

MAGNESIUM ISOTOPIC FRACTIONATION OF FORSTERITE DURING EVAPORATION FROM DIFFERENT CRYSTALLOGRAPHIC SURFACES. M. Yamada¹, S. Tachibana¹, H. Nagahara¹ and K. Ozawa¹,

¹Department of Earth and Planetary Science, Graduate School of Science, Univ. of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan. (maho@eps.s.u-tokyo.ac.jp)

Introduction: Chemical and isotopic fractionations are expected by evaporation of solid materials under low total pressure conditions in the early solar system. Kinetic isotopic fractionation inside evaporating solid is determined by the evaporation rate (v), diffusion rate (D), size of the solid (X) and isotopic fractionation factor (α).

Forsterite (Mg_2SiO_4), one of the major minerals in meteorites, evaporates congruently [1], and the evaporation rate in vacuum shows anisotropy, which is fastest along the c-axis and slowest along the b-axis in the temperature range of 1500-1800°C [1, 2]. The different crystallographic surfaces of evaporation residues also show different microstructures [3]. The Mg self-diffusion rate in forsterite is also anisotropic, which is fastest along the c-axis and slowest along the b-axis [4]. Therefore, the isotopic fractionation factor (α) is also expected to show anisotropy. However, it has not been investigated systematically yet.

In order to investigate anisotropy of α for evaporation of forsterite, we have analyzed Mg isotopic compositions of surfaces of evaporation residues by an ion microprobe. Our analytical approach is different from that in Wang et al. [5], who measured the depth profile of isotopic compositions from the surface of an evaporation residue using ion microprobe to obtain α and D along the a-axis of forsterite.

Samples and Analytical Methods: Magnesium isotopic compositions of the surfaces of synthetic forsterite, which was cut parallel to the a-, b-, and c-axes and evaporated in vacuum at 1504-1692 °C for 1-166 hours by [3], were measured using the Cameca 6f ion microprobe at Univ. Tokyo. A 0.3nA O⁻ primary beam with 12.5 keV energy and ~20µm diameter was used for measurements. Positive secondary ²⁴Mg, ²⁵Mg and ²⁶Mg ions, accelerated at 10kV, were detected by an electron multiplier. A mass resolving power was set to 4000 to resolve ²⁴MgH from ²⁵Mg. The typical counting rate of ²⁴Mg was ~10⁵ cps.

We measured about 10 points for a surface of both the sample and a starting material in each analytical session. The reported isotopic compositions here are relative differences between the mean of ~10 measurements for the sample and that for the starting material.

Results: The Mg isotopic compositions of three crystallographic surfaces of forsterite evaporated at 1504°C, 1598 °C, and 1692 °C are summarized in Fig. 1. Almost all the data lie on the mass-dependent fractionation line, and show heavier isotopic compositions than the starting material. For the present experimental conditions (temperature and heating duration), the degree of fractionation is smallest (or largest) along the b-axis (or c-axis) of which evaporation and diffusion rates are slowest (or fastest). This implies the presence of anisotropy in isotopic fractionation as expected. Temperature effects on isotopic fractionation are also seen; samples heated at higher temperatures tend to show larger isotopic fractionations. Note that the internal diffusive transportation of Mg ions should have reached the steady state for all the directions to discuss the anisotropy and the temperature dependence (see below).

Discussion: The surface isotopic compositions change with heating duration as a function of v , α and D [5]. The evaporating surface becomes richer in heavy isotopes as evaporation proceed, and therefore the diffusive transport flux of isotopes from the surface to the interior becomes larger with enrichment of heavy isotopes and is balanced with the evaporation flux after certain time (steady state). At the steady state, evaporation proceeds with keeping the surface isotopic composition constant, where the surface composition merely depends on α . Time required to reach the steady state is dependent on the ratio of D to v^2 .

We fitted the (010) surface isotopic compositions of samples heated at 1692 °C for 1-12 hours, the (001) samples heated at 1598 °C for 24 and 48 hours and the (100) samples heated at 1504 °C for 72 and 166 hours respectively, by the diffusion-controlled evaporation model [5] using v by [2].

The estimated α for ²⁶Mg or ²⁵Mg relative to ²⁴Mg along all the directions seem to be closer to unity than the square root of the mass ratio of isotopes, suggesting that the isotopic fractionation is smaller than that expected from the kinetic theory of gases. Although more data are needed for both shorter and longer heating durations, the present data imply that α may be closest to unity along the b-axis and farthest from unity along the c-axis. Kinetic isotopic

fractionation factors, different from square root of masses of gas species, have been also shown for Mg from CMAS melt [6].

There seems to be dependence of α on temperature. For all the surfaces, α at lower temperatures appears to be closer to unity, while α at higher temperatures tends to approach the square root of the mass ratio. This implies that kinetic isotopic effects on the surface of evaporating substance become smaller at higher temperatures.

Magnesium self-diffusion coefficients that can also be estimated from data-fitting, seem to be consistent with those in previous studies [e.g., 4].

References: [1] Nagahara H. and Ozawa K. (1996) *GCA*, 60, 1445-1459. [2] Ozawa K. et al. (1996) *LPS XXVII*, 989-990. [3] Nagahara H. and Ozawa K. (1999) *Proc. Japan Acad.*, 75B, 29-34. [4] Morioka M. (1980) *GCA*, 44, 759-762. [5] Wang J. et al. (1999) *GCA*, 63, 953-966. [6] Janney P. E. et al. (2003) *LPS XXXIV*, Abstract #1940.

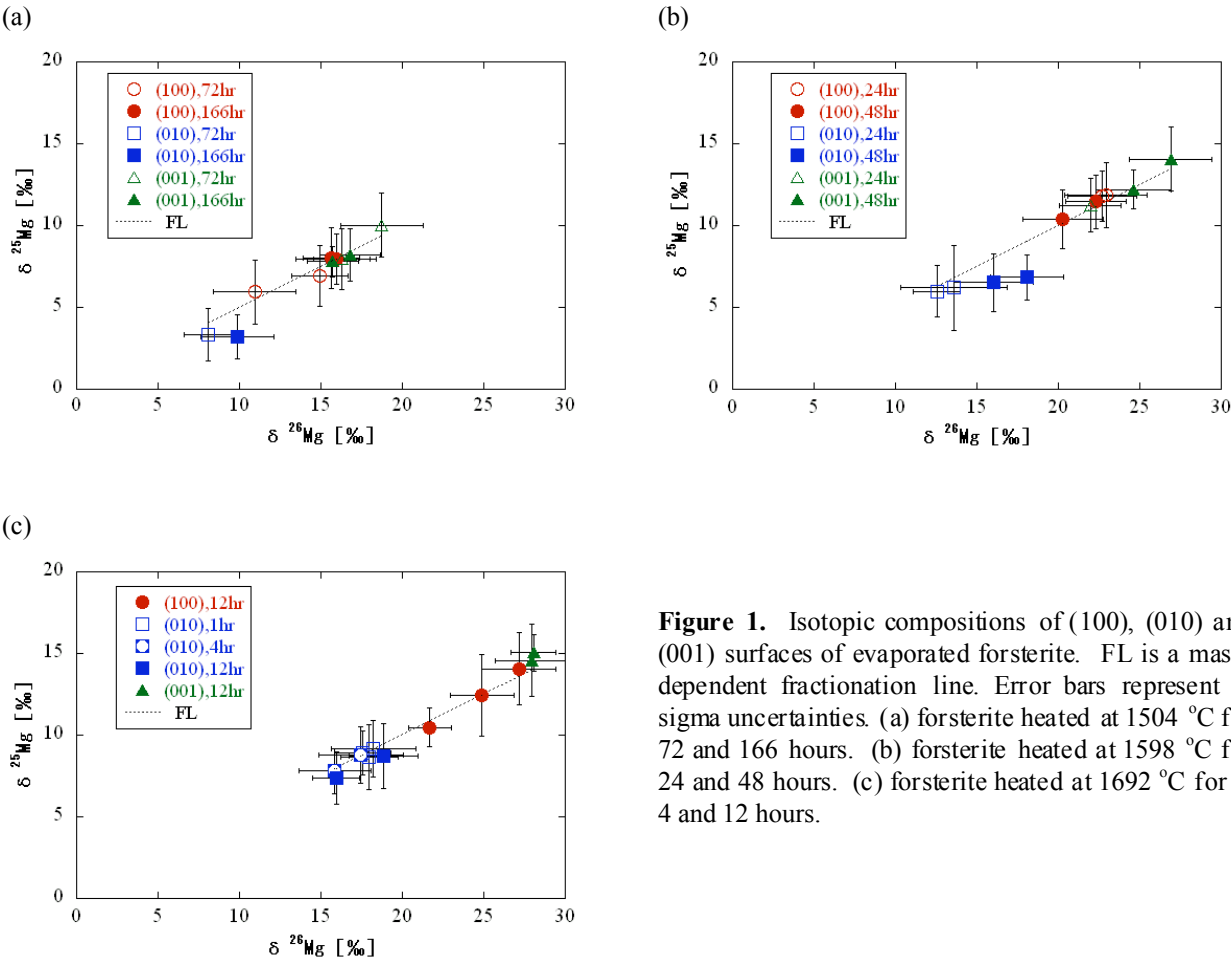


Figure 1. Isotopic compositions of (100), (010) and (001) surfaces of evaporated forsterite. FL is a mass-dependent fractionation line. Error bars represent 2-sigma uncertainties. (a) forsterite heated at 1504 °C for 72 and 166 hours. (b) forsterite heated at 1598 °C for 24 and 48 hours. (c) forsterite heated at 1692 °C for 1, 4 and 12 hours.