

Mg isotope constraints on the origin of Mg-rich olivines and mesostasis phases from Allende chondrules.

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Introduction: Reduced type I chondrules, that represent up to 95% of all chondrules from carbonaceous chondrites, are made of Mg-rich olivines (for the type IA), metal, variable fractions of glassy mesostasis, low-Ca pyroxene and other minor phases. These components are high-temperature phases but their origins remain unclear. A large fraction of Mg-rich olivines can be demonstrated on the basis of their chemical and oxygen isotopic compositions to be relict phases that partially re-equilibrated with the mesostasis during the last melting event underwent by the chondrules^[1-3]. Previous bulk Mg isotopic studies showed that individual chondrules in carbonaceous chondrites exhibited mass-dependent isotopic variations larger than 1000 ppm per amu^[4-6]. Despite these variations can be expected to result from evaporation and/or condensation during chondrule formation, the systematics that these processes have imprinted, at micron scale, on the Mg contents, the Mg mass-dependent and non mass-dependent isotopic variations, have never been explored. Here we present an in-situ Mg isotopic study of 20 Allende chondrules analyzed by LA-MC-ICP-MS, aiming at better constraining the origin of Allende chondrules.

Methods: A laser ablation system was coupled with a Thermo Fisher Neptune MC-ICP-MS to analyze in-situ Mg isotopic composition at a spatial resolution of 40-50 μm . This approach was preferred to MC-SIMS to better control matrix effects on Mg instrumental isotopic fractionation^[7]. The use of a HelEx sample cell improves the delivery efficiency of ablated aerosols. On MC-ICP-MS, four faraday cups are aligned to simultaneously measure intensities of $^{24}\text{Mg}^+$, $^{25}\text{Mg}^+$, $^{26}\text{Mg}^+$ and $^{27}\text{Al}^+$. Peak jumping from central mass 25 to 23 was conducted to monitor $^{44}\text{Ca}^{2+}/^{24}\text{Mg}^+$ each four cycles. A set of synthetic olivines and basalt glasses that have been chemically and isotopically characterized were used to correct matrix effects on the measured Mg isotopic ratios, to calibrate $^{27}\text{Al}/^{24}\text{Mg}$ measurements and to determine the instrumental $^{44}\text{Ca}^{2+}/^{48}\text{Ca}^{2+}$ ratio used for the correction of the $^{48}\text{Ca}^{2+}$ interference on $^{24}\text{Mg}^+$. The chemical compositions of the sections of Allende studied were mapped by SEM before laser ablation. This approach enabled us to measure $^{27}\text{Al}/^{24}\text{Mg}$ ratios together with mass-dependent ($\delta^{25}\text{Mg}$) and non mass-dependent (^{26}Mg excesses noted $\delta^{26}\text{Mg}^*$) Mg isotopic variations in

chondrules. We have precisions of $\pm 0.15\%$ for $\delta^{25}\text{Mg}$ and $\pm 0.03\%$ for $\delta^{26}\text{Mg}^*$ on San Carlos olivines.

Results: The Mg-rich olivines in Allende chondrules tend to have slightly negative $\delta^{25}\text{Mg}$ values, while the ferrous olivines and mesostasis phases have positive values. This is also observed in some cases within individual Mg-rich chondrules in CM2 chondrites^[5]. Mg-rich olivines show weak but detectable ^{26}Mg deficits (up to a few tens of ppm) relative to bulk Allende, while ^{26}Mg excesses are present in the mesostasis phases. Correlations of these excesses with $^{27}\text{Al}/^{24}\text{Mg}$ ratios define $^{26}\text{Al}-^{26}\text{Mg}$ isochrones. Assuming an homogeneous initial $^{26}\text{Al}/^{27}\text{Al}$ ratio of 5.23×10^{-5} at the time of formation of Ca, Al-rich inclusions (CAIs) from Allende^[8], the mesostasis in one Allende Type I chondrules formed at $1.64^{+0.19/-0.23}$ Ma after CAIs, while those of two Type II chondrules quenched at $2.23^{+0.19/-0.23}$ Ma and $2.69^{+0.27/-0.37}$ Ma, respectively.

Discussion: The $\delta^{25}\text{Mg}$ difference between Mg-rich olivines and mesostasis phases within individual chondrules suggests that the Mg-rich olivines did not crystallize from the parental melts of the mesostasis^[1-3]. The enrichment of light Mg isotopes in the Mg-rich olivines can point to fractionations due to non-equilibrium condensation^[9], or to a fractionated parental reservoir. The detectable ^{26}Mg deficits suggest that these olivines have been isolated early from the solar nebula. The heavier Mg isotopic composition of the mesostasis together with the higher Al/Mg ratios could imply fractionations due to partial Mg evaporation during chondrule formation. The timing of these chemical fractionations is constrained by the $^{26}\text{Al}/^{27}\text{Al}$ ratios of up to 1.0×10^{-5} . Modeling of evaporation is able to explain the variations in Mg contents together with $\delta^{25}\text{Mg}$ and $\delta^{26}\text{Mg}^*$ variations. These variations present within individual chondrules are new constraints on the origin of chondrules and are consistent with previous Mg data for bulk chondrules^[4-6].

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