

SILICON ISOTOPIC FRACTIONATION DURING EVAPORATION OF CAI-LIKE MELTS IN LOW-PRESSURE CONDITIONS.

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Introduction: The coarse-grained igneous texture of CAIs suggests that their precursors underwent melting followed by slow cooling. If such melting occurred under low-pressure conditions, evaporation of chemical elements and their mass-dependent isotopic fractionation would be expected. Isotopic fractionations observed in CAIs have been successfully reproduced by vacuum evaporation experiments using CAI-like melts; until recently, however, it was unclear if vacuum experiments adequately describe evaporation under the low P_{H_2} conditions of the solar nebula. Recent experiments showed that at 1600°C and 2×10^{-4} bar H_2 Mg and Si evaporate from FoB CAI-like melt ~40 times faster than in vacuum, but chemical evaporation trajectories and Mg isotopic fractionation remains the same [1, 2]. Here we present results on Si isotopic fractionation in samples evaporated in low P_{H_2} and in vacuum.

Experimental: In addition to the initial experiments at 2×10^{-4} bar H_2 and in vacuum using 2.5 mm samples [1, 2], new sets of low P_{H_2} evaporation experiments at 1600°C with the same starting material were conducted at Hokkaido University: at 2×10^{-4} bar H_2 using 1.5 mm sized samples and at 2×10^{-5} bar H_2 using 1.5 and 2.5 mm samples. The Si isotopic compositions of evaporation residues were measured using established solution techniques [3, 4] on a NuPlasma HR MC-ICP-MS at Trent University and a Neptune Plus MC-ICP-MS at University of St. Andrews. Chemical composition of run products was determined using a TESCAN LYRA3 FIB/FESEM equipped with an Oxford AZtec X-ray microanalysis system.

Results and Discussion: New experiments showed that evaporation trajectories of all run products follow the same trend established earlier [1, 2]. This further confirms the conclusion that presence of H_2 increases evaporation kinetics of Mg and Si (relative to vacuum), but relative evaporation rates of Mg and Si, and thus chemical evaporation trajectories, remain the same.

The Si isotopic composition measurements of evaporation residues from the first set of experiments at 2×10^{-4} bar H_2 give a fractionation factor of $\alpha_{29,28} = 0.9927 \pm 0.0004$ which is significantly different from $\alpha_{29,28} = 0.9901 \pm 0.0004$ obtained from previous vacuum evaporation experiments using different composition CAI-like melts [5, 6]. To check if smaller Si isotopic fractionation (larger $\alpha_{29,28}$) is due to Si recondensation (back reaction), we measured Si isotopes in evaporation run products from the new low P_{H_2} experiments. By lowering P_{H_2} and sample size we also lowered the amount of Si evaporating and thus limited possible Si recondensation. We also measured Si isotopes in evaporation residues from vacuum experiments.

Our new Si isotopic composition measurements of evaporation residues, including those from vacuum runs, resulted in $\alpha_{29,28} = 0.9917 \pm 0.0004$. The independence of $\alpha_{29,28}$ on run conditions (vacuum, 2×10^{-4} and 2×10^{-5} bars H_2) and sample size (2.5 and 1.5 mm) indicates that there is no detectable recondensation of Si in the experiments. This is in accord with the conclusion made based on Mg isotopic measurements [2].

It remains unclear why $\alpha_{29,28} = 0.9917 \pm 0.0004$ is different from $\alpha_{29,28} = 0.9901 \pm 0.0004$ reported for different composition melts [5, 6]. We should note that most previous experiments were conducted at 1900°C and thus we cannot exclude the temperature effect, as was observed for Mg isotopes [7]. However, no temperature effect on Si isotopic fractionation was reported from SIMS measurements of the same evaporation residues, but instead dependence of $\alpha_{29,28}$ on melt composition was observed [8]. However, it should be noted that this last conclusion was based on ion-probe measurements of Si isotopic composition, and no compositional effect on $\alpha_{29,28}$ (and $\alpha_{25,24}$ for Mg) was observed when solution ICP-MS data were interrogated [5].

Conclusions: The experiments show that, although evaporation of CAI-like melts in low-pressure H_2 is faster than in a vacuum, the chemical and isotopic fractionation of both Mg and Si remains the same. Therefore, the extensive experimental database on chemical and isotopic fractionation in vacuum can be safely used to model evaporation of CAI precursors in solar nebula, and previous interpretations of the isotopic fractionations of natural CAIs based on vacuum experiments do not require revision.

References: [1] Kamibayashi M. et al. (2018) 49th LPSC, Abst. #2432. [2] Mendybaev R. A. et al. (2018) 49th LPSC, Abst. #2580. [3] Georg R. B. et al. (2006) *Chem. Geol.* 235:95–104. [4] Savage P.S. and Moynier F. (2013) *EPSL* 361:487–496. [5] Mendybaev R. A. et al. (2013) *GCA* 123:368–384. [6] Mendybaev R. A. et al. (2017) *GCA* 201:49–64. [7] Richter F. M. et al. (2007) *GCA* 71:5544–5564. [8] Knight K. B. et al. (2009) *GCA* 73:6390–6401.