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Limited U isotope fractionation during evaporation: insights from gas levitation experiments

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Chondrites and their components exhibit discernible uranium isotope variations [1], which can significantly impact Pb-Pb chronology [2-3]. While the most extreme ^{235}U excess documented in CAIs (+59 ‰) is best explained by ^{247}Cm decay [4], the smaller effects seen in most CAIs (-6 to +1 ‰) and chondrules (-1 to -0.2 ‰) could stem from ^{247}Cm decay, evaporation, condensation, or a combination thereof. So far, the contribution of evaporation has not been experimentally evaluated. Since U is a refractory element with half condensation temperature of 1609 K [5], studying its behaviour during evaporation requires ultra-high temperature furnace. Here, we employed a gas-mixing aerodynamic levitation laser furnace to investigate the volatility and isotope behaviour of U in type I chondrule-like melt at temperature between 1633 and 2000°C, in various atmospheres (air, Ar, and H_2-Ar mixture), for durations of 180s to 900s. The chondrule-like starting material was doped with 100ppm U using HP-U std. Samples evaporated in Ar exhibit a maximum U loss of up to 60%, and homogeneous U content. The isotope data is consistent with a fractionation factor $\alpha^{238/235}U_{\text{gas-melt}}$ of 0.9998, higher than the predicted values (0.997) with 75 % saturation for UO_3 gas species. In contrast, experiments conducted in air show near total U loss (up to 97%), and the samples exhibit heterogeneous distribution of U across the hemispherical profile. Despite the extensive U loss, the samples display limited isotopic fractionation relative to the starting material (from -0.13 to +0.14 ‰), indicating diffusion-limit U evaporation in the melt. Overall, our results suggest that evaporation can hardly modify U isotopic composition in the melt even under oxidizing conditions. The wide range of d^{238}U seen in CAIs thus requires other drivers, such as ^{247}Cm decay, nucleosynthetic heterogeneity, and kinetically driven isotopic fractionation during condensation. More details will be presented at the conference.

Reference

[1] Li & Tissot, 2023, Chem Geol. 618, 121221
[2] Brennecka, Weyer, Wadhwa, et al. 2010, Science, 327, 449
[3] Tissot & Dauphas 2015 GeCoA, 167, 113
[4] Tissot, Dauphas, & Grossman. 2016, Sci Adv, 2, 1
[5] Wood, Smythe, & Harrison. 2019, Am Mineral, 104, 844