SURVIVABILITY OF PRESOLAR SIC GRAINS IN THE PROTOSOLAR DISK: AN EXPERIMENTAL STUDY OF EVAPORATION KIENTICS OF SIC IN THE LOW PRESSURE-H2-H2O GAS MIXTURE.

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Introduction: Persistence of presolar grains in primitive meteorites and interplanetary dust particles suggests that there were conditions in the early Solar System in which destruction and/or isotope equilibration of these presolar grains were kinetically inhibited. The presence and abundance of presolar grains, therefore, could be an indicator of physicochemical conditions of the early Solar System [e.g., 1–3].

Presolar SiC grains are one of the most abundant presolar grains [e.g., 4 and references therein]. They are suggested to be thermodynamically unstable in the disk gas of the solar composition [5]. Meyndybaev et al. (2002) [3] estimated the survivability of presolar SiC grains in the protosolar disk based on evaporation experiments of SiC under various temperature-oxygen fugacity (f_{O_2}) conditions at 1 atm. 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 H_2–H_2O gas mixture to determine the reaction mechanism and kinetics of evaporation of SiC in the early solar system.

Experimental procedure: The starting material was a polycrystalline β (3C)-SiC plate with dimensions of ~4 × (4–5) × 0.6 mm cut from a 20 cm-diameter polished disk synthesized by chemical vapor deposition (Ferrotec Material Technologies). β-SiC is a major polytype of presolar SiC grains (accounting for ~80% of all SiC grains found in the Murchison meteorite [6]). The sample was placed on a sample holder made of alumina, and evaporation experiments were conducted at 1450–1250°C and 0.5 Pa of an H_2-H_2O gas mixture with the H_2/H_2O ratio of ~57 in a high-temperature vertical vacuum furnace with a gas flow system. The heating duration ranged from 4 to 110.3 hours. 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).

Results and discussion: Thermodynamical calculations with FactSage software shows that solid SiO_2 is thermodynamically unstable at the experimental P-T conditions, suggesting that evaporation of SiC would proceed via active oxidation (SiC (s) + 2H_2O (g) = SiO (g) + CO (g) + 2H_2 (g)) during the experiments [e.g, 7 and references therein]. Weight losses were observed for all run products. Severe surface degradation (i.e., intergranular attack and pit formation) of the run products was observed, which is consistent with previous experiments in the H_2-H_2O gas at 1 atm [e.g., 8, 9]. The presence of SiO_2 layer on the sample surface was not confirmed by FE-SEM-EDS.

The evaporation rate k (cm s^{-1}) was estimated from the weight loss and original size of the sample as in [10], and the k was converted to the evaporation flux J (g cm^{-2} s^{-1}) with the density of SiC (3.22 g cm^{-3}). The J depends only weakly on temperatures higher than 1350°C, while larger temperature dependence is observed at temperatures lower than 1300°C, suggesting that the transition of the rate-limiting step occurs at ~1350°C. Similar transition was also observed in SiC evaporation experiments with the 1 atm H_2-H_2O gas mixture [9]. The flux of water vapor on SiC surface does not largely depend on temperature [e.g., 2], and independence of the evaporation flux on temperature above 1350°C indicates that the supply of water vapor is a rate-limiting step [2, 9]. On the other hand, the surface reaction should govern the overall reaction rate at lower temperatures [2, 9]. The evaporation flux at 1350–1250°C is similar to that obtained under reducing conditions in [3], and the estimated activation energy for the surface reaction in this study (~576 kJ mol^{-1}) is also identical to that at f_{O_2} = ~10^{-16}–10^{-18} bar in [3] (556 kJ mol^{-1}). These consistencies suggest that the reaction mechanism is the same below 1350°C in both experiments. We note that additional detailed microscopic observation is indispensable to confirm whether SiO_2 layer forms or not.

The present results suggest that the lifetime of SiC grains is determined by the supply of water vapor at high temperatures (>~1200–1400°C), while it is determined by the surface reaction at lower temperatures. SiC grains with 0.5–1 μm in diameter, which is typical sizes of presolar SiC grains [e.g., 4], could survive at temperatures lower than ~800 °C during the lifetime of disk gas (1–10 Myr). Presolar SiC could survive more efficiently than presolar silicates that may have experienced oxygen isotope exchange reaction in the disk [2].

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