ELEMENTAL AND ISOTOPE FRACTIONATION OF CAI-LIKE LIQUIDS BY EVAPORATION IN LOW PRESSURE H₂.

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Introduction. Laboratory determinations of the evaporation kinetics and isotopic fractionation of evaporation residues of silicate liquids exposed to vacuum or low-pressure hydrogen gas are essential for interpreting the thermal history of refractory inclusions in meteorites. The Type B CAIs are especially interesting in this regard because they are often enriched in the heavy isotopes of magnesium and silicon in a manner that is almost certainly the result of loss of magnesium and silicon by evaporation while these inclusions were molten. Richter [1] used model calculations to illustrate how a condensate from a solar composition gas subject to a particular pressure-temperature history (i.e., that due to a shock) can be translated into a specific, and quite realistic, elemental and isotopic CAI-like composition of the resulting residue. A key point is that the isotopic measurements of refractory inclusions have to be interpreted using high-precision experimental data on the evaporation kinetics and isotope fractionation during evaporation determined under conditions appropriate to those that prevailed in the early solar system. Aside from temperature, the key quantity affecting the evaporation kinetics of silicate liquids in the protoplanetary nebula is the pressure and oxygen fugacity of the surrounding gas (mainly hydrogen for a solar composition gas). Here we use laboratory data to contrast the evaporation kinetics and isotopic fractionation in vacuum and in finite hydrogen pressure.

Elemental Fractionations. For pressures less than about 10^{-7} bars H₂, the evaporation kinetics is effectively that in vacuum. For hydrogen pressures greater than 10^{-7} bars the evaporation kinetics increases in proportion to square root of the hydrogen pressure [2]. Figure 1 illustrates the effect of hydrogen pressure by comparing the evaporation rate in vacuum to that in 2×10^{-4} bars of hydrogen.

The increase in the evaporation rate when hydrogen is present is due to the effect of hydrogen on the saturation vapor pressure of gases in equilibrium with the silicate liquid. The Hertz-Knudsen equation for the evaporation rate J_i species i is

$$J_i = \frac{\gamma_i P_i^{sat}}{\sqrt{2\pi m_i RT}},$$

where γ_i is the evaporation coefficient, P_i^{sat} is the saturation vapor pressure of the gas species of mass m_i containing i, R is the gas constant and T is the absolute temperature. That the main effect of hydrogen on the evaporation
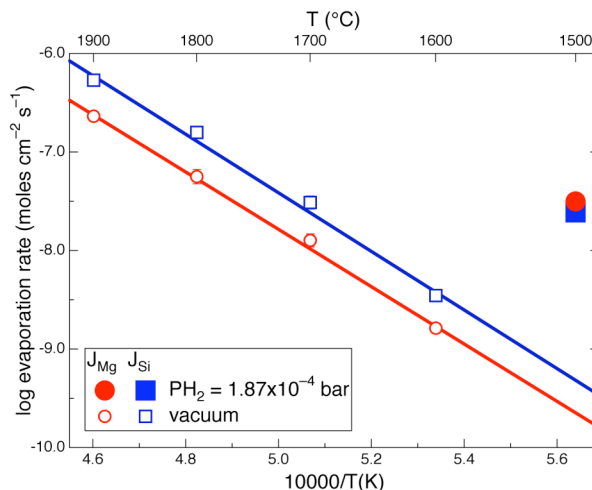

Figure 1. Evaporation kinetics as a function of temperature for a CAI-like liquid in vacuum and in 2×10^{-4} bars hydrogen. The evaporation rate in hydrogen (T=1500°C) is about two orders of magnitude larger that the vacuum rate extrapolated to 1500°C. Data from [2] and [3].

rate is due to the much larger saturation pressures of dominant gas species Mg and SiO when hydrogen is present is seen in the fact that the evaporation coefficients are much less sensitive to temperature than the evaporation rates themselves and that the evaporation coefficient for evaporation in hydrogen are much the same as that in vacuum at a comparable temperature (see Figure 3 in [3]).

Isotopic Fractionations. The exact pressure of gas present when the Type B CAIs were last molten is not known, but it is known that there was a gas present with approximately the same oxygen fugacity as that of a solar composition gas [4]. There already exist a wealth of high-precision data on the isotopic fractionation of CAI-like vacuum evaporation residues [3]. A key question is whether the kinetic fractionation factors determined in the vacuum experiments are relevant to evaporations in environments with finite hydrogen pressure. To address this with high-precision isotopic data, we remeasured the isotopic composition of a series of residues that were evaporated in 1.87×10^{-4} bars of hydrogen. The vacuum experiments are described in [2] and the magnesium isotopic measurements were done as described in [3] using the GV Instruments Isoprobe multicollector ICPMS at the University of Chicago. Al/Mg ratios were measured on the dissolved chips by

chips by multicollector ICPMS as described in [3]. Figure 2 shows the magnesium isotopic composition of the residues plotted in a manner such that if the process is one of Rayleigh fractionation, than the data will fall on a line with slope equal to $1-\alpha$ where α is the kinetic isotope fractionation factor defined as the ratio of the iso-
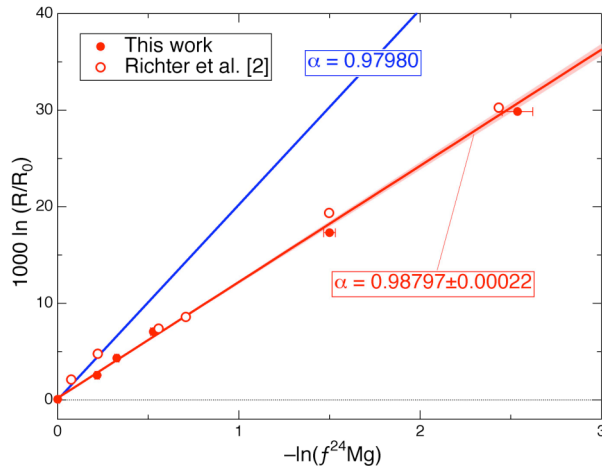

Figure 2: Magnesium isotopic composition of residues evaporated in 1.87×10^{4} bars H_2. Open symbols are earlier measurements by [2] and the small solid symbols are new high-precision data.

topic composition of the evaporation flux divided by the isotopic composition of the condensed phase. The kinetic isotopic fractionation factor for evaporation in hydrogen is compared to that in vacuum in Figure 3: the factors are approximately the same, but the low-pressure hydrogen value is slightly above the trend defined by the vacuum experiments. The experimentally determined values for α both in vacuum and in low-pressure hydrogen are much less fractionating (i.e., closer to one) than the often used value corresponding the to inverse square root of the mass of the isotope (shown as the "ideal" value in Figure 3.

Summary. Hydrogen (P>10^{-7} bars) significantly increases the evaporation rate at a given temperature (Fig. 1) because of its effect on the saturation vapor pressure of Mg and SiO. In contrast to this, the effect of hydro-
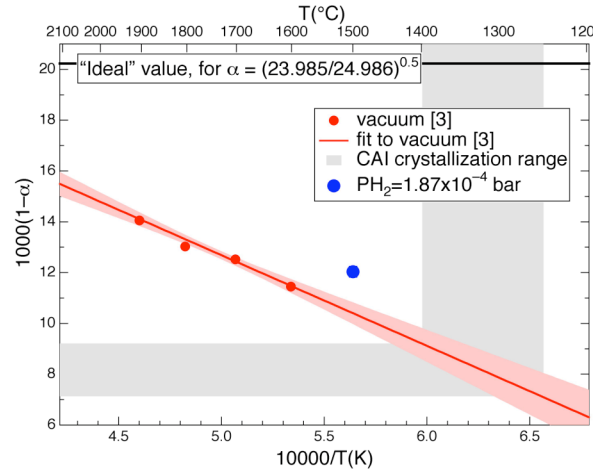
Figure 3. Experimentally determined kinetic isotope fractionation factor for residues evaporated at 1500°C into 1.87×10^{-4} bars hydrogen (blue symbol) compared to the isotope fractionation factor as a function of temperature for vacuum evaporation (red symbols) and the often-assumed value for kinetic fractionations corresponding to inverse square root of the masses of the magnesium isotopes (thick black line at the top of the figure).

gen on the kinetic isotope fractionation factor for magnesium is quite small (Figure 3). We are in the process of determining the effect of hydrogen on the fractionation of silicon isotopes by comparing the kinetic fractionation factor for residues evaporated in vacuum [5] to new measurements residues evaporated in low-pressure hydrogen.

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