Oxygen isotope fractionation of containerless silicate melts in gravity and microgravity

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High-temperature behaviors of stable isotopes are poorly understood especially during evaporation process. Generally, evaporation has been believed to accompany with heavy isotope enrichment in the residue by negative fractionation factor between vapor and liquid obeying mass-dependent law (Davis et al., 1990). However, in the conventional experimental system, container supporting the melt or solid is always present. In order to reproduce the evaporation in space, the experiment needs to be carried out under microgravity, which can achieve vapor emission without convective flow. We conducted CO2-laser fusion experiments on some containerless silicate melts (Mg2SiO4, MgSiO3, Ca2Al2O7, and CaO-MgO-Al2O3-SiO2 melts) under: (1) 1 bar Ar and gravity by aero-acoustic levitation (AAL at ISAS) and (2) 1 bar of air and low pressure of Ar by parabolic flight (G-II at DAS). The total durations of each laser irradiation including preparation of starting materials and runs were limited within a few minutes to avoid O-isotopic exchange with atmospheric O2 and each run under microgravity were completed within ~10 sec. Recovered samples (crystalline and glassy) were half-cut and investigated chemically and O-isotopically using electron and laser microprobes.

The initial O-isotopic ratios (d18O, permil) of Mg2SiO4, MgSiO3, Ca2Al2SiO7, and CMAS are 15.4, 12.8, 8.8, and 13.6 permil, respectively. After the experiments, obtained run products all showed isotopic depletion. For AAL experiments under gravity, Mg2SiO4 residues had d18Ofin of 2.5 to 8.3 permil, showing intense negative D18O (= d18Ofin - d18Oinit) of -12.8 to -7.1 permil, respectively. Ca2Al2O7 also showed a negative D18O of -17.8 permil (= -9.0 - 8.8). For microgravity experiments under 1 bar air, Mg2SiO4, MgSiO3, and CMAS showed D18O of -3.4 permil (= +12.0 - 15.4), -4.1 to -3.4 permil (= 8.7, 9.4 - 12.8), and -3.9 permil (= 9.7 - 13.6), respectively. Different results from above two experiments suggest that negative O-isotopic fractionation may have been enhanced by gravity. In contrast, microgravity experiments on MgSiO3 under low-pressure Ar significantly showed minimized negative fractionation as D18O of -1.4 to -1.0 permil (= 11.4, 11.8 - 12.8) among the other experiments. This result emphasizes that low-pressure condition also minimize the isotope fractionation.

The results of our experiments revealed that O-isotope behaviors during melt-evaporation could be affected both by gravity and pressure conditions. Conventional experiments for evaporation under gravity always accompanies with convective emission. It is likely that less gravity enhances diffusive boundary layer because of no convective flow near the evaporating melt. In-situ video observation displayed that vapor is not emitted vertically but radially to horizontally from MgSiO3 melt surface. Consequently, reasonable model explaining the reversed isotope fractionation can be simply expressed with av-l = as-l = as-DL x aDL-l, where as-DL = Rsolid/Rdiffusion layer (above 1.0, Uyeda et al., 1991) and aDL-l = Rdiffusion layer/Rliquid = 1.0. Thus, the evaporational loss can be interpreted as condensational erosion at the layer surface. Our results further provide important information to stable isotope cosmochemistry. If the possible mechanism producing isotope fractionation is attributed to stability of diffusion layer near the melt surface, it may give a constraint to the model for actual condition in the early solar nebula producing 'chondrules' and 'Calcium-Aluminum-rich Inclusions (CAIs)' or in the early planet-formation process constructing 'asteroids', which relate to igneous process under low-pressure. Our experiments suggest that formation of low d18O materials in the space (i.e., low-P and microG) requires melt evaporating with flow. Such a condition may be likely achieved in the early solar nebula accompanying with aerodynamic drag by bipolar outflow (e.g., Shu et al., 1996).