MEASUREMENT OF THE ISOTOPIC COMPOSITIONS OF SIX ELEMENTS IN INDIVIDUAL PRESOLAR SIC GRAINS

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Introduction: Presolar grains have been identified in primitive meteorites on the basis of their extreme isotopic deviations from solar compositions [1,2]. These grains, which formed in the mass-losing envelopes of stars or in supernova ejecta, survived formation of the solar system and retained the isotopic signatures of their parent stars. Approximately 93% of presolar SiC grains originated in carbon-rich Asymptotic Giant Branch (AGB) stars, have heavy elements with *s*-process signatures, and are referred to as "mainstream."

In most previous studies, the isotopic composition of only one heavy element was measured in each grain. In this study, the isotopic compositions of up to six elements (Mo, Zr, Ba, C, N, Si) were measured in each of 55 presolar SiC grains from the Murchison meteorite. Measurement of more than one element in each grain allowed identification of contamination with material of solar system isotopic composition in individual grains and may constrain the amount of ^{13}C in the intershell region between the H- and He-burning shells in parent AGB stars, in which the major source of neutrons for *s*-process nucleosynthesis is ^{13}C(α,n)^{16}O.

Experimental: Ninety-three grains were analyzed from the SiC KJH size fraction, separated from the Murchison meteorite [3]. Heavy element isotopic compositions in each were measured by resonant ionization mass spectrometry (RIMS) with CHARISMA [4]. Standards were terrestrial Mo metal, Zr metal, and $BaTiO_3$. After RIMS analysis, C, N and Si isotopic compositions in each grain were measured by secondary ionization mass spectrometry with the NanoSIMS at Washington University. Standards were synthetic SiC and Si_3N_4 grains.

Results: Fifty-five of the 93 grains yielded sufficient counts to obtain isotopic ratios of at least one heavy element; 30 yielded both Mo and Zr, and 18 yielded Mo, Zr, and Ba. Nine did not show the signature typical of the *s*-procss and were excluded from this study. Carbon, N and Si analysis confirmed that all grains reported here were mainstream except for one B-grain which, as we show below, was contaminated with Ba.

Molybdenum. Molybdenum isotopic ratios could be measured in 47 grains, 39 of which were confirmed to be mainstream by C, N, and Si analysis and were considered in this study. Their Mo isotopic compositions resemble those from previous studies on single grains [5]. When plotted on three-isotope plots normalized to *s*-only ^{96}Mo, they fall along mixing lines between a pure *s*-process component and a component with solar composition. These findings conform to model predictions for 1.5–3 $M_\odot$ AGB stars. The predictions span the range of all possible amounts of ^{13}C in the intershell region of an AGB star, from almost none (d12) to the maximum set by the ^{13}C(*p,γ*)^{14}N reaction (u2).

Zirconium. Zirconium isotopic ratios were measured in 30 of the 47 grains in which Mo was measured; the other 17 were either too small or too low in Zr for isotopic analysis. Previous work on Zr [6] shows that 1.5 and, rarely, 3 $M_\odot$ models best explain the data. As with Mo, model predictions are in excellent overall agreement with the data; all but five of these grains are within ± 2σ analytical errors of model predictions for either 1.5 or 3 $M_\odot$ AGB stars. Those five grains (crosses in Fig. 1) have compositions incompatible with model predictions. For the isotopic compositions of these outliers to be explained by stellar nucleosynthesis, the neutron flux due to the ^{13}C source would have to have been higher during production of Zr than during production of Mo, which is not feasible in light of their being contemporaneously synthesized in the same environment.

The simplest explanation for the outliers is contamination with solar system Mo. When we remove them from the dataset and reexamine a three-isotope Mo plot, it is evident that all remaining grains are within ±2σ of only three ^{13}C pocket amounts: st (the standard case), u1.3 (the standard case times 1.3), and d1.5 (the standard case divided by 1.5), where the standard case is the amount of ^{13}C required to reproduce the *s*-process main component of the solar system in a star of one-half solar metallicity [7]. This contrasts with findings of prior single-element studies in which all possible amounts of ^{13}C considered in the models were required to explain the data.

Barium. Eighteen grains were measured for Ba, Mo and Zr. The Ba results, like those for Mo, are consistent with AGB models for 1.5 $M_\odot$, as were Ba data collected on individual grains in a prior study [8]. Like the Mo findings, the present Ba plots have a larger proportion of strongly *s*-process-enriched grains than was found previously, but the range of compositions still spans the entire set of possible amounts of ^{13}C. A plot of Zr vs. Ba isotopic ratios (Fig. 2) shows seven grains with compositions inconsistent with model predictions. Again, the simplest explanation is solar system Ba contamination. After exclusion of these grains, on a Ba three-isotope plot, it is evident that, as with Mo, all measurements agree with the u1.3, st, and d1.5 cases.

Potential sources of contamination. Although its source has not been identified, solar system contamination remains the most likely explanation for the outlying grains. Opportunities for contact with solar system materials exist during aqueous activity on the meteorite parent body, in the host meteorite, during chemical separation of grains from the meteorite, and when mounting grains on gold foil.

Implications of ^{13}C pocket abundance constraints. Previous single-element measurements showed that the isotopic compositions of mainstream grains are generally consistent with predictions of AGB models and constrained the masses of parent stars to ~1.5–3 $M_{\odot}$. This work may further constrain parent stars to those with intershell ^{13}C amounts within a narrow range around the st case. However, our results do not necessarily indicate that smaller amounts of ^{13}C in the pocket do not exist in stars. It could be that SiC grains do form in AGB stars with pockets with low ^{13}C, but these grains have low concentrations of *s*-process elements such as Zr, Mo and Ba and are therefore not amenable to isotopic analysis. Indeed, the overproduction factors of *s*-process isotopes increase nonlinearly with ^{13}C pocket efficiency. Thus, one expects that grains from AGB stars with pockets low in ^{13}C will be much poorer in isotopes like ^{96}Mo, ^{94}Zr, and ^{136}Ba (the reference *s*-process isotopes in three-isotope plots and Figs. 1 and 2) than will grains from stars with more ^{13}C-rich pockets. We therefore cannot rule out selection bias as the cause of the apparent lack of grains from stars with small (< d1.5) amounts of ^{13}C.

Previous work on mainstream grains has shown that Sr isotopic ratios are not fully consistent with AGB model predictions and require the full range of ^{13}C pocket amounts [9]. Our work suggests the possibility that some grains are contamined with solar system Sr. This can be tested by multi-element isotopic analysis including Sr in individual grains.

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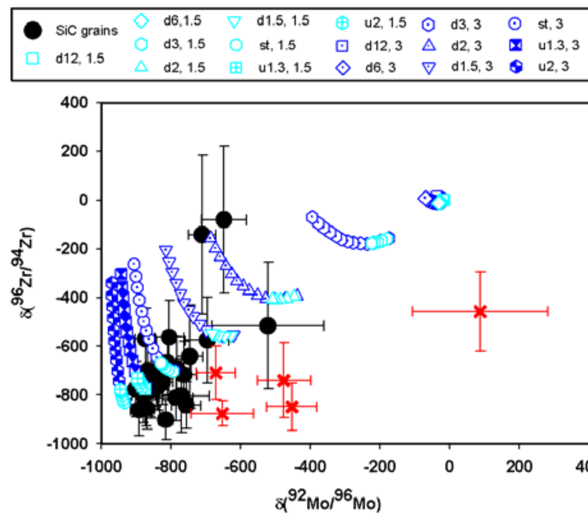

Figure 1. Two-element plot of Mo and Zr isotopic ratios in mainstream SiC grains compared with model predictions for 1.5 and 3 $M_{\odot}$ carbon-rich AGB stars with solar initial metallicity (open symbols). Filled circles are individual grains within 2σ of predictions. Crosses are grains believed to have been contaminated with Mo but not Zr.

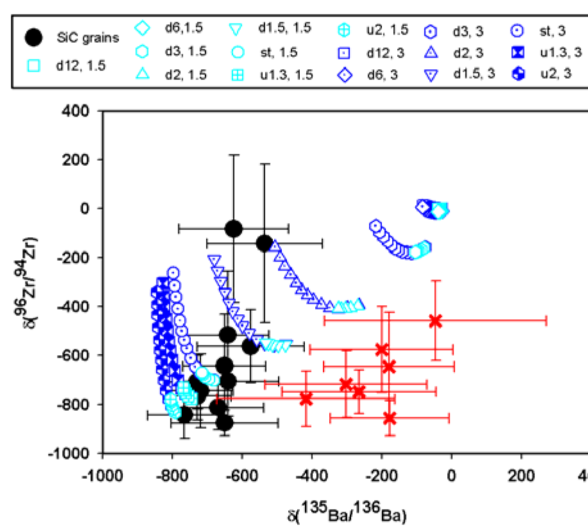

Figure 2. Two-element plot of Ba and Zr isotopic ratios in mainstream SiC grains compared with model predictions for 1.5 and 3 $M_{\odot}$ carbon-rich AGB stars with solar initial metallicity (open symbols).