

THE ROLE OF HYDROGEN AND CARBON IN EVAPORATION OF FORSTERITE IN THE SOLAR NEBULA: H. NAGAHARA AND K. OZAWA, Geol. Inst., Univ. Tokyo, Hongo, Tokyo 113, Japan.

Evaporation of silicate dusts in the solar nebula is largely dependent on the gas composition, which may have resulted in chemical and isotopic fractionation. Previous experiments have shown that evaporation rate of silicates is largely enhanced in the presence of H₂ gas [1]. The role of hydrogen and carbon in evaporation of forsterite was thermochemically investigated. The results show that carbon enhances evaporation more effectively, that is, vapor pressure of the system and evaporation rate increase more largely in the presence of carbon compared to the same amount of hydrogen. In the solar nebula, evaporation rate of silicate dusts would have accelerated if organic carbon was concentrated which evaporated at lower temperatures at the time of nebular heating.

Many geochemical evidence particularly isotopic heterogeneities in chondrites and physical calculations have shown that the solar nebula had not experienced total evaporation at the early stage of its evolution. In spite of it, chondrites, including CAIs, chondrules and chemical fractionation among chemical groups, have recorded high-temperature processes for their origin. In such a nebula, evaporation would be the most important process at the highest temperature. It is thought to be a kinetics-controlling process, and the rate should be investigated experimentally. We have shown that evaporation of silicates in H₂ gas, which is the major constituent of the nebular gas, is quite different from that in vacuum in that the former is a kind of chemical reaction and the latter is a breakdown reaction, and therefore, kinetics is different to each other [1]. Evaporation rate of forsterite in vacuum to pressures below about 1×10^{-6} bar is almost constant, at pressures between 1×10^{-6} to 1×10^{-4} bar it shows strong dependence on total pressure which almost corresponds to P_{H_2} at those conditions, and show smaller dependence at pressures above 1×10^{-4} bar. Evaporation rate indicated by evaporation flux j is related to its equilibrium vapor pressure and the condensation coefficient α at that temperature by the following equation, $j = \alpha (P_{eq} - P) / (2\pi m k T)^{-1}$ ---- (1), where j is the flux, P_{eq} is the equilibrium vapor pressure, P is the actual vapor pressure that reaches zero in vacuum, m is the weight of a molecule of gas species, k is the Boltzman constant, and T is the temperature. Alpha is a kinetic factor due to surface kinetics between gas and solid, which varies from zero to unity. The j value of [1] at 1700°C for vacuum experiments is 5.7×10^{-9} (in Si gram atom units) and the estimated α is ~ 0.05 . These values are about a half of those obtained by previous workers [2].

Because of the usage of carbon as capsule in our experiments, we thermochemically investigated the effect of carbon in addition to hydrogen on the vaporization processes. Gas-solid equilibria in the Forsterite-H-C ternary system was studied with the energy minimization technique by using free energy data taken from JANAF [3]. Twenty seven gas species are treated, which include C, CH, CH₄, CO, CO₂, H, HCO, H₂, H₂O, Mg, Mg₂, MgH, MgO, MgOH, Mg(OH)₂, O, OH, O₂, Si, Si₂, Si₂C, SiC, SiC₂, SiH, SiH₄, SiO, and SiO₂. Although solid Si, SiC, C and enstatite were taken into consideration, the results show that they do not coexist with Fo in this system. Because we are interested in the Fo-gas equilibrium, a part of the system where Fo is stable is considered here. The system is bounded by Fo-H or Fo-C binary joins. Although the effect of C/O ratio in condensation of the solar nebula has been fully stressed by [4] and more generally by [5], the ratio was not taken as a critical parameter in the present study because abundance of O is not independently variable but its ratios to Mg and Si is fixed to forsterite composition in order to investigate congruent evaporation process of forsterite [1, 2, 6]. The temperature was also fixed at 1700°C, which is the main experimental condition [1].

The results are summarized in Figs. 1 and 2. In Fig. 1, equilibrium evaporation flux in the Fo-H and Fo-C systems are plotted against total pressure. The lower left end is j_{Si} of forsterite in equilibrium with its own vapor without C or H gas. The flux along the vaporous curve is larger

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in the Fo-C system than the Fo-H system in all the pressure range, and the difference becomes larger with increasing total pressure which varies from several times at 10^{-5} bar total pressure to almost 2 orders at 10^{-3} bar. Since the flux is positively correlated with the equilibrium vapor pressure, increase of the flux represents the increase in total pressure. In other word, the presence of much smaller amount of carbon than hydrogen pushes up the vapor pressure of the system more effectively. Fig. 2 shows the contours of vaporous surface and iso-flux surface in the part of the Fo-H-C ternary system. The vaporous surfaces of the system are shown by thin lines of which vapor pressure are indicated by italic numbers. Along the Fo-H join, the gas composition changes continuously with pressure. On the contrary, the gas composition does not change largely along the Fo-C join up to the point G, which is the limit of forsterite vaporous. Because the vaporous is steep along the Fo-C join and the gas composition has limitation, the tie lines are parallel to the H-C join at low pressures but almost parallel to the H-G join at high pressures. The iso-flux surfaces are shown by thick lines with the flux value timed 10^{-7} by bold numbers. The flux increases gently along the Fo-H join, and steeply along the Fo-C join toward the point G. At low pressures, the iso-flux surface intersects the vaporous surface, but tend to be parallel near the H-G boundary.

The smaller flux of [1] than [2] may be partly due to the usage of single-crystal forsterite in [1], as also shown by the experiments for single-crystal forsterite done by [7]. The usage of carbon as capsule in [1] could be another factor to reduce the α value, but this cannot be verified yet. The experimentally obtained flux in the H_2 gas is shown by an equation, $j = 2.3 \times 10^{-3} P^{0.97} + 3.98 \times 10^{-9}$ [1]. Alpha estimated on the assumption that the system is nearly along Fo-H join, varies from 0.16 at 4×10^{-6} bar P_{H_2} to 0.91 at 4.7×10^{-5} bar P_{H_2} . Thus the effect of C should not necessarily be taken into consideration to explain the experimental results of [1].

Present results suggests that evaporation rate of silicate dusts in the solar nebula is also significantly affected if interstellar (amorphous or organic) carbon is concentrated which could evaporate at much lower temperatures. [1] calculated that the lifetime of forsterite grain in the solar nebula, but it would be much shortened if carbon is also concentrated in the region of dust enrichment.

References [1] Nagahara, H. and Ozawa, K. (1994) *Meteoritics*, 29, 508, [2] Hashimoto, A. (1990) *Nature* 347, 53, [3] JANAF (1975), [4] Larimer, J. W. (1979) *GCA* 43, 1455, [5] Lattimer, J. M. et al. (1978) *Ap. J.* 219, 230*, [6] Mysen, B. O. and Kushiro, I. (1988) *Ame. Miner.* 73, 1, [7] Wang, J. et al. (1993) *LPSC* XXIV, 1479.

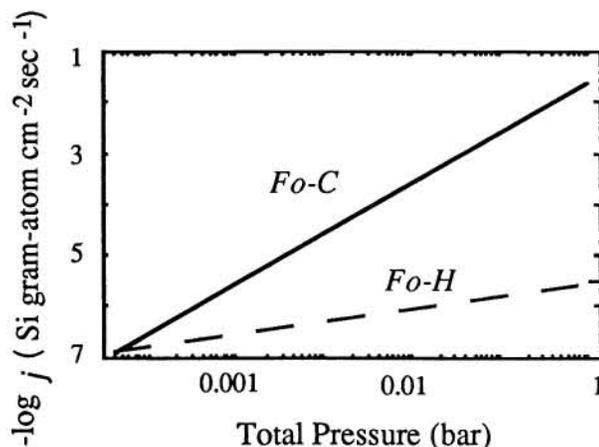


Fig. 1 Evaporation flux of forsterite shown as flux of silicon in the system Fo-H and F-C at 1700°C .

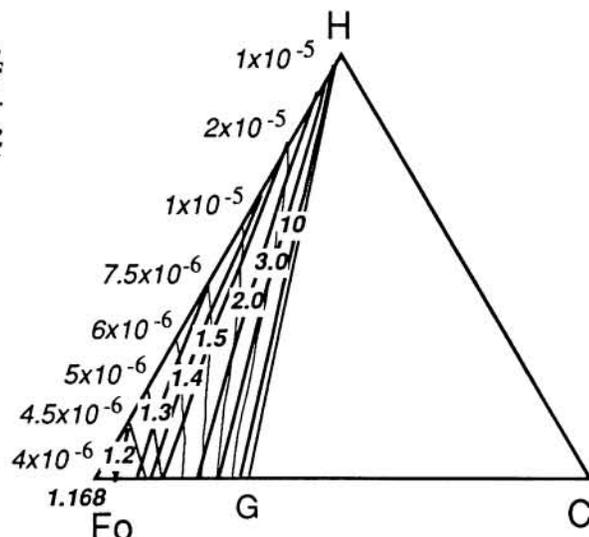


Fig. 2 The Fo-H-C ternary diagram at 1700°C in gram-atom unit. Solid lines: vaporous surface at the pressures numbered by italic (bar), thick lines: iso-evaporation rate surface of forsterite ($\text{Si gram-atom cm}^{-2} \text{sec}^{-1}$) with the number shown by bold italic ($\times 10^{-7}$).