SILICON ISOTOPE RATIO ANALYSIS OF A CAI BY LASER ABLATION MC-ICPMS AND IMPLICATIONS FOR THE ASTROPHYSICS OF CAI FORMATION.

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Introduction: While it is well known that igneous CAIs tend to have high $^{29}Si/^{28}Si$ and $^{25}Mg/^{24}Mg$ relative to chondritic values, there have been few systematic studies that relate observed isotope fractionations in CAIs to their conditions and timescales of formation. Together with a wealth of information about the physical chemistry associated with evaporation and condensation of CAI-like liquids [1] we apply silicon isotope ratios obtained *in situ* by UV laser ablation MC-ICPMS, and existing $^{25}Mg/^{24}Mg$ MC-ICPMS data [2], to construct a P_{H_2}-time curve unique to a CAI that constrains the astrophysical setting of CAI melting.

Analytical Methods: Silicon isotope ratios were measured using UV laser ablation MC-ICPMS. Mass interferences were eliminated by operating with a mass resolving power (m/Δm) of ~9000. Corrections for instrumental mass bias were perfomed by sample-standard bracketing with our in-house standard (San Carlos olivine). We used a 193 nm excimer laser operated with a pulse repetition rate of 2 to 6 Hz and UV fluence of 21 to 27 J/cm^2. *In situ* analyses were acquired from spots measuring 100 μm in diameter and ~20-30 μm deep. Precision of the LA-MC-ICPMS analyses is on the order of +/− 0.2‰ 1σ for both $\delta^{29}Si$ and $\delta^{30}Si$. The accuracy of our Si isotope ratio results was assessed using a synthetic glass on the $CaMgSi_2O_6$-$CaAl_2Si_2O_6$ join prepared with a 1% spike of ^{28}Si.

Sample Description: For this study we analyzed Leoville 144A, an igneous, compact type A CAI from a CV3 meteorite. It is composed mainly of melilite, magnesian spinel, minor Al-Ti rich diopside and minor perovskite typically associated with diopside.

Modeling: Isotopic fractionation in a volatilizing CAI is controlled by the relative rates of evaporation and diffusion of the constituent phases. In order to model the isotopic evolution of the CAIs we obtained numerical solutions to the problem of elemental and isotopic fractionation at the moving surface of an evaporating sphere coupled with diffusional transport within the sphere. The shrinking sphere represents a volatilizing protoCAI where the rate of surface retreat is a function of the average volatility of the object as prescribed by differences between ambient and equilibrium vapor pressures of the constituents of the object. The implicit finite difference scheme for solving this problem, akin to an inverse Stefan problem, was described previously [3] and has been modified to account for the effect of Si and Mg isotope fractionation that includes isotope-specific rates of diffusion within the sphere.

In order to validate our calculation scheme we calculated models for the measured changes in Mg isotope ratios and chemical compositions attending evaporation of type B CAI-like liquids in the laboratory reported by Richter et al. [1] and Si isotope fractionation measured for analogous experiments reported by Janney et al. [4].

The calculations show that our models accurately reproduce the experimental results for both Si and Mg isotope fractionation using the $^{29}Si/^{28}Si$ and $^{25}Mg/^{24}Mg$ fractionation factors of 0.9898 and 0.9869, respectively (Fig. 1). These fractionation factors are the same as those reported in the original studies [1,4] and verify that diffusion played a negligible role in determining the buk isotopic compositions of the the experimental products.

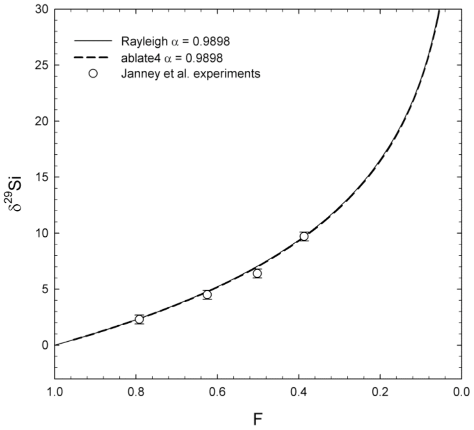

Fig. 1 Comparison of experimental $\delta^{29}Si$ vs. Si remaining [4] and model calculations for experiments using code ablate4 (this study).

Our calculations also faithfully reproduce the chemical evolution of the bulk products with up to 60 or 70% mass loss (Fig. 2).

Results: Fifteen LA-MC-ICPMS analyses of $^{29}Si/^{28}Si$ and $^{30}Si/^{28}Si$ were obtained for CAI Leoville 144A. The $\delta^{29}Si$ values are essentially uniform across the interior of the object with an average of 3.4 +/−0.3‰ (Fig 3). There is a detectable increase in $\delta^{29}Si$ at the margin of the object of 1‰ relative to the interior that correlates with a similar increase in δ^{25}Mg found previously [2].

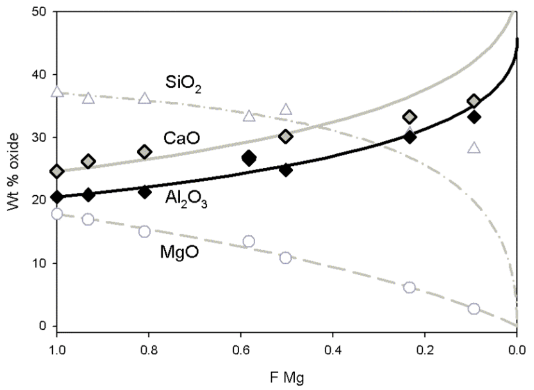

Fig. 2 Comparison of experimental chemical evolution vs. Mg loss (symbols, [1]) and model calculations (curves) for experiments using code ablate4.

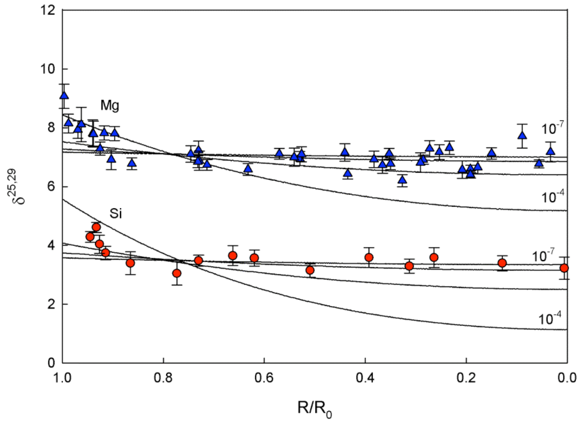

Fig. 3 Silicon (this study) and Mg [2] isotope ratio data vs. distance across Leoville CAI 144A. Also shown are model evaporation curves at indicated P_{H_2}.

Comparisons with Models: Modeling the high δ^{29}Si and δ^{25}Mg in 144A constrains the initial bulk composition, duration, and pressure (P_{H_2}) attending melting of the object. We emphasize that a unique relationship between composition, time and pressure would not be constrained with either δ^{29}Si or δ^{25}Mg alone, and that it is the combination of the two isotope ratios that yields the sought after result.

We calculated both bulk isotopic values (Fig. 4) and zoning profiles (Fig. 3) as a function of pressure. The bulk model results are used to produce a unique univariant relationship between P_{H_2} and time during melting (Fig. 4). The resulting univariancy corresponds to a unique initial composition that is similar to condensation models for CAIs. Similar curves can be constructed for other CAIs where both Si and Mg isotope ratio data are obtained.

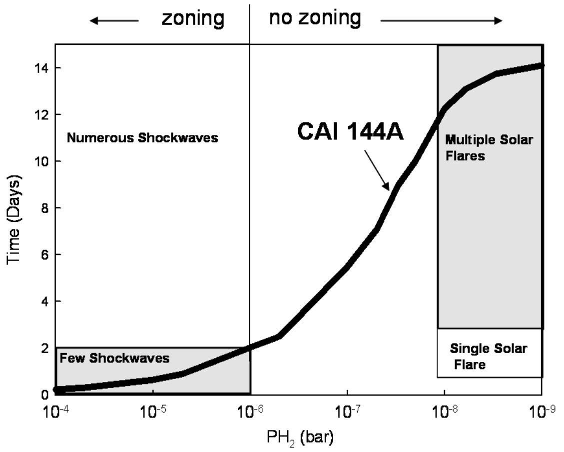

Fig. 4 P_{H_2} – time univariancy uniquely defined by Si and Mg isotope ratio data for CAI 144A.

Astrophysical Implications: The shockwave model for heating chondrules and igneous CAIs predicts melting temperatures for a few hours at pressures that exceed ambient by a factor of 10 to 100 [5]. If melting of 144A occurred in shockwaves, the likely pressure was between 10^{-6} bar to 10^{-3} bar. Figure 4 indicates that one or perhaps a few shockwaves could explain the isotope data for 144A. Shockwave heating also implies that the CAI was at one time zoned in ^{29}Si/^{28}Si and ^{25}Mg/^{24}Mg because of the rapid rate of evaporation at high P_{H_2}. Elimination of the zoning profile would take ~500 years of exposure to high temperatures of ~1300K.

An alternative hypothesis for CAI formation consistent with Fig. 4 is exposure to numerous solar flare events (each event lasts on the order of 1 day). This would occur if CAIs formed in the inner annulus of the solar protoplanetary disk and were later dispersed to more distal regions of the disk, perhaps by x-winds [6]. Our modeling of 144A suggests a duration of melting of ~14 days at vacuum. In this case the lack of zoning in the body of 144A is a primary feature of the CAI.

We calculate that the sharp gradients in δ^{29}Si and δ^{25}Mg at the margins of 144A require ~10 years of solid-state evaporation (sublimation) at near-solidus temperature following melting.

References: [1] Richter F. M. et al. (2002) *GCA, 66,* 521–540. [2] Young E. D. et al. (2005) *EPSL, 238,* 272–283. [3] Young E. D. (2005) *Science, 308,* Supplemental Material. [4] Janney P. E. (2005) *LPS XXXVI,* Abstract #2123. [5] Desch S. J. and H. C. Connolly (2002) *Meteoritics, 37,* 183–202. [6] Shu F. H. et al. (2001) *Astrophys. Jour., 548,* 1029–1050.