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Kinetics of evaporation of forsterite and Fe-Mg olivine in vacuum

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Evaporation of forsterite, which plays an important role in chemical and isotope fractionation in the early solar nebula, is governed by surface kinetics strongly affected by surface conditions, such as surface roughness as well as density and orientation of dislocation outcrops. We have revealed anisotropies in evaporation rate and surface microstructure of forsterite and Fe-Mg olivine [1-3]. In order to better understand the kinetics of evaporation of forsteriete and olivine, we carried out experiments in a wider range of temperature and examined evaporation mode on the basis of temperature dependence of evaporation rates and surface microstructures.

Experiments were carried out in a vacuum chamber internally heated by W mesh heater. Starting materials are single crystals of synthetic forsterite and natural Fe-Mg olivine (Fo∼90), which are cut into crystallographically oriented rectangular parallelepipeds. The experimental temperature in the present and our previous studies ranged from 1300 to 1600 °C for Fe-Mg olivine and from 1500 to 1800 °C for forsterite. Surface microstructures of experimental residues were observed with SEM and EBSD, and face-specific evaporation rates were calculated from sample sizes and weight losses on at least three parallelepipeds with different [001]:[010]:[001] ratios. Development of Fe-Mg zoning due to preferential evaporation of Fe and Fe-Mg lattice diffusion in the sample was taken into consideration in rate estimation for olivine evaporation.

The experimental results for both forsterite and olivine experiments demonstrated systematic temperature dependence of anisotropy in evaporation rate: (010)>(001)>(010) above ∼1750°C, (001)>(100)>(010) at temperatures between ∼1750 and ∼1500°C, and (001)>(010)>(100) below ∼1500°C. The maximum anistoropy in the evaporation rate is factor of 5 below ∼1750°C, but the anisotropy is significantly suppressed above ∼1750°C, where the differences among three evaporation rates are within 70% at ∼1800°C. These crossovers in evaporation rate were intimately associated with changes in surface microstructures not directly related to surface morphologies originated from dislocation outcrops. Facets of (010) disappear on the (010) surface above ∼1750°C, and (100) facets appear on the (100) surface below ∼1500°C, which is consistently observed both for forsterite and olivine. The facets observed in SEM were confirmed to be atomistically flat consisting of stacking of layers with one or few unit-cells height through STM observations. On the contrary, non-facetted surfaces were confirmed to be atomistically rough. Therefore, the rate crossovers are attributed to rough-smooth transitions [4-5] at ∼1500°C for (100) and at ∼1750°C for (010). Such rough-smooth transition for the (001) surface is expected to exist below ∼1500°C. The anisotropy in the rough-smooth transition temperature identified for forsterite and olivine cannot be explained solely by the differences in slice energy or attachment energy (anisotropy in bond stength) for the three crystallographic faces [6], which predicts that the transition temperture decreases in the order of (010), (001), and (100).

The presence of Fe notably enhances stoichiometric evaporation of Fe-Mg olivine at 1500°C without forming any reaction product [3]. This suggests that stoichiometric evaporation from olivine or congruent evaporation from forsterite is controlled by removal of Mg2+ and Fe2+ from either the M1 or M2 site followed by spontaneous destruction of SiO4 tetrahedron at least above 1500°C, which is required not to result in incongruent evaporation forming enstatite layer. On the contrary, Fe-Mg olivine evaporates nonstoichiometrically to form enstatite at the forsterite surface at ∼1300°C (Ozawa and Nagahara, 2002), where removal of Fe2+ or Mg2+ is not the rate-controlling process but Si removal or breaking Si-O bonds governs the overall reaction. The slow removal of Si results in nonstoichiometric evaporation via reaction with olivine residue to produce enstatite on the surface. It is, therefore, expected that the order of roughening transition for (100) and (001), which is in conflict with the order of attachment energy, is attributed to the arrangement of M-O and Si-O bonds in surface layers of each orientation, either promoting or inhibiting preferential removal of Mg2+ and Fe2+.

[1] Nagahara and Ozawa (1999); [2] Nagahara and Ozawa (2000); [3] Ozawa and Nagahara, (2001); [4] Burton and Cabrera (1949); [5] Burton et al. (1951); [6] Watson et al. (1997).