Mg diffusion in melilite: constraints on the thermal history of refractory meteorite inclusions

T. LaTourrette

Department of Earth & Space Sciences, University of California, Los Angeles, CA 90095 1567, USA

The preservation of primordial Mg isotopic heterogeneity in refractory meteorite inclusions (see MacPherson et al., 1995) indicates that these inclusions have never experienced a thermal history sufficient to induce diffusive isotopic homogenization between coexisting phases. Knowledge of the Mg self diffusion rates in key minerals such as melilite and anorthite can therefore be used to place constraints on the maximum thermal history experienced by these inclusions and their parent bodies since they formed 4.56 billion years ago. In addition, the reliability of the short-lived ²⁶Al-²⁶Mg chronometer is contingent upon the assumption that Mg isotopes have not been redistributed since the in situ decay of ²⁶Al. In order to help understand the conditions under which Mg isotopic heterogeneity in refractory inclusions can be preserved, I have measured Mg diffusion coefficient in the åkermanite.

Experiments and analyses

Self diffusion coefficients for Mg were measured in synthetic åkermanite ($Ca_2MgSi_2O_7$) (Morioka and Nagasawa, 1991). Crystal sections cut perpendicular to the c-axis were polished and pre-annealed at the temperature and duration of the intended experiment.

Approximately 0.25 μ l/mm² of a solution of 1000 ppm ²⁵Mg in HNO₃ was applied to the polished surface of a crystal and dried at ~50°C, followed by firing for 1 h at 600°C to decompose the nitrate to MgO. This precipitation process typically resulted in some minor etching of the crystal surface. Charges were annealed at 1 bar in air or argon; experimental details are summarized in Table 1.

Recovered crystals were ultrasonicated in water and ethanol and then mounted in epoxy. Isotopic concentration profiles were measured by depth profiling with the Cameca ims-1270 ion microprobe at UCLA. A ~65 nA O⁻ primary beam was rastered over a 100 µm square and sputtered positive secondary ions were accelerated to 10 kV. ²⁴Mg, ²⁵Mg, ²⁶Mg, and ³⁰Si were measured at a massresolving power of ~3000-5000, which is sufficient to resolve all significant molecular interferences. A field aperture inserted in the secondary ion beam path restricted the analysed region of the sputtered crater to the central 50 µm. Sputter crater depths were measured with a Dektak surface profilometer. The depth resolution is limited by propagation of surface roughness and is estimated to be ± 10 nm. The amount of ²⁵Mg added to the surface of the crystals is too small to assume an infinite source; consequently,



FIG. 1. (a) (*left*) Measured diffusion profile from run 106 and fit to thin source equation. (b) (*right*) inverted diffusion profile and linear fit. D is determined from slope.

Run	t (hr)	Т	Atm	$\log D_{Mg}(m^2/s)$
108	28	900	air	-19.51
113	19.3	900	air	-19.57
106	16	950	air	-19.09
111	3.1	1000	Ar	-18.47
112	0.5	1100	Ar	-17.58
107	1	1100	Ar	-18.03
$D_0 (m^2/s)$				2.0×10^{-8}
E (kJ/mol)				265

TABLE 1. Experimental details

diffusion profiles were fit according to the conditions for a thin-source (e.g. Crank, 1975):

$$\mathbf{R} = \mathbf{R}_{\mathrm{xl}} + (\mathbf{R}_{\mathrm{s}} - \mathbf{R}_{\mathrm{xl}}) \exp\left[-\frac{x^2}{4\mathrm{Dt}}\right],$$

where $R = {}^{25}Mg/\Sigma Mg$, the subscripts xl and s refer to the crystal interior and surface, respectively, x is distance, D is the diffusion coefficient, and t is time. Diffusion coefficients were calculated from the slope of a plot of $-\ln[(R - R_{xl})/(R_s - R_{xl})] vs x^2$. A typical measured profile and measured fit are shown in Fig. 1a, and an inverse profile and linear fit are shown in Fig. 1b.

Results and discussion

Preliminary diffusion coefficients are listed in Table 1, and an Arrhenius plot summarizing the results is shown in Fig. 2. Two or three profiles were measured on each sample, with separate profiles agreeing to within $\pm 25\%$. Replicate experiments at 900°C also agree well, although there is a factor of 2.8 discrepancy between the two runs at 1100°C. Based on the co-linearity of the results, experiments run in air and Ar appear to be consistent with each other. The activation energy derived from the data is 265 kJ/mol, which is consistent with the results for diffusion of several other divalent cations in åkermanite determined by Morioka and Nagasawa (1991). The pre-exponential factor, however, is significantly lower.

As shown in Fig. 2, the Mg diffusion in åkermanite is about an order of magnitude slower at any temperature than Mg diffusion in anorthite as recently determined by LaTourrette and Wasserburg (1998). This contrasts with the results for O, which show that diffusion in åkermanite is about an order of magnitude faster than in anorthite (Ryerson and McKeegan, 1994). This indicates that Mg isotopic anomalies in åkermanitic melilite in refractory



FIG. 2. Arrhenius plot summarizing results for Mg diffusion in åkermanite from this study and in anorthite from LaTourrette and Wasserburg (1998).

meteorite inclusions will be more resistant to redistribution during thermal disturbances. Under isothermal conditions, melilite would need to be ~100°C hotter than anorthite to undergo the same extent of isotopic exchange. In the case of a planetesimal heated by ²⁶Al-decay, melilite should be more likely to retain its isotopic signature. Thus, the constraints on thermal histories of refractory inclusions and planetesimals determined from Mg diffusion in anorthite by LaTourrette and Wasserburg (1998) remain valid.

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