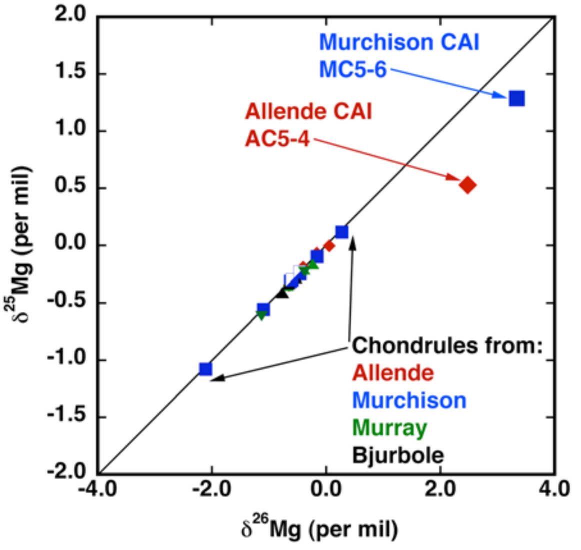

Figure 1. $\delta^{25}Mg$ vs. $\delta^{26}Mg$ in chondrules of Allende (small red diamonds), Murchison (small blue squares), Murray (green triangles) and Bjurbole (black traingles), and in one CAI each from Allende (AC5-4; large red diamond) and Murchison (MC5-6; large blue square); data for olivine grains from Murchison (half-filled blue squares) and Murray (inverted green triangles) are also shown. Errors (2SE) are smaller than the symbols. Solid line is the best-fit line to the chondrule data and defines a slope of 0.502 ± 0.013.