

HIGH RESOLUTION ^{26}Al - ^{26}Mg CHRONOMETRY OF CAIs FROM THE ALLENDE METEORITE S. B. Jacobsen, R. Chakrabarti, O. Bogdanovski, M.C. Ranen and M. I. Petaev, Department of Earth and Planetary Sciences, Harvard University, 20 Oxford St. Cambridge, MA 02138, USA (jacobsen@neodymium.harvard.edu).

Introduction: Most evidence for the presence of ^{26}Al ($\tau_{1/2} = 0.73$ Ma) in the early Solar System comes from the study of CAIs, with most of them apparently having an initial $^{26}\text{Al}/^{27}\text{Al}$ of $\sim 5 \times 10^{-5}$ [1-3]. The recent comparison of ^{26}Al - ^{26}Mg , ^{53}Mn - ^{53}Cr and ^{207}Pb - ^{206}Pb systems [4] seems to validate ^{26}Al as a chronometer and suggests that ^{26}Al was widely and uniformly distributed in the early Solar System and can be used as a fine-scale chronometer. This evidence suggests that (i) ^{26}Al was injected from a nearby stellar source and homogenized within the solar nebula in a short time period compared to its half-life and (ii) ^{26}Al must have been an important heat source for melting small planetesimals. The exact initial value is debated, with estimates ranging from ~ 4.5 to 7×10^{-5} [5-10].

The MC-ICPMS measurements provide the most precise data for the Al-Mg system. [5-7] suggested that most CAIs formed within an extremely short time interval of $\sim 20,000$ years. However, two research groups have obtained similarly precise isochrons for CAIs, but quite different inferred initial $^{26}\text{Al}/^{27}\text{Al}$ values of 5.8×10^{-5} and 4.9×10^{-5} [7-8]. At this stage it is not clear what is the cause of this discrepancy: sample selection, interlaboratory calibration problems, or differences in data reduction procedures. We have initiated a study of ^{26}Al - ^{26}Mg isotope systematics of CAIs by MC-ICPMS techniques aiming to resolve these problems and, in particular, to learn whether most CAIs formed over a very short time interval or not.

Samples: We selected one very large (~ 6.34 grams) potato-shaped CAI (SJ101) and several smaller CAIs from the Allende CV3 meteorite, with more CAIs to be added later. To obtain a representative “whole rock sample” of SJ101, the middle section (2.11 grams) was crushed to a fine powder, with one side of the CAI being used for a detailed petrologic and chemical study. This forsterite-bearing CAI is described in detail in an accompanying abstract [11]. Its Si isotope composition ($\delta^{29}\text{Si} = -0.30 \pm 0.05$ and $\delta^{30}\text{Si} = -0.60 \pm 0.08$) is close to the values of bulk chondrites [12], implying that this CAI experienced little or no evaporation and, as discussed below, is ideal for a chronological study. From a second ~ 1 cm irregular coarse-grained CAI (SJ103) we have extracted a small (8 mg) fragment for analysis.

Methods: The samples were dissolved with HF-HNO₃ mixture in a steel-jacketed Teflon bomb. The

spinel residue left after the initial dissolution was dissolved separately and then recombined with the main sample. Mass spectrometric measurements were carried out using a GV Isoprobe P MC-ICPMS in conjunction with a desolvating nebulizer (Elemental Scientific APEX with SPIRO). The ^{24}Mg , ^{25}Mg , ^{26}Mg and ^{27}Al ion beams were measured simultaneously in low and high mass Faraday cups (L3, Ax, H4 and H6, respectively). The $^{27}\text{Al}/^{24}\text{Mg}$ as well as the preliminary Mg isotope compositions were determined on aliquots without any chemical processing. These measurements were interspersed with our gravimetrically determined $^{27}\text{Al}/^{24}\text{Mg}$ ratio (~ 1.09) standard as well as with DSM3 [13-14] to allow sample-standard bracketing for (i) preliminary determination of $^{25}\text{Mg}/^{24}\text{Mg}$ and $^{26}\text{Mg}/^{24}\text{Mg}$ isotope ratios relative to the DSM3 standard expressed as $\delta^{25}\text{Mg}$ and $\delta^{26}\text{Mg}$ values (DSM3 \sim the bulk Silicate Earth (BSE) Mg isotope composition [13]), and (ii) precise determination of the $^{27}\text{Al}/^{24}\text{Mg}$ ratios. Then the Mg was separated from the dissolved samples by cation exchange procedures (in 0.8N HNO₃ with AG50WX12 resin) to obtain $\sim 99\%$ pure Mg ($\sim 99\%$ yield) for mass spectrometric analysis. The isotope composition of the chemically purified Mg was measured by MC-ICPMS. The mass spectrometric mass fractionation was cancelled out by the sample-standard bracketing method. The natural fractionation in a sample is then obtained with an uncertainty of ± 0.05 per mil or better.

Data Evaluation: With MC-ICPMS it is possible to obtain very high precision Mg isotope data, but it is recognized that use of this precision to obtain precise estimates of the radiogenic excess of ^{26}Mg in a particular sample may be limited because of insufficient understanding of the fractionation laws both in the mass spectrometer measurements as well as in nature [15]. The radiogenic effect due to ^{26}Al decay is represented as

$$\varepsilon_{26\text{Mg}} = \left[\frac{\left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}} \right)_{\text{sample}}}{\left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}} \right)_{\text{BSE}}} - 1 \right] \times 10^4 \quad (1)$$

where the $^{26}\text{Mg}/^{24}\text{Mg}$ ratios have been corrected for both instrumental and natural fractionation. The radiogenic effect in ε -units ($\varepsilon_{26\text{Mg}}$) can be calculated from the measured $\delta^{25}\text{Mg}$ and $\delta^{26}\text{Mg}$ values relative to the bulk Earth value:

$$\varepsilon_{26Mg} = \left\{ [1 + 10^{-3}\delta^{26}Mg][1 + 10^{-3}\delta^{25}Mg]^{-1/\rho} - 1 \right\} \times 10^4 \quad (2)$$

where ρ is a fractionation coefficient that is, in general, not precisely known. In the exponential law commonly used for correcting for mass fractionation in TIMS measurements, the value of ρ is 0.51101 [for this law $\rho = \ln(m_{25Mg}/m_{24Mg}) / \ln(m_{26Mg}/m_{24Mg})$ and the m_{iMg} are the masses for the Mg isotopes]. The raw $^{25}Mg/^{24}Mg$ and $^{26}Mg/^{24}Mg$ ratios measured with the Isoprobe-P yield a ρ value of ~ 0.52 which is substantially higher than the exponential law value and close to the equilibrium value [13]. Using our experimentally determined slope, we obtain fractionation corrected $^{26}Mg/^{24}Mg$ ratios for terrestrial samples and the standards that are essentially the same as the value of 0.139828 reported by [13-14] (when normalized to $^{25}Mg/^{24}Mg = 0.12663$). We can achieve an external reproducibility of 5 ppm for $^{26}Mg/^{24}Mg$ ratios if they are normalized this way. If, instead, we use the exponential law alone, then the reproducibility is substantially worse (10-15 ppm or worse) and we obtain a much lower $^{26}Mg/^{24}Mg$ ratio. It is clear that the exponential law is not correcting properly for mass spectrometric fractionation in the Isoprobe-P. We use the "law" determined by data obtained with this instrument. This is effectively done by first using a sample-standard bracketing method to obtain $\delta^{25}Mg$ and $\delta^{26}Mg$ values as explained above and then to apply a correction for non-instrumental (natural) mass fractionation to obtain a precise estimate of ε_{26Mg} (equation 2). Based on evaporation experiments an experimentally determined value of $\rho = 0.514$ was recommended by Davis et al. [15]. Since there is some ambiguity of what ρ value should be used for CAIs that have experienced substantial Mg isotopic fractionation from the average solar system value, one of our goals is to determine this value by analyzing CAIs with a large range of Mg isotope fractionations, rather than relying on the exponential law or the laboratory-determined evaporation coefficient. Until this is achieved it is best to base the chronology on samples with $\delta^{25}Mg \sim 0$.

Results: The forsterite bearing inclusion SJ101 has little or no excess ^{26}Mg . Thus, it does not appear to have had live ^{26}Al like FUN inclusions, however, it does not have the fractionation effects of such inclusions (both $\delta^{29}Si$ and $\delta^{25}Mg \sim 0$). It may thus appear to be an UN-inclusion. These data were all obtained with the low mass resolution mode (~ 500) of our Isoprobe P. This instrument has both true (for the axial channel) and pseudo (for all detectors) high mass resolution that is variable up to $\sim 10,000$. We will repeat these measurements using the high-resolution capabilities to verify that our measurements are not an artifact of unre-

solved interferences. At the meeting we will also present data for normal CAIs with the aim of having resolved the main issues about the high resolution chronology of CAIs.

References: [1] Lee T. et al. (1977) *ApJ*, 211, L107-L110. [2] Macpherson et al. (1995) *Meteoritics* 30, 365-386. [3] Hsu W. et al. (2000) *EPSL*, 182, 15-29. [4] Kita N. T. et al. (2005) *ASP Conference Series*, 341, pp.558-587. [5] Bizzarro M. et al. (2004) *Nature*, 431, 275-278. [6] Bizzarro M. et al. (2005) *Nature*, 435, 1280. [7] Thrane K. et al. (2006) *ApJ*, 646, L159-L162. [8] Jacobsen B. et al. (2007) *LPS*, XXXVIII, 1491. [9] Young E.D. et al. (2005) *Science*, 308, 223-227. [10] Cosarinsky M. et al. (2006) *LPS*, 37, 2357. [11] Petaev M.I. & Jacobsen S.B. (2007) this volume. [12] Georg R.B. et al. (2007) *Nature*, 447, 1102-1106. [13] Young E.D. & Galy A. (2004) *Reviews in mineralogy and geochemistry*, 55, 197-230. [14] Galy A. et al. (2001) *Intern. J. Mass Spectrometry* 208, 89-98. [15] Davis A.M. et al. (2005) *LPS*, 36, 2334.