THE EFFECT OF TOTAL PRESSURE ON VAPORIZATION OF ALKALIS FROM PARTIALLY MOLTEN CHONDRITIC MATERIAL

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ABSTRACT. In order to examine the effect of total pressure on vaporization of alkalis (Na, K, Rb) from a partially molten chondritic material, heating experiments were carried out under various He gas pressures ($\sim 10^{-5}$–$\sim 10^{-1}$ torr) at 1300°C. The rate of vaporization decreased in the order of Na > K > Rb with the increasing of the pressure, and reached a minimum at $\sim 10^{-1}$ torr.

1. Introduction

Alkali metals are moderately volatile at high temperatures and their vaporization behavior can constrain melting conditions of lunar and meteoritic materials (Gibson and Hubbard, 1972; Kreutzberger et al., 1985; Tsuchiyama et al., 1981; Matsuda et al., 1990).

We have conducted a series of vaporization experiments to study vaporization behavior of alkalis from a partially molten chondritic material (Shimaoka and Nakamura, 1989). Our results suggest that the vaporization is controlled mainly by the loss of elements from the partial melt, which is similar to the case of total melt (Tsuchiyama et al., 1981). However, there exists a systematic difference in vaporization rates between the two results. The vaporization rate can be influenced by total pressure, oxygen fugacity, and the chemical composition of charge as well as temperature. In this work, a heating experiment of a chondritic material was carried out under various total pressures ($\sim 10^{-5}$–$\sim 10^{-1}$ torr) at a constant temperature (1300°C) to investigate the effect of total pressure on vaporization of alkalis.

2. Experimental

The apparatus and the general procedure used for the vaporization experiment were described previously (Shimaoka and Nakamura, 1989). In this work, the starting material with fine grain-size ($\phi < 10$ um), similar to sample A in our previous work, was newly prepared from the Etter (L5) chondrite. Total pressure ($\sim 10^{-5}$–$\sim 10^{-1}$ torr) was adjusted using helium

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gas with high purity (99.999%). The sample was heated at a constant rate (~30°C/min) up to 1300°C. After maintaining this temperature for 40min, the power supply of the furnace was cut off and then the sample was quenched with a cooling rate of ~600°C/min. A run product was broken into fragments and divided into two parts. One part was used for preparation of thin section. The other part was crushed to fine powder and subjected to analyses of Na by atomic absorption spectroscopy, and of K and Rb by isotope dilution mass spectrometry. The proportions of holes and melt in the run products were estimated from the SEM back-scattered electron images (Figure 1) of run products (Shimaoka and Nakamura, 1989).
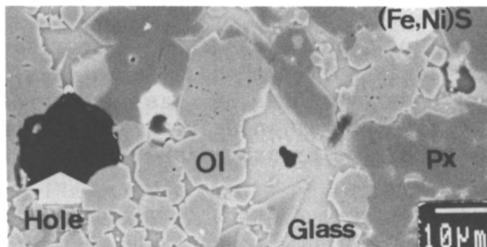
Figure 1. Back-scattered electron image of one of run products (1300°C, 40min). Ol: olivine, Px: pyroxene, (Fe,Ni)S: pentrandite

3. Result and Discussion

3.1. PREVIOUS STUDY

Previously, we obtained the following equation applicable to the vaporization of Na from a partial melt (Shimaoka and Nakamura, 1989):

$$\ln(C/C_0) = -k(f \cdot S_{tot}/V_p)t \qquad (1)$$

where C and C_0 are the concentrations in run product and starting material respectively, k, S_{tot} and V_p are the vaporization rate, the overall surface area and the volume of partial melt of charge respectively, t is the heating duration and f is the overall proportion of effective surface area of melt to total surface area of charge. The f value is tentatively estimated to be 0.2. Assuming that f, S_{tot} and V_p are constant during the heating, a linear correlation between t and $\ln(C/C_0)$ is expected. In Figure 2, results of an additional heating experiment for Na of this work are compared with those of previous work. In this diagram, all data points distribute along a linear curve, indicating a good reproducibility of vaporization rate in our heating experiments. As previously mentioned, the vaporization rate (k) calculated from the slope of regression lines are systematically high compared to those of the same temperature range obtained by extrapolating Tsuchiyama et al.(1981)'s values.
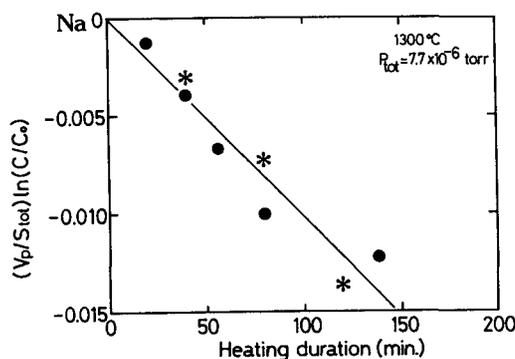
Figure 2. Plot of relative concentration of Na in heated samples corrected for effects of holes and partial melts as a function of heating duration. Symbols of stars and circles represent the present and previous results, respectively (Shimaoka and Nakamura, 1989).

3.2. EFFECT OF TOTAL PRESSURE

Applying equation (1), the vaporization rates of alkalis at various total pressures are obtained (Table 1 and Figure 3): in general, the vaporization rates of alkalis decrease with increasing total pressures. It is noted that the rate of vaporization decreases gradually for Na but abruptly for K and Rb at $\sim 10^{-1}$torr. Actually, almost no vaporization loss of Rb was detected for the run product at $\sim 10^{-1}$torr. It is inter-esting to note, that the pressure effect is greatest for Rb and smallest for Na.

TABLE 1. Results for run products at 1300°C

Run No.	89A	88A	86A	87A	90A
Duration (min)	40	40	40	40	40
Total pressure (torr)	7.7×10^{-2}	7.7×10^{-3}	7.7×10^{-4}	7.7×10^{-5}	7.7×10^{-6}
Holes (%)*1	10.4	9.2	11.0	8.3	9.5
Deg. of melt (%)*1	41.9	48.9	43.6	55.9	39.2
Olivine Fa (mol%)	27.5	26.9	25.1	25.0	26.3
Na (ppm)	2074	790	239	384	548
K (ppm)	454	315	235	238	286
Rb (ppm)	1.32	0.962	0.836	0.822	0.979
k_{Na} (cm^3/min·cm^2)*2	1.07×10^{-4}	1.89×10^{-4}	2.43×10^{-4}	2.93×10^{-4}	2.81×10^{-4}
k_K (cm^3/min·cm^2)*2	9.83×10^{-6}	4.18×10^{-5}	5.59×10^{-5}	7.69×10^{-5}	6.39×10^{-5}
k_{Rb} (cm^3/min·cm^2)*2	0	2.79×10^{-5}	3.33×10^{-5}	4.84×10^{-5}	3.34×10^{-5}

*1 See Shimaoka and Nakamura (1989).
*2 Vaporization rates of alkali metals (Na, K, Rb) calculated using eq.(1) with f=1 (see text).

Concentration of alkalis in the starting material;
Na=6550ppm, K=506ppm, Rb=1.32ppm
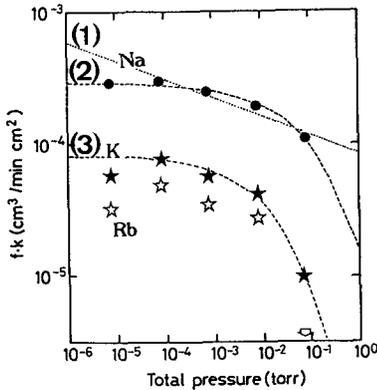
Figure 3. Plot of the vaporization rate of alkali metals as a function of total pressure. The regression lines are:
(1) $\log(f \cdot k) = -0.142 \log P_{tot} - 4.08$
(2) $\log(f \cdot k) = -1.34 \exp(\log P_{tot}) - 3.54$
(3) $\log(f \cdot k) = -2.79 \exp(\log P_{tot}) - 4.08$

Inspection of the least square fits indicates that an exponential curve ((2) and (3) in Figure 3) is best fitted to the data points. However, the physical meaning of exponential function is not clear. For simplification, we tested a linear curve ((1) in Figure 3) to compare with the data obtained by Tsuchiyama et al.(1981). The vaporization rate of Na was calculated to be 1.6×10^{-4} $cm^3/min \cdot cm^2$ at 1300°C (f=0.2) by extrapolating this line to 1 atm. This value seems to be consistent with the value $(0.5-1.7 \times 10^{-4} cm^3/min \cdot cm^2)$ calculated from Tsuchiyama's experiment at the same temperature and oxygen partial pressure ($10^{-11.6}$ atm). Since data points (particularly at high pressure) are still limited, more work may be needed to confirm the present result.

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