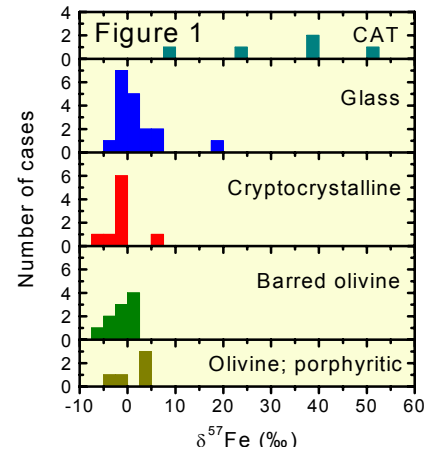


ISOTOPIC FRACTIONATION OF FE-57 IN STONY COSMIC SPHERULES: IMPLICATIONS FOR IRON LOSS S. Taylor¹, C. M. O'D. Alexander², J. Delaney^{3,5}, P. Ma^{4,5}, G. F. Herzog^{4,5}, and C. Engrand⁶
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Introduction: The extent of alteration due to atmospheric heating differs greatly among micrometeorites, even those that melted completely. Taylor et al. [1] have argued that a specific group of micrometeorites, the stony cosmic spherules (sCS) with unusually high Mg/Si ratios, low FeO contents, and high relative abundances of Ca, Al, and Ti, experienced the most intense heating. Corroboratory isotopic evidence indicates significant but variable evaporative losses of Fe, Si, and Mg in three such particles - from 1% to 50% of the pre-atmospheric mass [2]. If heating was indeed exceptionally intense for these particles, then most other sCS (and, *a fortiori*, unmelted micrometeorites) should have sustained smaller mass losses due to evaporation. To test this inference we measured the ⁵⁷Fe/⁵⁶Fe ratios of a suite of 43 sCS. The ⁵⁷Fe/⁵⁶Fe ratios effectively monitor evaporation of the relatively volatile FeO. We hoped also to learn from these measurements more about how sCS lose Fe.

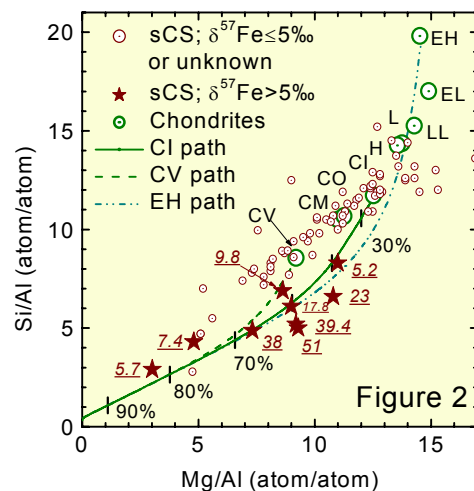
Experimental Methods: Two of the 43 sCS studied were mounted and polished at Rutgers University [see, 2]. The others had been prepared previously for electron microprobe analysis [1]. We analyzed the ⁵⁷Fe/⁵⁶Fe ratios of the samples with the Carnegie Cameca 6f ion microprobe using a 12.5-kV O⁻ primary beam in the shaped or flat-bottomed illumination mode, a 10-kV secondary accelerating voltage, a 50-eV energy window, and a 100- μ m field aperture. Primary beam currents were 1-4 nA and spot sizes were \sim 20 μ m across. As in [2], the standards used to measure the instrumental mass fractionation factors for all three elements were a BHVO basaltic glass and San Carlos olivine [3]. The 1- σ reproducibility of this standard was \sim 0.5 ‰, but estimates of the accuracy of the sample analyses are typically \sim 1‰. The influence of composition on the instrumental fractionation factor for Fe is not known but is thought to be relatively small, provided that magnetite and metal are not present. The presence of magnetite and olivine in some sCS may explain the spread to low values of $\delta^{57}\text{Fe}$.

Results: Figure 1 shows the results of our new and previous [2] analyses of $\delta^{57}\text{Fe}$. After [1], by using textures and color we classified the 47 particles as follows: 18 glass; 9 cryptocrystalline, 10 barred olivine; 5 CAT; 3 porphyritic; and 2 single-mineral (olivine). As a criterion for significant isotopic frac-



tionation of Fe, we adopt the conservative threshold of $\delta^{57}\text{Fe} > 5$. The measured values of $\delta^{57}\text{Fe}$ exceed this threshold for all 5 CAT spherules, for 3 of 18 glass spherules, and for 1 of 9 cryptocrystalline spherules, but not for any barred olivine or porphyritic particle.

Discussion: Alexander [4] has modeled the compositional evolution of chondritic melts evaporating at high temperature. Figure 2 shows these evaporative trajectories along with compositional data for the main chondrite groups and for numerous sCS [1, 2, this work]. Stars denote sCS with values of $\delta^{57}\text{Fe}$ (labels) > 5 ‰. The sCS without Fe isotopic fractionation fall on a line of roughly constant Si/Mg, but variable Al. Many data points lie close to the CC- and OC-, but not to the E-chondrite bulk compositions. E-chondrites, evidently, are not prolific sources



of sCS.

Qualitatively, isotopic fractionation of Fe occurs as predicted by the evaporation model of [4], *i.e.*, in sCS with Si/Al and Mg/Al ratios lowered by evaporation of Si and Mg. A weak correlation between $\delta^{57}\text{Fe}$ and the percentage of total evaporative loss (marked with vertical hatches on the CI trajectory of Fig. 2) probably reflects the range of initial compositions as indicated by the sCS with $\delta^{57}\text{Fe} \leq 5\text{‰}$.

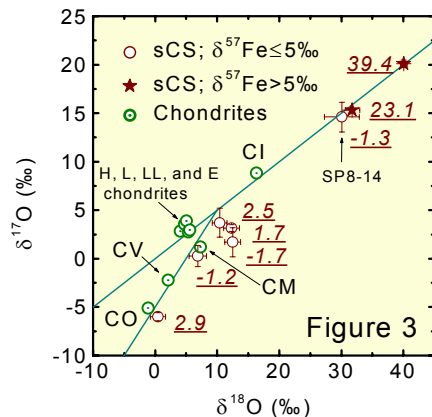


Figure 3 shows isotopic data for O, obtained with the UCLA ion microprobe [5], in sCS. Where available, values of $\delta^{57}\text{Fe}$ appear next to the data points. We find high values of $\delta^{57}\text{Fe}$ in two of three sCS with $\delta^{18}\text{O} \geq 25$. If these two sCS were initially CM-, CO- or CV-like, then both atmospheric exchange and evaporative loss (with enrichment of ^{17}O and ^{18}O) must have affected the oxygen isotopic abundances. Overall, the correlation between $\delta^{57}\text{Fe}$ and $\delta^{18}\text{O}$ is quantitatively weak, probably because of variations in initial oxygen composition. The texturally unusual sCS SP8-14 has an FeS rim, an inhomogeneous glass composition, and low $\delta^{57}\text{Fe}$, implying relatively mild heating. In this case, the high values of $\delta^{18}\text{O}$ may reflect atmospheric exchange and/or the presence in the starting material of high $\delta^{18}\text{O}$ phases such as carbonates and H_2O [e.g., 6] that were lost at relatively low temperature.

With the assumption of Rayleigh fractionation, the Fe isotopic data indicate the fractional but not the absolute evaporative losses of Fe. The size of the absolute loss depends on the starting composition and the detailed history of each particle. Much evidence suggests that precursors for many sCS were CI- or CM-like in composition [e.g., 7]. For illustration, we consider particle SP8-10, a CAT spherule with $\delta^{57}\text{Fe} = 39.4\text{‰}$, assuming the initial Fe/Al (wt) ratio was 17.8 (the CM value), and that the particle lost no Al. The total initial Fe content is then $17.8 \times \text{Al}$. The difference between this value and the measured Fe content represents the total loss of Fe due to evapora-

tion and ablation. In particle 10 we have Fe=0.05 wt %, Al=3.48 wt %, and $\delta^{57}\text{Fe} = 39.4\text{‰}$ (which implies a fractional evaporative loss of 98.7%). Assuming a final mass for the sCS of 100 μg , we calculate an evaporative loss of Fe between 61 μg and 3.6 μg depending on whether metal loss came after or before evaporation, respectively. Several lines of evidence point toward early metal loss: Measured Fe/Al ratios are less than CM values even in the sCS with the lowest $\delta^{57}\text{Fe}$ values; metal beads occur in 32% of the cryptocrystalline spherules, but in only 7% of the glass spherules and have not been observed in CAT spherules; and preliminary measurements give small values of $\delta^{57}\text{Fe}$ for metal beads. Perhaps early metal loss results from reduction-ejection of metal high in the atmosphere, where low O pressures mean that the competing process of oxidation is less efficient. From our data and simple mass balance calculations we infer that losses due to evaporation seldom exceed and are typically smaller than those due to ejection.

What becomes of the ejected metal? In an extreme case, a CM-like mass of 135 μg could lose up to 28 μg of Fe (along with ~ 7 μg O) to produce an unusually large residual sCS with a radius of 200 μm . If all the Fe were ejected intact, it might form a metallic spherule with a radius of 20 μm or a wüstite spherule with a radius of 25 μm . Most of the ablated metal would probably form smaller objects.

Conclusions: Isotopic fractionation of Fe in stony cosmic spherules (sCS) increases as Fe content decreases and is prevalent in sCS with high Ca and Al contents. In a survey of sCS, we found 9 of 47 sCS with $\delta^{57}\text{Fe} \geq 5\text{‰}$, which corresponds to a fractional evaporative loss of more than 40%. The highest value of $\delta^{57}\text{Fe}$ observed, 51.1 ± 6.3 ‰ corresponds to an evaporative loss of Fe of 99.6%. In general, however, depletions of Fe from sCS due to ejection of metal probably precede and exceed in size those due to evaporation. More precise measurements of $\delta^{57}\text{Fe}$ in sCS with higher iron contents would be useful. Preliminary measurements in such samples show sizable fractionation of K isotopic abundances.

References: [1] Taylor S. et al. (2000) *M&PS*, 35, 651-666. [2] Alexander C.M.O'D. et al. (2001) *GCA*, in press. [3] Alexander C.M.O'D. and Wang J. (2001) *MPS*, 36, 419-428. [4] Alexander C.M.O'D. (2001) *MPS*, 36, 255-283. [5] Engrand C. et al. (1998) *LPS*, XXIX, 1473.pdf. [6] Brown P.G. et al., (2001). *Science*, 290, 320-325. [7] Brownlee D.E. et al. (1997) *MPS*, 32, 157-175; Kurat G. et al. (1994) *GCA*, 58, 3879-3904; Matrajt G. et al. (2001) *MPS*, 36, A127; Maurette M. et al. (2000) *Planet. Space Sci.*, 48, 1117-1137.