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Experimental constraints on sulfur diffusion and isotopic fractionation during evaporation of reduced silicate melts: Implications for magma ocean evaporation

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Sulfur isotopic ratios represent a key tool for investigating planetary processes, such as magma ocean evaporation, as they may record isotopic fractionation. To constrain the effects of temperature and oxygen fugacity on S behavior during magma ocean evaporation, we conducted 73 experiments, under reduced conditions, in an atmosphere-controlled vertical furnace.

A starting Fe-free basaltic glass (~5000 ppm S, $\delta^{34}S$ = 16.1±0.5‰) was synthesized near the C–CO buffer in sealed silica tubes. This material was evaporated for 5 to 90 minutes, at 1400- 1600°C and fO_2 conditions ranging from IW-2 to IW+2, under both hydrous and anhydrous conditions. S distribution and concentrations were characterized by Electron Probe Micro-Analyzer (EPMA) mapping and core-to-rim line profiles, whereas S isotopic compositions were measured by Secondary Ion Mass Spectrometry (SIMS) along the same profiles.

Mapping revealed heterogeneous S distributions, including some samples showing strong, roughly uniform S depletion and others with convection and diffusion features. Diffusion patterns are restricted to experiments (19 samples) at lower temperatures (1400 and 1450°C) and relatively oxidizing conditions (IW+2 and IW).

Sulfur concentration profiles from samples showing diffusion reveal increasing S loss with temperature and duration, greater at IW+2 than at IW, and further enhanced in hydrous experiments. This trend is consistent with faster S diffusion under oxidizing conditions and under hydrous conditions, the latter likely related to changes in S speciation.

In anhydrous samples evaporated at IW and IW+2, the longest experiments display significant isotopic fractionation, with $\delta^{34}S$ variations of at least ~-5‰ from core to rim. Their cores also appear enriched in ^{34}S relative to the initial bulk composition, resulting in higher mean $\delta^{34}S$ values. Hydrous and S-depleted anhydrous samples are isotopically homogeneous, though shifted toward heavier $\delta^{34}S$ values relative to the starting material.

These results indicate isotopic fractionation triggered by surface evaporation, likely reflecting preferential diffusion and loss of the lighter ^{32}S isotope. The absence of isotopic gradients in hydrous and depleted anhydrous samples may suggest isotopic re-equilibration following ^{32}S loss.

Numerical modeling will further constrain S behavior in reduced silicate melts and its implications for magma ocean processes.