SIMS MG ISOTOPE ANALYSES FOR METEORITES AND COMETARY SAMPLES.

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Introduction: Secondary Ion Mass Spectrometry (SIMS) has been applied to in-situ Mg isotope analyses of meteoritic Ca- Al-rich inclusions (CAIs), chondrules, as well as cometary particles to investigate early solar system chronology using ^{26}Al-^{26}Mg method [e.g. 1-3] and high temperature heating events that resulted in natural mass dependent isotope fractionation [4]. While the SIMS techniques can provide high ionization yields (≥10%), high spatial resolution (2-30 µm) and high precisions (≤0.1‰) using state-of-the-art large radius SIMS [1-3, 5], Mg isotope analyses of extraterrestrial samples often encounter difficulty in aiming the primary ion beam at target minerals and phases due to their small size, or require extraordinary long analyses time to reach sufficient precision, such as 3-11 hours per spot in comet and chondrules samples that do not show large ^{26}Mg excesses [2-3].

Improvements in Mg isotope analysis at WiscSIMS takes advantage of (1) a new high-brightness primary ion source to analyze small areas with higher ion intensities, the use of (2) three electron multipliers (EM) to detect three Mg isotope simultaneously, and (3) Faraday cup (FC) amplifiers with 10^{12} ohm resistor with low thermal noise. Our major goals are (1) to identify Mg isotope fractionation among ^{16}O-rich forsterite and enstatite particles from comet Wild 2 [6], and (2) to determine small ^{26}Mg excess in cometary particles that were never resolved before [e.g., 3].

RF Plasma Oxygen Ion Source: WiscSIMS IMS 1280 is upgraded to have radio-frequency (RF) Plasma oxygen ion source that improved the primary beam density by a factor of 5 to 20 times compared to the original Duoplasmatron (DP) ion source [7]. We use O_2^- primary ions instead of O^- ions because of a higher secondary Mg^+ ion yield (~3×) and overall better ionization efficiency (~1.4×) [5]. Typical spot sizes used for chondrule analyses [7-8] are 4-5 µm and 7 µm with intensities of 50 pA and 1 nA for plagioclase and olivine analyses, respectively.

Multi-EM Analyses of Plagioclase: With a finely focused intense primary beam the depth of SIMS analyses reaches a few µm within 1 hour. Secondary ion intensities fluctuate significantly as the pits deepen. In order to obtain Mg isotope ratios at higher precisions, three Mg isotopes were simultaneously detected using three EM on the multicollection system [7-8]. In each analysis, the high voltage (HV) applied to each EM was automatically adjusted so that changes in EM relative gains are minimized [9]. For plagioclase standard with low MgO (~0.1%), the external reproducibility of mass fractionation corrected Mg isotope ratios (δ^{26}Mg*) was better than 2‰ at ^{24}Mg intensities of 2×10^4 cps [7]. For plagioclase standard with higher MgO contents (1%), the HV of the EM detector for ^{24}Mg (≥10^5 cps) increased rapidly after each analysis (~+3V). We observed that raw δ^{25}Mg and δ^{26}Mg values increased monotonically more than 5‰ in 50 consecutive analysis, which resulted in 10‰ drop in the raw δ^{26}Mg*. It is likely that the EM pulse height distribution changed slightly with EM usage and affected the relative gains between major and minor isotopes. By using a more intense primary beam (0.1-0.2 nA; ~5 µm spot size), $^{24}Mg^+$ (≥3×10^5 cps) can be detected on a FC through amplifier board with a 10^{12} ohm resistor. In this condition, no drift in raw δ^{25}Mg and δ^{26}Mg values were observed. The external reproducibility of δ^{26}Mg* was reduced to ~0.7‰ (2SD).

In order to analyze cometary plagioclase similar to those by [3], primary beam should be reduced to 2 µm and ~20 pA. If plagioclase contains 0.4% MgO as in [3], ^{24}Mg ion intensities would be ~3×10^4 cps and the uncertainty of ^{26}Mg excess could be better than 2‰. It is a significant improvement over 3-5 ‰ in [3].

Multi-FC Analyses of Olivine with 2 µm spot size: Mg isotope analyses of olivine using primary O_2^- beam at 30 pA intensity with a spot size of 2 µm resulted in secondary $^{24}Mg^+$ and $^{25,26}Mg^+$ intensities of ~5×10^6 cps and (6-7)$\times10^5$ cps that were detected on multicollection FCs using 10^{11} ohm and 10^{12} ohm resistors, respectively. A single analysis takes 11 min. The external reproducibility of raw δ^{25}Mg, δ^{26}Mg, and δ^{26}Mg* of San Carlos olivine was 0.2‰, 0.3‰, and 0.5‰, respectively (2SD). The uncertainty of measured δ^{25}Mg is sufficiently small to identify mass dependent fractionation at ≥1‰ levels.

Application to chondrule chronology: Al-Mg isotope data from FeO-rich chondrules in Acfer 094 (ungrouped carbonaceous chondrite) were obtained using the techniques described above [8]. The inferred ages among FeO-rich chondrules show a similar range to those of FeO-poor chondrules in Acfer 094 [2], suggesting no significant time difference between the formation of FeO-rich and FeO-poor chondrules. Including data from [2], two chondrules show Al-Mg ages that are older by ~0.8 Ma compared to 15 other chondrules in Acfer 094. These chondrules have distinct oxygen isotope ratios. This might suggest that chondrules originated from a different disk region and the ~0.8 Ma age difference would correspond to the transfer time; a possible transport mechanism could be radial mixing [e.g., 10].

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