

SILICON AND OXYGEN ISOTOPIC FRACTIONATIONS DURING EVAPORATION OF SILICATE MELTS: EFFECT OF MELT STRUCTURE.

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Introduction: Some early Solar System materials have experienced high-temperature melting events that resulted in a loss of moderately volatile elements by evaporation. If evaporation occurs under low-pressure conditions, evaporation residues are expected to be enriched in heavy isotopes. Such enrichments are typical in coarse-grained igneous Type B and compact Type A CAIs. To model isotopic evolution of CAI precursors and to reconstruct their starting chemical composition, knowledge of isotopic fractionation factors (α_i) is required. The experimentally determined α_i s of Mg, Si and O isotopes for melilitic and forsteritic melts ([1] and references therein) show that isotope fractionation is essentially independent of temperature, and chemical and isotopic evaporation trajectories are the same in both low- P_{H_2} and vacuum conditions, despite much faster evaporation kinetics of Mg and Si at low- P_{H_2} . However, no data are available for anorthitic melts, despite anorthite being one of the major minerals of coarse-grained CAIs. Here we present new experimentally determined α_{Si} for molten anorthite, as well as for molten SiO_2 which is a major component of silicate melts.

Experimental and methods: The evaporation experiments were conducted at 1800°C in a vacuum ($<10^{-6}$ bar) using premolten samples loaded into 2.5 mm dia Ir-wire loops. Since the effective temperature of the transparent molten SiO_2 is significantly lower than the effective furnace temperature, the starting SiO_2 powder was mixed with 1 wt % of ultrafine metallic iridium. Anorthite (AN) melt was prepared by mixing appropriate amounts of high-purity SiO_2 , $CaCO_3$ and Al_2O_3 powders. Chemical composition of the starting materials and evaporation residues was obtained using the TESCAN LYRA3 FIB/FESEM. Silicon isotopic composition of bulk samples were measured by MC-ICPMS [2] and Si and O isotopic compositions in SiO_2 residues were measured using MC-SIMS [3, 4].

Results and discussion: Vacuum evaporation of AN melt at 1800°C resulted in strong enrichments of evaporation residues in heavy Si isotopes with $\delta^{29}Si = 9.1 \pm 0.1\%$ and $\delta^{30}Si = 18.2 \pm 0.2\%$ (2sd) in the most evaporated sample that lost 69% of Si. The obtained α_{Si} for AN melt are $\alpha_{29,28} = 0.9922 \pm 0.0003$ and $\alpha_{30,28} = 0.9845 \pm 0.0006$, which are the same as reported for molten Mg_2SiO_4 ($\alpha_{29,28} = 0.9928 \pm 0.0002$ at 1800°C [5]) and only slightly larger than those for melilitic and forsteritic melts ($\alpha_{29,28} = 0.9899 \pm 0.0004$ [6, 7]).

Although evaporation rates of molten SiO_2 are similar to those for other silicate melts, no heavy Si isotope enrichments were found in the SiO_2 residues: $\delta^{29}Si = 0.03 \pm 0.04\%$ in the least evaporated (1% loss) and $\delta^{29}Si = -0.02 \pm 0.02\%$ (2sd) in the most evaporated (32% loss) samples [8]. SIMS profiles through the residues showed that all samples studied are homogeneous isotopically. However, we cannot exclude the possibility of heavy Si isotope enrichments at the evaporating surface (on the μm scale), similar to that observed in experiments with single crystal forsterite [9]. The Si isotopic homogeneity suggests a similar mobility for all Si isotopes in the SiO_2 melt which is in agreement with experiments of Richter et al. [10] who demonstrated the same mobility of ^{70}Ge and ^{76}Ge in molten GeO_2 (an analog of Si in SiO_2).

The SIMS study showed that the outer parts (~25–75 μm from the evaporating surface) of the most evaporated SiO_2 residue appear to be enriched in $\delta^{18}O$ by ~1‰ compared to the central parts. One would expect much larger fractionations at the surface, if the fractionation is controlled by evaporation. However, we cannot completely rule out the possibility that the apparent enrichment in $\delta^{18}O$ at the edges is due to topographic effects: e.g., no such $\delta^{18}O$ variability was observed in a residue that lost 23% of SiO_2 . $\delta^{18}O$ up to 8‰ have been reported in bulk samples from experiments in which powdered and sintered SiO_2 was evaporated in vacuum at 1600°C [11]. Such high enrichments in $\delta^{18}O$ most likely are due to large effective surface area of the evaporating material.

Comparable α_{Si} values for melilitic, forsteritic and anorthitic melts suggest only a weak dependence of α_{Si} on melt structure and composition. Lack of isotopic fractionation in SiO_2 residues most likely is due to a slow isotopic exchange in very viscous SiO_2 melt. Going forward, our results from SiO_2 experiments can be applied to understand the origin of SiO_2 -dominated tektites, such as Libyan Desert Glasses.

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