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) is required. The experimentally determined αSi 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-PH2 and vacuum conditions, despite much faster evaporation kinetics of Mg and Si at low-PH2. 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 αSi for molten anorthite, as well as for molten SiO2 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 SiO2 is significantly lower than the effective furnace temperature, the starting SiO2 powder was mixed with 1 wt% of ultrafine metallic iridium. Anorthite (AN) melt was prepared by mixing appropriate amounts of high-purity SiO2, CaCO3 and Al2O3 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 SiO2 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 δ29Si = 9.1±1.0‰ and δ30Si = 18.2±0.2‰ (2sd) in the most evaporated sample that lost 69% of Si. The obtained αSi for AN melt are α29,28Si = 0.9922±0.0003 and α30,28Si = 0.9845±0.0006, which are the same as reported for molten Mg2SiO4 (α29,28Si = 0.9928±0.0002 at 1800°C [5]) and only slightly larger than those for melilitic and forsteritic melts (α29,28Si = 0.9899±0.0004 [6, 7]).

Although evaporation rates of molten SiO2 are similar to those for other silicate melts, no heavy Si isotope enrichments were found in the SiO2 residues: δ29Si = 0.03±0.04‰ in the least evaporated (1% loss) and δ29Si = –0.02±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 SiO2 melt which is in agreement with experiments of Richter et al. [10] who demonstrated the same mobility of 70Ge and 76Ge in molten GeO2 (an analog of Si in SiO2).

The SIMS study showed that the outer parts (~25–75 µm from the evaporating surface) of the most evaporated SiO2 residue appear to be enriched in δ18O 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 δ18O at the edges is due to topographic effects: e.g., no such δ18O variability was observed in a residue that lost 23% of SiO2, δ18O up to 8‰ have been reported in bulk samples from experiments in which powdered and sintered SiO2 was evaporated in vacuum at 1600°C [11]. Such high enrichments in δ18O most likely are due to large effective surface area of the evaporating material.

Comparable αSi values for melilitic, forsteritic and anorthitic melts suggest only a weak dependence of αSi on melt structure and composition. Lack of isotopic fractionation in SiO2 residues most likely is due to a slow isotopic exchange in very viscous SiO2 melt. Going forward, our results from SiO2 experiments can be applied to understand the origin of SiO2-dominated tektites, such as Libyan Desert Glasses.