SILICON ISOTOPIC COMPOSTION OF ALLENDE CHONDRULES AND NEBULAR PROCESSES.

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Introduction: There are many theories that have been advanced to explain the origin of chondrules [1]. One tool that can be used to probe for information relating to the physical processes associated with chondrule formation is stable isotope measurements. Possible mechanisms to generate isotopic variation in chondrules include (i) closed system formation from isotopically heterogeneous precursor material; (ii) fractionation during chondrule formation (whether nebular or planetesimally based); (iii) secondary alteration processes on the chondrite parent body or in the nebula, that are either aqueous or thermally metamorphic in nature. However, one caveat associated with measurements of bulk chondrules is that the isotopic composition of an individual chondrule could be the result of one or more than one fractionation event.

The early gas-source mass spectrometry Si isotopic data of [2] for chondrules, showed a limited isotopic range of δ^{30}Si~1.9‰, compared to CAIs, δ^{30}Si~20‰ [3]. They did not find any simple correlations of δ^{30}Si with chondrule texture, mineralogy or size. However, the range of δ^{30}Si in chondrules was somewhat greater than that in bulk chondrites measured by [4] (δ^{30}Si~1.2‰), using similar analytical methods. The external precision at that time was ±0.4‰ ($2\sigma_{SD}$), which is more that a factor of two greater than modern multi-collector inductively-coupled-plasma mass spectrometry (MC-ICPMS) methods. The recent high precision Si isotope measurements of meteoritic material [5-9], have been focused primarily on bulk samples, with a much more limited range in Si isotopic composition than observed by [4]. However, the limited δ^{30}Si data that exist for individual chondrules ([5, 10]), also point to a larger range in chondrule δ^{30}Si compositions relative to bulk meteorites.

The aim of this study was to investigate the extent and cause of Si isotopic variability in chondrules. One particular interest was to determine how consistent the greater δ^{30}Si variability in chondrules relative to chondrites was, and whether this variability was the result of precursor heterogeneities or open system chondrule formation processes.

Method: Ten chondrules were picked from Allende, each of which was split into two fragments. One half was preserved to mount for petrological examination, and the other half was used for the isotopic and major element analysis. The Si isotopic compositions were measured using multi-collector ICP-MS in a similar fashion to that outlined in [8]. One half of each chondrule was powdered and was processed with an alkaline flux with the resulting "fusion-cake" made up into an acidic solution with HCl.

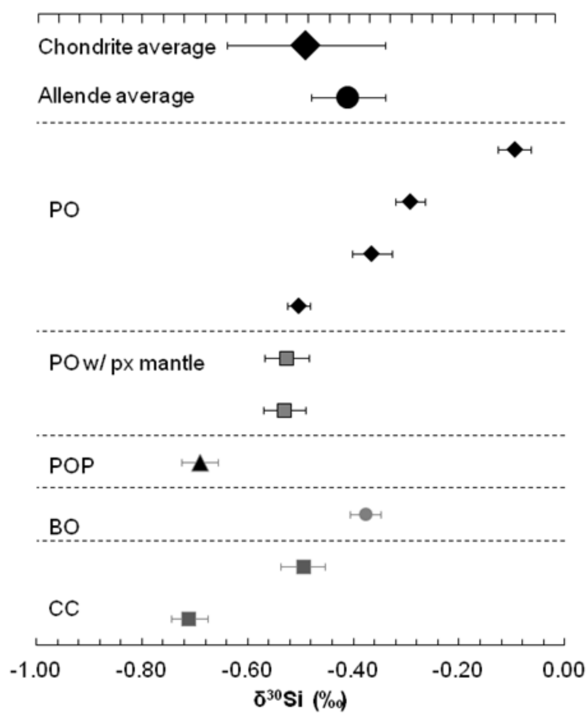

Figure 1 δ^{30}Si of ten Allende chondrules and their petrographic description. PO= porphyritic olivine; px mantle=pyroxene mantle; POP=porphyritic olivine and pyroxene; BO= barred olivine; CC=compound chondrule (containing more than one lithology). The error bars on the chondrules are $\pm2\sigma_{SEM}$. The chondrite average and Allende average (bulk) are from [8] with error bars of $\pm2\sigma_{SD}$.

The Si isotope ratios were measured on a Nu Plasma HR MC-ICP-MS at medium resolution using the sample-standard bracketing technique. The external reproducibility of a chemically processed pure Si standard (Diatomite) and rock standard (BHVO-2) is shown to be <0.15‰ ($2\sigma_{SD}$) for δ^{30}Si. All three stable silicon isotopes (^{30}Si, ^{29}Si, ^{28}Si) were simultaneously measured and fractionations from the standard for all the chondrules were shown to be mass dependent.

Results: The range of $\delta^{30}Si$ compositions for the Allende chondrules was from −0.71‰ to −0.10‰, which is larger than the range currently observed for bulk meteorites (~0.3‰) [8]. However, this is still a limited range in comparison to recent CAI data [11, 12]. There is no consistent correlation between the bulk chondrule $\delta^{30}Si$ with petrological grouping (Fig 1), nor is there any strong inverse correlation with size or Si/refractory element ratio. The average Si isotopic composition of the ten Allende chondrules is $\delta^{30}Si = -0.46 \pm 0.36$ ($2\sigma_{SD}$), which is a similar value to the average of seven aliquots of Allende ($\delta^{30}Si = -0.46 \pm 0.07$, $2\sigma_{SD}$) [8].

Discussion: One mechanism to generate limited non-systematic isotope variation would be if the chondrules formed as closed system objects that inherited their Si isotopic composition from precursor materials. If so, this would make a nebular origin more likely as the bulk compositions of all the meteorites analysed to date show a much more limited range than these Allende chondrules [8]. In addition fire fountaining processes, which are likely in planetary models of chondrule formation, do not appear to measurably fractionate Si isotopes [13]. Potentially these heterogeneities could have been generated by the processes associated with CAI formation, which again would make a nebular origin more likely. The similarity in the average Si isotopic composition of bulk chondrites and chondrules (Fig 1) argues for chondrules being small samplers of very small scale heterogeneity, while chondrites, being larger, average out this same heterogeneity better. This fits with findings from this and previous studies [2, 5, 10], that chondrules display a greater range in Si isotopic composition than bulk chondrites. While the Si isotope data are consistent with closed system formation for chondrules, other evidence from chondrules such as redox state and oxygen isotopic composition [14] is harder to explain in terms of a closed system origin.

The limited fractionation in the bulk chondrule Si data is consistent with other isotope systems, including K [15], Fe [16] and Mg [17], where bulk chondrules show relatively limited fractionations (<1‰ amu^{-1}) when compared with igneous CAIs (~5‰ amu^{-1}). These data, as well as the Si isotopic data from this study, require some alternate explanation for fractionation other than chondrules forming during simple evaporation into free space. The K data has been explained in terms of open-system exchange with a "normal" isotopic reservoir, most likely the evaporated gas [15]. Varying degrees of evaporation and re-condensation, most likely in the solar nebula, could also be a plausible explanation for the measured $\delta^{30}Si$ data if the observed variation is the result of chondrule formation processes rather than inherited heterogeneities.

It is also possible that post-formation processes may have played a role in the creating the measured variation in Si isotopic composition of the bulk chondrules. Allende is known to have undergone thermal metamorphism. However, Si diffuses very slowly, and at the estimated cooling rates for chondrules, the closure temperatures for mm scale objects would be significantly higher than the temperatures reached on the Allende parent body. Aqueous alteration, as the cause of the variation in $\delta^{30}Si$ in bulk chondrules would only be a noteworthy factor if there was significant clay formation, changing the Si isotopic composition of the fluids. While there is evidence for phyllosilicate minerals in the CV3 chondrites [18], it does not explain the variation seen in the LL3 meteorite Chainpur [5]. Secondary alteration processes cannot be completely ruled out contributing to the variation in $\delta^{30}Si$ observed in bulk chondrules; however, it is unlikely to be the primary cause.

Conclusions: The $\delta^{30}Si$ chondrule data do not display an obvious evaporation control as there is no strong inverse correlation with size or Si/refractory element ratio. There are no solid arguments for post-formation processes controlling the variation observed in $\delta^{30}Si$ of the bulk chondrules. The variation in the Si isotopic composition of the chondrules is most likely a nebular process, whether inherited heterogeneities related to CAI formation or evaporation and re-condensation forming the chondrules in the solar nebula.

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