

## 非晶質ケイ酸塩と水蒸気との酸素同位体交換速度論から制約される始原的ケイ酸塩ダストの原始惑星系円盤での熱的プロセス

### Thermal processes of primary silicate dust in protoplanetary disk: Constraints from oxygen isotope exchange kinetics between amorphous silicate and water vapor

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Mass-independent oxygen isotope variations of extraterrestrial materials is considered to be a result of oxygen isotope exchange between <sup>16</sup>O-rich primitive materials and <sup>16</sup>O-poor gaseous reservoirs (Yurimoto & Kuramoto, 2004; Lyons & Young, 2005). Because the isotope exchange rate is controlled by the physicochemical condition of protoplanetary disks, oxygen isotopic compositions of extraterrestrial materials could be a good tracer for the disk conditions. However, little has been known about the kinetics of isotope exchange reaction for oxygen in the disks. We focused on oxygen isotope evolution of amorphous silicate dust, which is recognized as primitive material in protoplanetary disks, and investigated oxygen isotope exchange kinetics between amorphous silicates and water vapor.

A series of oxygen isotope exchange experiments between submicron-sized grains of amorphous silicate with forsterite and enstatite stoichiometry and <sup>18</sup>O-enriched water vapor (97% <sup>18</sup>O) were carried out at 803–1123 K and water vapor pressure ( $P_{\text{H}_2\text{O}}$ ) of 0.01–1 Pa using a gold-mirror vacuum furnace equipped with a gas flow system (Yamamoto & Tachibana, 2018). The run products were analyzed with FT-IR (JASCO FT-IR 4200), XRD (Rigaku SmartLab), and FE-SEM (JEOL JSM-7000F). The bulk oxygen isotope composition was determined as an average of multiple measurements for a pelletized sample by SIMS (Cameca ims-6f and -1280HR).

An infrared absorption peak of amorphous silicate at ~10 micron shifted to longer wavelengths with time by annealing with H<sub>2</sub><sup>18</sup>O vapor of 0.3 Pa at 803–883 K for amorphous forsterite and at 923–1003 K for amorphous enstatite, whereas such a shift was not observed for samples annealed in the presence of isotopically normal water vapor of 0.3 Pa. The relative peak shifts of the 10 micron feature of amorphous forsterite and enstatite correlate with their oxygen isotopic compositions, suggesting that the peak shift was caused by the isotope exchange of oxygen between amorphous silicates and H<sub>2</sub><sup>18</sup>O vapor. Temporal change of the degree of isotope exchange under these conditions was governed by diffusive isotope exchange with the isotope exchange rate of  $D \text{ (m}^2 \text{ s}^{-1}\text{)} = (1.5 \pm 1.0) \times 10^{-19} \exp[-161.5 \pm 14.1 \text{ (kJ mol}^{-1}\text{)} R^{-1} (1/T-1/1200)]$  for amorphous forsterite and  $D \text{ (m}^2 \text{ s}^{-1}\text{)} = (5.0 \pm 0.2) \times 10^{-21} \exp[-161.3 \pm 1.7 \text{ (kJ mol}^{-1}\text{)} R^{-1} (1/T-1/1200)]$  for amorphous enstatite. At  $P_{\text{H}_2\text{O}} = 0.01$  Pa, the isotope exchange reaction for amorphous forsterite is controlled by a supply of water vapor at 853 and 883 K, and the supply-controlled isotope exchange rate was evaluated. Crystallization of amorphous forsterite dominated over oxygen isotope exchange at a higher temperature of 1073 K because crystallization precedes the oxygen isotope

exchange reaction. The oxygen isotope exchange timescales for amorphous forsterite and enstatite dust under protoplanetary disk conditions suggest that the primary oxygen isotope signatures of amorphous silicate dust would be erased by thermal annealing at temperatures above 500–650 K within the lifetime of protoplanetary disks.

Cometary silicate dust collected by the Stardust mission has  $^{16}\text{O}$ -poor isotopic compositions close to the terrestrial value (McKeegan et al., 2006; Nakamura et al., 2008). If primitive silicate dust in the protosolar disk has the same  $^{16}\text{O}$ -rich composition as the Sun, this observation implies that comets accreted silicate dust that was thermally processed at temperature above  $\sim 500\text{--}650\text{ K}$  in the inner Solar System and transported to the comet formation region (e.g., Nuth, 1999; Ciesla, 2007). This could also explain that presolar silicate grains are not a major component in cometary silicates (Floss et al., 2013). The effective radial transport of inner disk materials to the outer part of protoplanetary disks may provide some constraints on the dynamics of protoplanetary disks prior to planet formation.

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