

AN EXPERIMENTAL STUDY ON EVAPORATION OF SILICON CARBIDE IN A LOW-PRESSURE HYDROGEN-WATER VAPOR GAS MIXTURE. D. Yamamoto¹, A. Takigawa², and S. Tachibana², ¹Kyushu University, Japan (yamamoto.daiki.182@m.kyushu-u.ac.jp), ²The University of Tokyo, Japan.

Introduction: Presolar grains found in primitive meteorites and interplanetary dust particles are survivors of interstellar dust processes, disk thermal processes, and thermal/aqueous alteration processes on meteorite parent bodies [e.g., 1, 2, 3, 4], and they would record these processes.

Presolar SiC, one of the most abundant presolar grains [e.g., 5 and references therein], is a good indicator of the physicochemical conditions of the protosolar disk because of their thermodynamically unstable nature under relatively oxidative conditions of the protosolar disk gas [6]. Mendybaev et al. (2002) [2] conducted evaporation experiments of SiC under various temperature-oxygen fugacity (f_{O_2}) conditions at 1 atm and discussed the survivability of presolar SiC grains in the protosolar disk. Because the kinetics obtained at 1 atm may not be applicable to the low-pressure disk conditions, we carried out evaporation experiments of SiC in the low-pressure gas of an H₂-H₂O gas mixture to determine the reaction mechanism and kinetics of evaporation of SiC in the disk.

Experiments: The starting material was a polycrystalline β (3C)-SiC plate with dimensions of $\sim 4 \times (4-5) \times 0.6$ mm cut from a 20 cm-diameter polished disk synthesized through the chemical vapor deposition technique (Ferrotec Material Technologies). β -SiC is a major polytype of presolar SiC grains (accounting for $\sim 80\%$ of all SiC grains found in the Murchison meteorite [7]). The sample was placed on a sample holder made of alumina, and evaporation experiments were conducted at 1450–1250°C and 0.5 and 2.5 Pa of an H₂-H₂O gas mixture (P_{tot}) for 4–110.3 h in a high-

temperature vertical vacuum furnace with a gas flow system. In the gas flow system, deionized pure liquid water was put in a silica glass tube and water vapor from the liquid water was carried by hydrogen gas supplied from a hydrogen gas cylinder. The H₂/H₂O ratio in the experiments at 0.5 and 2.5 Pa was estimated to be ~ 225 and 420, (i.e., $P_{\text{H}_2\text{O}}$ of 2.2×10^{-3} and 6.0×10^{-3} Pa), respectively, based on the amount of liquid water consumed and the gas flow rate.

The sample weight and dimensions were measured before and after the experiments with an ultra-micron balance (Mettler Toledo XP2U) and a micrometer, respectively. The sample was analyzed with a FE-SEM-EDS (JEOL JSM-7000F). FIB lift-out sections of some samples were prepared with a FIB-SEM (Hitachi NX2000), and the thin sections were analyzed with a STEM-EDS (JEOL JEM-2800).

Results: From the thermodynamical calculations with FactSage software, the formation of solid SiO₂ is not expected under the experimental P - T conditions. FE-SEM-EDS and STEM-EDS analysis of the heated samples showed that there was no excess of oxygen near their surface in EDS spectra, suggesting that evaporation of SiC would have proceeded via active oxidation ($\text{SiC (s)} + 2\text{H}_2\text{O (g)} = \text{SiO (g)} + \text{CO (g)} + 2\text{H}_2 \text{(g)}$) [e.g., 8 and references therein].

Weight losses were observed for all run products (Fig. 1). Surface degradation for the samples heated at higher temperatures (1350–1450°C) was more severe than those heated at a lower temperature (1250°C). The evaporation rate k (cm s⁻¹) was estimated from the weight loss and original size of the sample as in [9]. Because of the different initial sizes, the modeled curves were different for each sample even for the same k (Fig. 1). The value of k was evaluated for each sample independently. The k was converted to the evaporation flux J (g cm⁻² s⁻¹) with the density of SiC (3.22 g cm⁻³), and the J under each temperature-pressure condition is shown in Fig. 2. The J depends only weakly on temperatures higher than 1350–1400°C, while larger temperature dependence was observed at temperatures lower than 1300°C, suggesting that the transition of the rate-limiting step occurs at ~ 1300 – 1350 °C. A similar transition was also observed in SiC evaporation experiments with the H₂-H₂O gas mixture at 1 atm where the transition occurred at ~ 1450 °C [10] (Fig. 2). The J at P_{tot} of 2.5 Pa is ~ 2 – 3 times larger than that at P_{tot} of 0.5 Pa at temperatures higher than 1400°C, while the J is the same within the variance of several experimental results at temperatures below 1300°C at

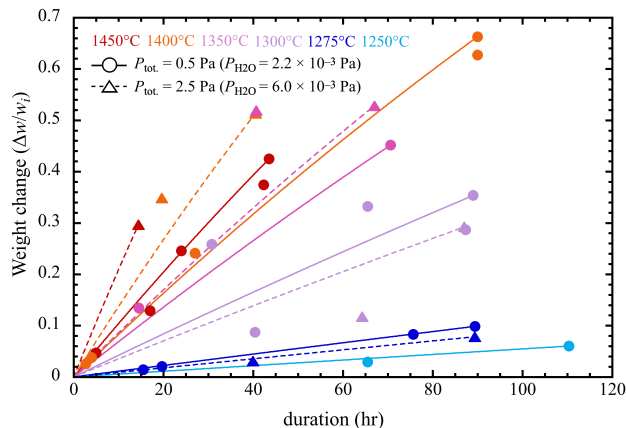


Fig. 1. Weight changes of the samples heated at different P - T conditions. The modeled curves for the samples heated for the longest duration under each P - T condition are shown as solid and dotted curves.

$P_{\text{tot.}}$ of 0.5 and 2.5 Pa. The temperature-dependent evaporation flux at $T \sim 1300^\circ\text{C}$ is similar to those obtained at f_{O_2} of IW-3 and IW-6 reported by Mendybaev et al. (2002) [2] (Fig. 2).

Discussion: Present results suggest that the mechanism and associated kinetics of evaporation of SiC largely depend on temperature (Fig. 2).

Under low-pressure conditions, supply of water vapor required for the active oxidation is limited and would thus control the reaction rate [e.g., 3, 12]. The flux of water vapor on SiC surface does not largely depend on temperature [e.g., 3, 11, 12], and the little or no dependence of the evaporation flux on temperature above 1300–1350°C indicates that the supply of water vapor is a rate-limiting step [3, 11, 12]. Because the flux of water vapor is proportional to $P_{\text{H}_2\text{O}}$, the difference between the values of J at $P_{\text{tot.}}$ of 0.5 and 2.5 Pa is broadly consistent with the difference in $P_{\text{H}_2\text{O}}$. The surface reaction frequency of colliding water molecules with SiC was estimated in the range of 0.05 to 0.1.

In contrast, the surface chemical reaction should control the overall reaction rate at lower temperatures [10]. Based on the J at 1275 and 1250°C, an apparent activation energy for the surface chemical reaction was $\sim 531 \text{ kJ mol}^{-1}$, which is identical to those at f_{O_2} of IW-3 and IW-6 (~ 563 and 556 kJ mol^{-1} , respectively) in [2].

The lifetime of 1 μm -diameter presolar SiC grains in the protosolar disk with $P_{\text{H}_2\text{O}}$ of 0.1 Pa was calculated based on the kinetic data in this study (Fig. 3). The timescales for oxygen isotopic exchange of 100 nm-sized amorphous forsterite (Mg_2SiO_4) grain and 1 μm -sized corundum (Al_2O_3) grain with disk gas were also shown based on the experimental oxygen diffusion data [11, 13]. The lifetime of SiC grain has little temperature dependence at temperatures higher than 1500°C where the supply of water vapor governs the reaction rate, while it has large temperature dependence at lower temperatures because the reaction rate is controlled by the surface chemical reaction (Fig. 3). The lifetime determined by the surface chemical reaction in this study is quite similar to that in [2] because of the similar J and activation energy each other (Fig. 2). Presolar SiC grains are unlikely to survive heating events associated with the formation of igneous CAIs, which would have been at $\sim 1400^\circ\text{C}$ and $P_{\text{H}_2\text{O}} > 0.1 \text{ Pa}$ for ~ 2 –3 days [12]. Presolar SiC could survive better than presolar silicates over a wider range of disk conditions, but could not escape complete evaporation under the conditions where original O-isotopic anomalies in presolar corundum were completely lost through isotopic exchange.

References: [1] Heck P. R. et al. (2009) *ApJ*, 698, 1155–1164. [2] Mendybaev R. A. (2002) *GCA*, 66, 661–682. [3]

Yamamoto D. et al. (2020) *MaPS*, 55, 1281–1292. [4] Wakita S. et al. (1976) *ApJ*, 836, 106 (6pp). [5] Lodders K. and Amari S. (2005) *Chem. Erde-Geochem.* 65, 93–166. [6] Larimer J. W. and Bartholomay M. (1979) *GCA* 43:1455–1466. [7] Daulton T. L. et al. (2003) *GCA* 67:4743–4767. [8] Narushima T. et al. (1997) *Mater. Trans. JIM* 38:821–835. [9] Takigawa A. et al. (2009) *ApJ.*, 707, L97–L101. [10] Kim H.-E. and Readey D. W. (1989) In *Silicon Carbide '87, Am. Ceram. Soc.*, 301–312. [11] Yamamoto D. et al. (2018) *ApJ*, 865, 98 (14pp). [12] Yamamoto D. et al. (2022) *GCA*, 336, 104–112. [13] Prot D. and Monty C. (1996) *Philos. Mag. A*, 73, 899–917. [14] Lodders K. (2003) *ApJ*, 591, 1220–1247.

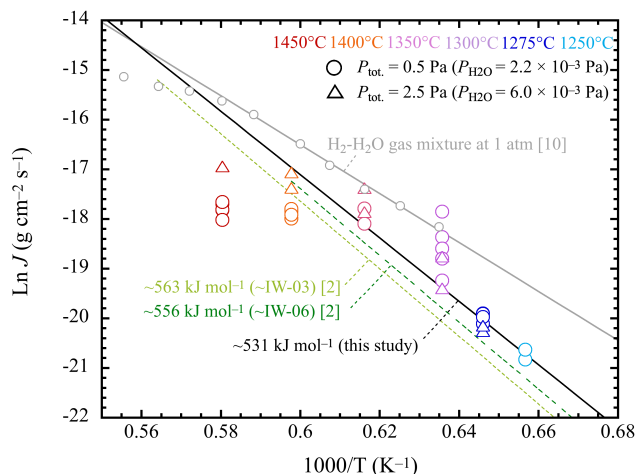


Fig. 2. Arrhenius plot of the evaporation flux J at different P - T conditions in this study. Evaporation flux J taken from the previous experimental studies [2, 10] are also shown.

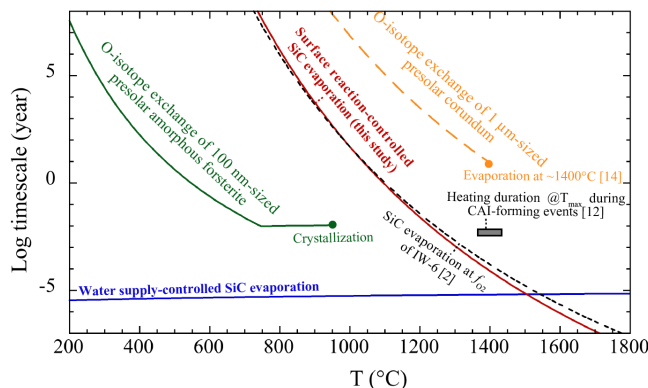


Fig. 3. Timescales of 1 μm -sized SiC evaporation controlled by the supply of water molecules to the grain surface (blue solid line) and the surface chemical reaction (red solid curve). The timescale of SiC evaporation based on the evaporation kinetics determined by [2] is shown as a black dotted curve. Oxygen isotope exchange timescales of 100 nm-sized presolar amorphous silicate and 1 μm -sized crystalline corundum grains [11, 13] are also shown for comparison. Gray shaded area represents the timescale at the maximum temperature during the igneous CAI formation.