

Oxygen isotopic exchange between primitive amorphous silicate dust and water vapor in the protosolar disk

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Meteoritic evidences suggest that oxygen isotopic exchange between ¹⁶O-rich primitive silicate dust, which would be dominantly in amorphous form, and ¹⁶O-poor nebular water vapor occurred in the early Solar System, and the oxygen isotopic compositions of extraterrestrial dust may put constraints on disk processes. However, it has been little known about the efficiency of oxygen isotopic exchange in the disk. Here we experimentally investigate oxygen isotopic exchange between amorphous forsterite and water vapor.

A series of oxygen isotopic exchange experiments between sub-micron-sized amorphous forsterite grain (average diameter of ~80 nm) with H₂¹⁸O vapor (97% ¹⁸O) were carried out at 803–1123 K under water vapor pressure (P_{H₂O}) of 10⁻², 0.3 Pa using a gold-mirror vacuum furnace. The run products were analyzed with FT-IR (JASCO FT-IR 4200) and XRD (Rigaku SmartLab). Bulk oxygen isotopic measurements were conducted for pelletized samples by SIMS (Cameca ims-6f).

Infrared absorption peak of amorphous forsterite at ~10 micron shifted to higher wavelengths with time by annealing at H₂¹⁸O vapor of 0.3 Pa at 803–883 K, whereas such a shift was not observed for samples annealed in the presence of isotopically normal water vapor of 0.3 Pa. This peak shift is caused by the oxygen isotopic exchange with H₂¹⁸O. SIMS analyses showed that relative shifts of the 10 μm peak of amorphous forsterite correlate linearly with oxygen isotopic composition up to ¹⁸O/(¹⁸O+¹⁶O)~0.4. Time evolution of the degree of isotopic exchange with amorphous forsterite at P_{H₂O}~0.3 Pa was governed by three-dimensional spherical diffusion (Crank, 1975), yielding the diffusive exchange rate coefficient of $\ln D = -27.1 - (162.3 \pm 16.1 \text{ kJ mol}^{-1})/RT$. At 953 K and P_{H₂O} of 0.3 Pa, all typical peaks of crystalline forsterite shifted to higher wavelengths with peak broadening, which can be interpreted as simultaneous isotopic exchange and crystallization. At 1073 K and P_{H₂O}~0.3 Pa for 2 hr, infrared spectrum of a run product shows several peaks of isotopically normal crystalline forsterite in spite of heating in the presence of H₂¹⁸O. This is most likely because crystallization occurred more rapidly than isotopic exchange and a slower isotopic exchange rate for crystalline forsterite prevented the efficient isotope replacement.

At P_{H₂O} of 10⁻² Pa, the reaction rates at 883 K and 853 K were similar to each other. This little temperature dependence of the reaction rates implies that the supply of water vapor is a rate-limiting step. At 803 K, the reaction rate at P_{H₂O}~10⁻² Pa was almost identical to that at P_{H₂O}~0.3 Pa, which is explained by the diffusion-controlled isotopic exchange that should not have significant pressure dependence on water vapor pressure.

Based on the experimental results, we estimated timescales for complete isotopic exchange between amorphous forsterite (80 nm, 1 μm in diameter) in the protosolar disk. We found that the original oxygen isotopic signatures of amorphous forsterite could be preserved only if the dust was kept at temperatures lower than ~500 K within the disk lifetime of 1–10 Myr.

Keywords: amorphous silicate, dust, oxygen, isotopic exchange, protosolar disk