

DETERMINATION OF PRIMORDIAL REFRACTORY INCLUSION COMPOSITIONS. S. B. Simon¹, L. Grossman^{1,2}, I. D. Hutcheon³, R. W. Williams³, A. Galy⁴, A. V. Fedkin¹, R. N. Clayton^{1,2}, and T. K. Mayeda². ¹Dept. of the Geophys. Sci., 5734 S. Ellis Ave; ²Enrico Fermi Inst., 5640 S. Ellis Ave., Univ. of Chicago, Chicago, IL 60637; ³Lawrence Livermore Nat. Lab., Livermore, CA 94551-0808; ⁴Dept. Earth Sci., Univ. of Cambridge, Cambridge CB2 3EQ England. (sbs8@midway.uchicago.edu)

Introduction: Both the chemical and isotopic bulk compositions of Types A (melilite-rich) and B (pyroxene-rich) refractory inclusions from CV3 chondrites suggest that they may be evaporative residues of high-temperature condensate assemblages [1]. The MgO and SiO₂ contents of these objects are generally consistent with the degrees of evaporation implied by the isotopic compositions that have been measured, but direct evidence of this process is sparse because there are few inclusions for which bulk chemical and bulk Mg and Si isotopic data exist. We are addressing this problem by obtaining these data for a suite of inclusions from Allende, Efremovka, and Leoville. Here, we report new bulk Mg isotopic compositions for inclusions whose bulk chemical compositions we have recently determined [2]. Their bulk Si and O isotopic compositions will also be measured as part of this study, allowing us to calculate their original bulk compositions based on the implied amounts of evaporative losses.

Sampling and analytical techniques: For each inclusion, 1 or 2 thin sections were prepared. Material was excavated from the remaining portions, ground into a homogeneous powder (5-200 mg, most ~40 mg) and divided into aliquots for instrumental neutron activation analysis (INAA), bulk Mg isotopic analysis, and bulk Si and O isotopic analysis. Our modal recombination and INAA methods are described elsewhere [2]. Bulk Mg isotopic data were obtained with the IsoProbe multi-collector, magnetic sector ICP-MS at LLNL and the Nu-Plasma double focussing MC-ICP-MS at U. Cambridge. For Mg isotopic analysis, powders were dissolved in acid and insoluble residues were removed. Mg was purified on resin columns with quantitative recovery. Analytical techniques are similar to those described in [3].

Results: The Mg isotopic data are given in Table 1 and are reported relative to the DSM3 standard [4]. Analyses of 6 of 7 inclusions analyzed by both labs are within analytical uncertainty of each other, demonstrating there is no significant intra-laboratory bias between LLNL and UC. The results from the two labs were averaged together for each of these samples, and the errors were mathematically propagated. For the other sample, TS32, the uncertainty given below represents the range of the two analyses. There is no clear relationship between isotopic composition and petrographic type. The inclusion with the highest F_{Mg} is TS32, a compact Type A (CTA), but other

Table 1. Mg Isotope Compositions of CAIs.

Sample	Type	F _{Mg} (‰/amu)	% Evap.
F2 (TS65)	B2	6.12±0.03	26.0±0.1
E107	B2	1.99±0.06	9.4±0.3
3537-2	B1	3.26±0.14	14.9±0.5
TS33	B1	4.01±0.04	18.0±0.2
TS34	B1	5.63±0.28	24.2±1.1
Golfball	B	1.61±0.06	7.7±0.3
TS32	CTA	8.05±0.50	32.7±1.7
E13	CTA	-1.03±0.07	—
E49	CTA	5.41±0.07	23.4±0.3
E49A	CTA	2.17±0.05	10.2±0.3
E55	CTA	1.35±0.23	6.5±1.1
E56	CTA	2.45±0.11	11.4±0.5
E62	CTA	-1.25±0.11	—

CTAs we analyzed have low F_{Mg}s (Table 1). The isotopic compositions of the three Type B1 inclusions are within the range of the Type B2 inclusions we analyzed. Thus, we cannot identify any one type as generally having undergone either more or less evaporation than the other types. Two samples have negative F_{Mg}, indicating that either the evaporative loss of Mg from these samples was overprinted by some other process, or that the nebular Mg isotopic composition was lighter than our terrestrial standard. The solar Mg isotopic composition is not known with certainty, and if bulk chondrites are representative of the nebula, it is fractionated by ~-0.5‰ relative to DSM3, which would increase our calculated evaporative losses by ~2% absolute. Isotopic heterogeneity in the nebula cannot be ruled out. For samples with positive F_{Mg}, evaporative losses were calculated using the Rayleigh equation, assuming monatomic Mg to be the evaporating species.

Discussion: We can compare the present results to estimates of Mg evaporative losses based on inclusion bulk compositions, bulk condensate compositions, and evaporation calculations. Previously [1], on a plot of SiO₂ vs MgO, we constructed contours of evaporative Mg loss based on calculated composition trends for isothermal evaporation at 1700K and

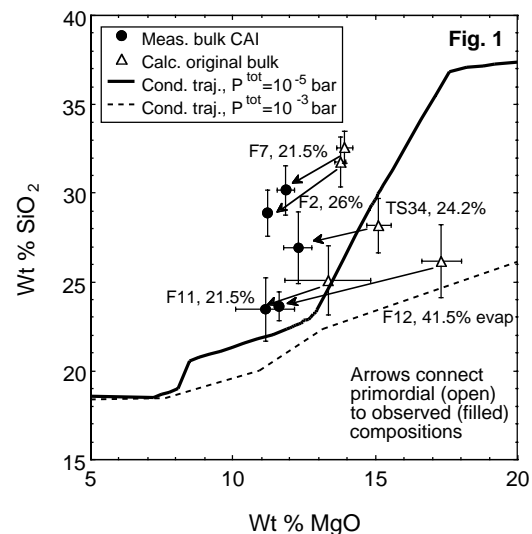
$P^{\text{tot}}=10^{-6}$ bar of various compositions that started out along a trajectory of equilibrium condensate assemblages predicted to form from a solar gas at $P^{\text{tot}}=10^{-5}$ bar. When the bulk chemical compositions of CAIs, corrected for sampling errors to the solar CaO/Al₂O₃ ratio [2] and normalized to 100% CMAS, are plotted with the contours from [1], they yield estimates of Mg loss that are greater than the losses indicated by their bulk Mg isotopic compositions (Table 1). We would expect the degree of fractionation to increase with distance from the condensation trajectory, but this is not observed. This probably simply means that the inclusions did not experience evaporation under the one set of conditions modeled and/or the precursors were not condensates from the one trajectory used in [1].

Thus far, combining our bulk chemical and isotopic data with published bulk isotopic data [5], there are five inclusions for which the bulk chemical and Mg and Si isotopic compositions are available: F2/TS65 (B2); F7/TS67 (B2); TS34/A13S4 (B1); F11/TS68 and F12/TS69 (CTAs). Calculation of their original, pre-evaporation compositions is thus possible. Starting with the observed bulk compositions corrected to the solar CaO/Al₂O₃ ratio [2] and normalized to 100% CMAS, we add the proportions of MgO and SiO₂ that the isotopic compositions indicate were lost by assuming Rayleigh fractionation and evaporation of Mg and SiO, and renormalize to obtain the original bulk compositions. Our simple model has some limitations, in that it assumes complete melting of the precursors, a single stage of evaporation, no fractional crystallization during evaporation into a vacuum, and that the precursors were isotopically normal. Incomplete melting or fractional crystallization in an inclusion would lead to preservation of relatively isotopically unfractionated material and F_{Mg} s that are lower than would be predicted from the Rayleigh equation for the fraction of ²⁴Mg remaining [6]. Another problem is that there are variations among terrestrial Mg isotopic standards [4]. Thus, published [5] Mg isotopic data (F7, F11, F12) may not be directly comparable with the data in Table 1.

The results are illustrated in Fig. 1. In spite of the above caveats, some first-order observations can be made. The original compositions are spinel-saturated and project into the melilite field of the gehlenite-anorthite-forsterite system. Had melts of the original compositions crystallized without any evaporation, they still would have had the mineralogy of CAIs. The original compositions almost certainly do not plot along a single condensation trajectory, as all except F11 define a trend in which SiO₂ and MgO are

inversely correlated. Two of the calculated original compositions (F11, TS34) plot within error of the reference condensation trajectory (solar gas, $P^{\text{tot}}=10^{-5}$ bar) we have used previously, and one other (F12) plots within error of the trajectory for a solar gas at $P^{\text{tot}}=10^{-3}$ bar. Note that the slopes of arrows connecting original and observed compositions increase with increasing initial SiO₂ content. This behavior is consistent with the hypothesis that CAIs are evaporation residues, because proportions of SiO₂ lost relative to MgO increase with increasing SiO₂ content [1].

The relationships between the original and the observed compositions can also be used to place constraints on the physico-chemical conditions that the inclusions experienced. Only certain combinations of peak temperature (T_{max}), cooling rate, P^{tot} , etc. will lead from the primordial compositions to inclusions with the observed chemical and isotopic compositions. Preliminary calculations for a $P_{\text{H}}^{\text{tot}}$ of 2×10^{-5} bar, using the method of [6], which account for fractional crystallization during evaporation, droplet radius and proportions of crystals at droplet surfaces, indicate that the precursor of F7 could have evaporated during cooling at 0.3K/hr from a T_{max} of 1630K; F2, 0.2K/hr from 1640K; TS34 at 0.9K/hr from 1720K; and F11 at 5K/hr from 1770K.



References: [1] Grossman L. et al. (2000) *GCA* 64, 2879-2894. [2] Simon S. and Grossman L. (2004) *GCA*, in review. [3] Galy A. et al. (2001) *Int. J. Mass Spectrom.* 208, 89-98. [4] Galy A. et al. (2003) *J. Anal. At. Spectrom.* 18, 1352-1356. [5] Clayton R. et al. (1988) *Phil Trans. R. Soc. Lond. A*, 325, 483-501. [6] Fedkin A. and Grossman L. (2003) *Evol. Solar Syst. Mat., NIPR*, 25-26.