**SILICON ISOTOPES IN TYPE 1AB CHONDRULES AND IMPLICATIONS FOR THE CONDITIONS ATTENDING GAS-LIQUID REACTIONS IN CHONDRULES.** E. R. Harju[1] and E. D. Young[1], [1]Department of Earth and Space Sciences, University of California Los Angeles, Los Angeles, CA, USA (harju@ucla.edu, eyoung@ess.ucla.edu).

Introduction: Some chondrules are known to have mantles of pyroxene. One model [1] suggests that olivine in molten or partially molten CR and CV chondrules interacted with SiO gas to create pyroxene by a reaction similar to:

$$Mg_2SiO_4 + SiO + H_2O \rightarrow Mg_2Si_2O_6 + H_2$$
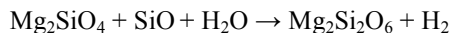
where SiO gas is here shown to be oxidized by water to form SiO_2 in the melt. This model is used to explain oxygen isotope data by interaction of chondrules with a ^{16}O-poor gaseous reservoir (e.g., water [2]). If a silicon-rich gas did interact with chondrules in the nebula there may be evidence of this in the silicon isotopes present in chondrule rims and interiors. The above reaction essentially represents condensation of SiO. The isotope fractionation due to condensation can be calculated using the following condensation model developed by [3,4]:

$$\alpha_{cond} = \frac{\alpha_{Eq}\alpha_{Kin}s_i}{\alpha_{Eq}(s_i - 1) + \alpha_{Kin}} \qquad (1)$$

where s_i is equal to the partial pressure of gaseous species i divided by the equilibrium partial pressure of of gas i and is a measure of the degree of undercooling in the system. The fractionation factor α_{kin} embodies the isotope fractionation associated with condensation inclusive of collision frequency and zero point energy effects [3]. We are testing the hypothesis that the reaction between SiO in the gas phase and chondrules might lead to silicon isotope fractionation by measuring Si isotope ratios in interiors and pyroxene margins of chondrules by UV laser ablation MC-ICPMS.

Methods: Thin and thick sections of CV and CR chondrites were examined to find Type 1AB chondrules appropriate for the study. The presence of pyroxene rims was then confirmed by EDS in variable pressure mode. Silicon isotopes were measured in-situ using UV laser ablation and the ThermoFinnigan Neptune MC-ICPMS at UCLA. The sections inspected with the UCLA JEOL electron microprobe following laser ablation studies to confirm the mineralogy of the phases analyzed.

Results: Figure 1 is a silicon three-isotope plot showing data obtained from one type 1AB chondrule from the CV chondrite Allende. The mass-dependent fractionation line is shown for reference. Olivine is higher in $^{29}Si/^{28}Si$ compared to pyroxene by ~ 0.3‰.

Figure 2 shows the isotope shift expected for $\delta^{29}Si$ as a function of the fraction of silicon that remains in the gas, *F*, in a Raleigh-type condensation at 1700 K for different values of s_{SiO} based on equation (1). In this calculation, there is a distinct reservoir effect represented by the fraction of silicon (SiO) remaining in the gas phase that results in an increase in $^{29}Si/^{28}Si$ in the condensed phase as isotopically lighter Si is lost to the condenation reaction. This reservoir effect competes with the lowering of $^{29}Si/^{28}Si$ due to the kinetics of condensation alone. The difference in $\delta^{29}Si$ between pyroxene and olivine shown in Figure 1 is shown as a horizontal line in Figure 2.

Figure 3 is a plot of the isotope shift expected versus the oversaturation represented by s_{SiO} that drives condensation in the absence of Raleigh-type reservoir effects at 1700 K. The intersection of the curve and the experimentally determined isotope shift is at $s_{SiO} = 1.04$, which corresponds to an undercooling of -0.36 K.
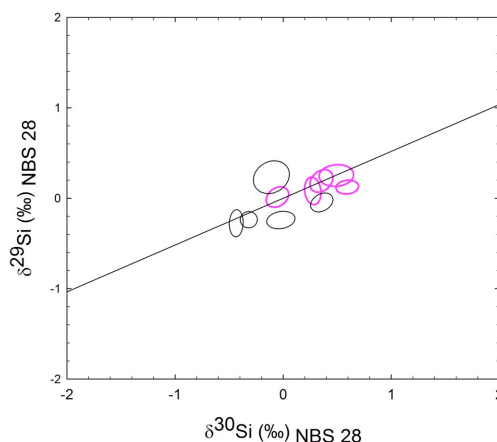

Figure 1. Silicon three-isotope plot for an Allende chondrule obtained by LA-MC-ICPMS from a type 1AB chondrule in Allende. The pink ellipses are olivine and black ellipses are pyroxene. The ellipses represent 2σ analytical errors.

Discussion: Figure 2 suggests multiple scenarios that are consistent with the experimentally determined
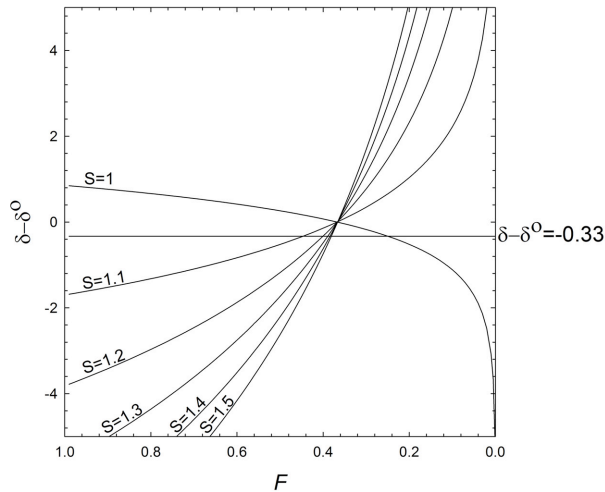
Figure 2. The expected isotpe shift in δ^{29}Si as a function of the fraction of silicon remaining in the gas, F, in a Raleigh-type condensation. The curves represent different values of $s_{SiO} = p_{SiO}/p_{SiO, Eq}$. The horizontal line represents the experimentally determined value of $\delta-\delta°$.

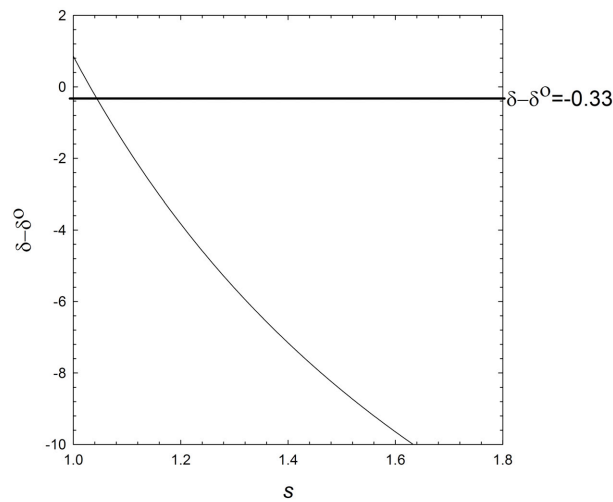**Figure 3.** The expected isotope shift for δ^{29}Si as a function of $s_{SiO} = p_{SiO}/p_{SiO, Eq}$. The horizontal line represent the experimentally determined value of $\delta-\delta°$. The intersection of the two curves is at $s_{SiO} = 1.04$, which corresponds to an undercooling of -0.36 K.

fractionation (shift in δ^{29}Si, $\delta-\delta°$) of -0.33 per mil. The case of $s_{SiO} = 1$ represents equilibrium were $p_{SiO} = p_{SiO, Eq}$. If the process took place at equilibrium then approximately 75% of the silicon residing in the gas would have condensed in a Raleigh-type process. A value of $\delta-\delta° = -0.33$ could also be obtained with

higher partial pressures of SiO compared to the equilibrium partial pressure of SiO (increasing s_{SiO}) if less of the gas from the reservoir condensed. In the case of $s_{SiO} = 1.1$ approximately 55% of the silicon in the gas has condensed. The values of s_{SiO} from 1.1 to 1.5 correspond to undercoolings of -0.86 to -3.67 K.

The curve in Figure 3 gives theoretical values for $\delta-\delta°$ based on s_{SiO} alone with no reservoir effects, and shows that a very small amount of undercooling could explain the data. In this case, $\delta-\delta° = -0.33$ intersects the curve once at a value of $s_{SiO} = 1.04$, slightly above the equilibrium s_{SiO}. This value of s_{SiO} is not sensitive to temperature in the range of 1400-1700 K. If the value of s_{SiO} was 1.1 or greater (i.e., undercooling was on the order of 1 degree or more) then Figure 3 implies that a shift in δ^{29}Si of -2‰ or greater is expected. We would easily detect such a shift.

Based on the available data the most robust solution appears to be a process where the pyroxene in the chondrule rim formed under near-equilibrium conditions, with the partial pressure of SiO slightly greater than the equilibrium partial pressure of SiO, corresponding to undercooling of < 1 degree. This result is consistent with the notion that chondrules in general were molten in near equilibrium with enveloping gas [5]. Just as chondrules lack evidence for evaporation (elevated stable isotope ratios), so too do they exhibit no obvious isotopic consequences of condensation where condensation has been proposed as a reaction.

Isotope data from additional type 1AB chondrules will be collected in the future. A larger data set should help to better constrain the conditions under which the pyroxene rims formed.

References: [1] Chaussidon, M. et al. (2008) *GCA, 72,* 1924-1938. [2] Clayton, R. N. et al. (1983) *Chondrules and Their Origins,* 37-43. [3] Young, E. D. and Schauble, E. A. (2012) *Meteoritics & Planet. Sci., 47,* Abstract #4382. [4] Simon, J. I. and DePaolo, D. J. (2010) *EPSL, 289,* 457-466. [5] Galy, A. et al. (2000), *Science, 290,* 1751-1753.