

The role of back reaction on the chemical and isotopic fractionation in evaporation

Hiroko Nagahara[1], Kazuhito Ozawa[2]

[1] Geol. Inst., Univ. Tokyo, [2] ISEI

The effect of back reaction on chemical and isotopic fractionations is examined theoretically and experimentally. The calculation results show that the decoupling of chemical and isotopic fractionations is expected even if the dust enrichment factor is much smaller than unity, and is more marked for strongly volatile elements. Slow diffusion in the condensed phase further enhances the decoupling. Evaporation experiments with varying degree of back reaction were conducted by changing the size of orifice of a Knudsen cell for multi-component system of a chondrite. Comparison of the experimental results with the model calculation implies that the evaporated fraction obtained above is expected to show the largest degree of isotopic fractionation, which we are planning to measure.

Evaporation and condensation played key roles on cosmochemical fractionation took place in the solar nebula. Despite significant chemical fractionation among planets, asteroids, and meteorites regarding major and trace element concentrations, there appears to be little isotopic fractionation. Several different explanations are proposed for the decoupling between chemical and isotopic fractionations of volatile elements: (1) fractionation before formation of chondritic materials, (2) evaporation in the presence of ambient gas, and (3) evaporation of very volatile elements at large evaporation Peclet number ($Pe=Jr/D$). In this study, the role of back reaction is examined quantitatively, and experiments on evaporation of alkalis from silicate melt with various magnitudes of back reaction were conducted to evaluate the role of back reaction.

The effect of back reaction on chemical and isotopic fractionations is examined by using a model for stoichiometrically evaporating binary system. The calculation results show that the back reaction brings high degree of isotopic fractionation for volatile elements in the earlier stage and negligibly small fractionation substantially before complete evaporation, although the elements are progressively fractionated as evaporation proceeds. The decoupling is expected even if the dust enrichment factor is much smaller than unity, and is more marked for strongly volatile elements. Slow diffusion in the condensed phase further enhances the decoupling by more preferentially suppressing the isotopic fractionation.

Evaporation experiments with varying degree of back reaction were conducted by changing the size of orifice of a Knudsen cell. The silicate portion of Jilin (H5) chondrite was used in the experiment. Temperature was 1300 C and the duration was 40 minutes. The weight loss increases with increasing orifice size. The composition near the surface shows systematic variation with increasing orifice size. Na, K, and Si decrease and Ca and Al increase. The total evaporated fraction estimated by Al_2O_3 and CaO contents in the glass is up to 35%, and the evaporated fractions of Na and K increase from a few % to 70-80 % with increasing orifice size from 0.25 to 1 mm. The experimental conditions for volatile elements are characterized by large K and large Pe. Comparison of the experimental results with the model calculation implies that the evaporated fraction obtained above is expected to show the largest degree of isotopic fractionation, which we are planning to measure. Partial pressure of Na in a capsule is estimated by using the Clausing factor and the area of the orifice of the Knudsen cell. They decrease by factor of ~5 and the average evaporation rates increase by a similar factor as the orifice size increases from 0.25 to 1 mm. The open capsule shows a marked decrease in the partial pressure (by one orders of magnitude smaller than 1mm orifice), although the average evaporation rate is only slightly enhanced. Thus an increase of the partial pressure of the element considered above the threshold value is necessary to suppress chemical fractionation by evaporation.