

**EXPERIMENTAL AND THEORETICAL DEMONSTRATION OF THE POTENTIAL IMPORTANCE OF GAS FLOW FOR CONTROLLING ISOTOPE FRACTIONATION DURING EVAPORATION OF “PEBBLES”.** H. Tang<sup>1‡</sup>, E. D. Young<sup>1</sup>, C. A. Macris<sup>2</sup>, A. A. Hogan<sup>2</sup>, and Q. R. Shollenberger<sup>3</sup> <sup>1</sup>University of California, Los Angeles, <sup>2</sup>Indiana University-Purdue University Indianapolis, <sup>3</sup>Lawrence Livermore National Laboratory, <sup>‡</sup>now at University of Science and Technology of China.

**Introduction:** Evaporation of molten rock in the Solar System is evidenced unequivocally by the millimeter to centimeter sized calcium-aluminum-rich inclusions (CAIs) found in chondrite meteorites (e.g., [1]). Many of these objects display enrichments in heavy isotopes of their primary constituents, including the elements Mg, Fe and Si. At the planetary scale, the effects of evaporation on planetesimals and larger bodies during the magma ocean stage of evolution has become a long-standing issue in order to interpret the isotopic shifts relative to chondritic compositions.

An outstanding issue is the effect when evaporation takes place with the presence of gas flow. Important analogous circumstances may include pebble accretion into planetary envelopes and the possibility that chondrules might form in collisional plumes. Indeed, the importance of the gas flow during evaporation has gone underappreciated, though it can accelerate evaporation rate by suppressing return flux from vapor to melts.

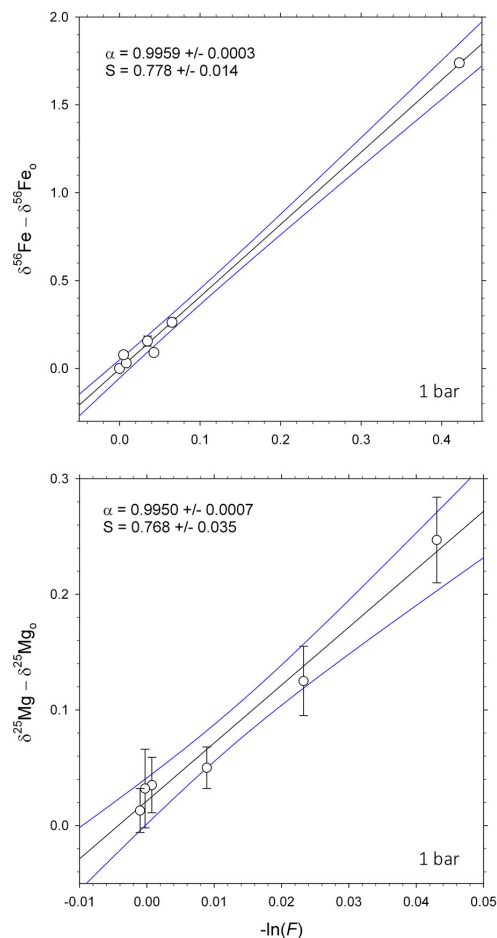
To quantitatively assess the impact of gas flow on the chemical and isotopic effects of evaporation, here we report results of evaporation experiments in which spherules of synthetic enstatite chondrite composition were melted in gas flow in a levitation system. Evaporative residues were analyzed for their Fe and Mg isotopic compositions as well as their Fe and Mg concentrations. In addition, we show with a mathematical model that the results can be explained by the primary control that the velocity of the gas has on the evaporation process. This interpretation can successfully explain the isotopic effects obtained using similar conditions reported previously [2-4].

**Methods:** Vaporization experiments were performed in the High-Temperature Conical Nozzle Levitation System at the High Pressure and Temperature Geochemistry lab at Indiana University-Purdue University Indianapolis (IUPUI). The EH-like spherical samples with ~2mm in diameter were levitated by a gas while being heated with a CO<sub>2</sub> IR laser. The pre-fused spherical samples were heated to 2273 or 2323 K for 120 to 480 s in pure N<sub>2</sub> or a mixture of Ar+CO+CO<sub>2</sub> (IW-0.5) (with the pressure of 0.33 bar and 1 bar). Samples were then quenched to glass for chemical and isotopic analyses.

Fe and Mg isotopic compositions in the experimental products were measured using MC-

ICPMS at UCLA following the methods described in [5-8]. The Fe/Al and Mg/Al ratios of the products were also measured on the MC-ICPMS using samples prior to purification for chemical variation resulted from evaporation.

Oxygen isotopic compositions of the samples were also analyzed using the fluorination system and Panorama mass spectrometry at UCLA. These results will be presented at the conference.



**Fig 1.** Plots of shifts in <sup>56</sup>Fe/<sup>54</sup>Fe and <sup>25</sup>Mg/<sup>24</sup>Mg vs fraction of Fe and Mg in the evaporated residues. The slopes corresponding to 1- $\alpha$  reflect evaporation at a saturation of ~0.7.

**Results and discussion:** The selected results are shown in Fig 1, where F is the fraction remaining of the

element of interest in the residues. The slopes indicate the value of  $1-\alpha$  according to the Rayleigh fractionation equation, reflecting the degree of saturation  $S$ . A simple relationship between the derived fractionation factor  $\alpha$  and  $S$  is

$$\Delta = \Delta_{eq} + (1 - S)\Delta_{kin} \quad (1)$$

where  $\Delta = \delta_{gas} - \delta_{melt} \approx 10^3 \ln \alpha$ . As a result, despite the different volatilities of Mg and Fe, the saturations of  $\sim 0.77$  are similar to each other within analytical uncertainties. This saturation value is also consistent with those obtained for other elements under similar conditions [2-4].

The relatively high saturation obtained here reflects the presence of gas flow that can promote removal of evaporated molecules from the evaporating surface, while allowing for a finite return flux. A first-order estimate of saturation in the similar P-T conditions without gas flow comes from comparing the rate of evaporation to the rate of return flux limited by diffusion through the ambient gas. According to [9, 10], the predicted  $S$  in this case is  $>0.992$ , much greater than our measured values of  $\sim 0.77$ .

To interpret the isotope fractionation observed in our experiments, we treat the flowing gas as an inviscid fluid and derive the drift velocity from the force balance that leads to stability against gravity during aerodynamic levitation [11]. Solving the force balance equation the drift gas velocity  $V_d$  leads to

$$V_d = \frac{\sqrt{2gm_{sphere}}}{2\sqrt{\pi C_D \rho_{gas}}} \frac{1}{r}, \quad (2)$$

where the drag coefficient  $C_D$  relies on the Reynolds number ( $Re$ ), and  $Re$  relies in turn on  $V_d$ . Values for the drift gas velocity  $V_d$  can be solved by iteration. The estimated  $V_d$  then is applied to derive the saturation by balancing the net flux over the surface of the evaporating sphere and the total escape flux of the evaporated product, yielding

$$S = 2\sqrt{\pi}V_t / \left( 4\pi V_d + 2\pi \left( \frac{(V_t^2 + 2V_d^2) \operatorname{erf}(V_d/V_t)}{4V_d} + \frac{V_t \exp(-V_d^2/V_t^2)}{2\sqrt{\pi}} \right) \right) \quad (3)$$

where  $V_t$  indicates the thermal velocity of the vapor.

Applying the estimated  $V_d$  and  $V_t$  of the corresponding levitation gas, the saturations calculated based on Eq (3) range from 0.77 to 0.80 for Fe and Mg isotopic fractionation, which, within the uncertainties, are identical to our measured saturation values. The

match between calculated measured saturation occurs where the flowing gas is treated as an inviscid fluid. This behavior may be due to the evaporation at the surface, which is known to reduce friction drag on the surface when the rock vapor is the dominant species at the surface of the evaporating sphere, which is plausible in our case [12].

Our model explains all of previous isotopic data obtained from aerodynamic levitation experiments [2-4], suggesting that it is the aerodynamics of the gas flow over the spheres that mainly controls saturation. The effects for oxygen are currently being measured.

This model may be applied to pebble accretion during growth of rocky bodies within the lifetime of protoplanetary disks [13]. Once approaching the mass of Mars, a growing rocky embryo can retain an  $H_2$ -rich atmosphere acquired from the protoplanetary disk. The atmosphere mass can be on the order of a per cent of that of the planetary core [14]. In this case cm-sized pebbles could encounter melting temperatures at terminal velocities when accreting onto the embryo through the atmosphere, resulting in mass loss and isotopic shifts due to evaporation as they descend to the surface. Evaporation of pebbles during pebble accretion could be at least partially responsible for the isotopic and chemical compositions of terrestrial planetary bodies. One such calculation has been presented [11].

#### References:

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