MASS-DEPENDENT FRACTIONATION OF OXYGEN ISOTOPES IN CAIs: TRACING THE EVOLUTION OF GAS IN THE EARLY SOLAR SYSTEM

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Introduction: Mass-dependent fractionation of oxygen isotopes during evaporation of calcium-aluminum-rich inclusions (CAIs) in the early solar system is expected. Experiments with forsterite melts show that with evaporation $^{18}O/^{16}O$, $^{25}Mg/^{24}Mg$ and $^{29}Si/^{28}Si$ increase such that $\delta^{18}O \geq \delta^{25}Mg >> \delta^{29}Si$ relative to initial ratios [1]. Similar results occur in experiments on CAI-like melts [2]. Models for evaporation are used here to explore the significance of oxygen mass-dependent fractionation, or lack thereof, in CAIs. An evolution in $\Delta^{17}O$ in H_2O gas over timescales commensurate with multiple episodes of heating of CAIs is implied.

Calculations: In this study calculations that simulate evaporation from the surface of a molten CAI as a function of composition, temperature, and $P(H_2)$ [3] show that $^{18}O/^{16}O$ should have increased during evaporation by easily-measured magnitudes. For example, typical $\delta^{25}Mg$ of 7 ‰ and $\delta^{29}Si$ of 3 ‰ in igneous CAIs should be accompanied by mass-dependent $\delta^{18}O$ of 10 ‰. The calculations show that FUN inclusions have $\delta^{25}Mg$, $\delta^{29}Si$, and $\delta^{18}O$ consistent with expectations. Non-Fun inclusions, however, do not record such simple behaviors during evaporation.

Comparisons with data: The magnitudes of $^{18}O/^{16}O$ mass-dependent fractionations in real non-FUN igneous CAIs can be estimated from their deviations from the slope-1.0 line in oxygen three-isotope space (the alternative of assuming the CCAM line to be primordial precludes all evaporative effects in oxygen, and only exacerbates the problem). Minerals with low-^{16}O, usually melilite (Mel) and anorthite (An), have deviations from the proposed primordial slope-1 line of ~8 ‰ at $\Delta^{17}O$ ~ 0, consistent with model results in this study. However, high-^{16}O phases, usually spinel (Sp) and Ti-rich clinopyroxene (Cpx), with $\Delta^{17}O$ ~ –20 ‰, exhibit little sign of mass-dependent fractionation of $^{18}O/^{16}O$. These phases instead cluster tightly on the slope-1 line. Yet these same minerals also have high $\delta^{25}Mg$ and $\delta^{29}Si$ indicative of evaporation.

One explanation is that molten CAIs exchanged oxygen with O-bearing gas in the disk subsequent to evaporation, erasing the evaporative effects on O isotopes. Exchange with CO is sluggish [4] and so H_2O as the most likely exchangeable gaseous reservoir for oxygen. In this scenario, two episodes of exchange between the CAI and gas are required because of the abundant evidence for ^{16}O-poor H_2O in the solar nebula. The first episode of exchange had to occur when the gas was ^{16}O rich to preserve low $\Delta^{17}O$ but erase positive deviations from the slope-1 line. The second phase of exchange, increasing $\Delta^{17}O$ in Mel and An, must have occurred after crystallization of Sp and Cpx so the latter were inoculated from exchange by slow self diffusion of O in their crystalline structures.

References: [1] Davis A.M. et al. 1990. *Nature* 347: 655-658. [2] Mendybaev R.A. et al. 2010. *41st Lunar and Planetary Science Conference*. pp. 2725. [3] Shahar A. and Young E.D. 2007. *Earth and Planetary Science Letters* 257: 497-510. [4] Bosenberg J.S. et al. 2005. *68th Annual Meteoritical Society Meeting*. pp. 5111.