EXTINCT ^{93}Zr IN SINGLE PRESOLAR SiC GRAINS AND CONDENSATION FROM ZIRCONIUM-DEPLETED GAS.

Y. Kashiv[1,2,3,8], A. M. Davis[1,2,3], Z. Cai[6], B. Lai[6], S. R. Sutton[1,3,4], R. S. Lewis[2,3], R. Gallino[7] and R. N. Clayton[1,2,3,5], [1]Department of the Geophysical Sciences, [2]Enrico Fermi Institute, [3]Chicago Center for Cosmochemistry, [4]Consortium for Advanced Radiation Sources, [5]Department of Chemistry, University of Chicago, Chicago, IL 60637; [6]Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439, [7]Dipartimento di Fisica Generale, Università di Torino, I-10025 Torino, Italy. ([8]yoavk@geosci.uchicago.edu)

Introduction: Eighty five percent of ^{93}Nb, the only stable isotope of niobium, is produced by *s*-process nucleosynthesis in low mass asymptotic giant branch (AGB) stars as radioactive ^{93}Zr ($t_{1/2} = 1.5\times10^6$ y) [1]. Due to its short half-life compared to the lifetime of low mass stars (~10^9 y), ^{93}Zr in stars must be synthesized locally and cannot be inherited from other stars (the AGB phase occurs at the end of the star's life). On the other hand, the half-life of ^{93}Zr is long compared with the total time of the AGB phase (~10^6 y [2]) and the timescale for condensation of a ~1 µm SiC grain in the stellar wind (a few years [3]). This means that ^{93}Zr was still alive at the time grains condensed in the source AGB stars. Hence, evidence for live ^{93}Zr in mainstream presolar SiC grains would further strengthen their association with AGB stars. Here, we evaluate Zr and Nb concentration measurements in individual presolar SiC grains and their implications for *in situ* decay of ^{93}Zr. The data were collected by synchrotron x-ray fluorescence; preliminary analytical results and experimental details have already been presented [4].

Results and Discussion: *Extinct ^{93}Zr in the grains.* Niobium abundances, expressed as enrichment factors relative to Si and CI chondrites, are plotted versus zirconium abundances in Fig. 1. The isotopic composition of the grains was not measured, so it was assumed that the grains are mainstream grains (which make up ≈90% of all SiC grains [5]). The ion microprobe results of Amari et al. [6] for mainstream grains are plotted as well. The experimental data are compared with predictions of elemental abundances in SiC grains condensing from the envelope of a 1.5 $M_\odot$ AGB star of initially solar metallicity and composition. In plotting the results of the calculations it was assumed that 100% of zirconium and niobium and 50% of silicon condensed into the grains. SiS is a very stable gas phase species, so only silicon in excess of the abundance of sulfur is available to condense as SiC [7]. New solar system zirconium and niobium abundances, based on recent measurements in meteorites [8], were derived and used in the calculations and to calculate the enrichment factors (Zr = 10.26 and Nb = 0.7572, in units of atoms per 10^6 Si atoms).

Two arrays of calculations are plotted in Fig. 1. One array, labeled "Nonradiogenic array", represents only the niobium present in the stellar envelope at the time of SiC condensation, without any subsequent radiogenic contribution from ^{93}Zr. The second array, labeled "Radiogenic array", represents the expected niobium abundance in the grains after ^{93}Zr has fully decayed. As can be seen in Fig. 1, the two experimental data sets agree with one another (except for one grain with high Zr/Nb in the ion probe data [6]), and both agree with the radiogenic array.

The good agreement between the grain data and the calculated radiogenic array, and the correlation between niobium and zirconium, combined with the relatively short ^{93}Zr half-life, strongly suggest that freshly synthesized *s*-process ^{93}Zr condensed into the SiC grains as they were forming in the winds from low mass AGB stars, and that most of the niobium in the grains is the product of *in situ* decay of ^{93}Zr. This result strengthens the identification of low mass AGB stars as being both the source stars of presolar mainstream SiC grains and an *s*-process site. In addition, it adds to the recent evidence for condensation of *s*-process ^{99}Tc ($t_{1/2} = 2.13\times10^5$ y) [9] and lack of condensation of volatile *s*-process ^{135}Cs ($t_{1/2} = 2.3\times10^6$ y) [10] into the grains.

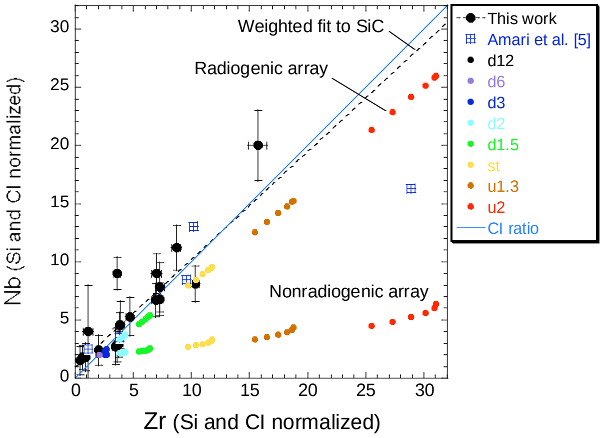

Figure 1. *Nb vs. Zr abundances in single mainstream SiC grains compared with predictions of abundances for SiC grains condensed from the envelope of a 1.5 $M_\odot$ AGB star. d12,…,u2 are the different ^{13}C pocket amounts (for more details see [10]). Uncertainties are 2σ.*

Grain condensation from zirconium-depleted gas. Another interesting observation is that most of the experimental data points of this study (15 of 19 grains) plot above the radiogenic array in Fig. 1. A linear fit to these data point, weighted by the uncertainties in the Nb abundances (which are much larger than the uncertainties in the Zr abundances), is plotted as a dashed curve. This curve is within uncertainty parallel to and displaced above the radiogenic array. The most likely explanation for the displacement of the data points above and parallel to the radiogenic array, and to the spread of the data points, including the extreme points, is condensation of the SiC grains from zirconium-depleted stellar gas (the displacement is greater than potential systematic experimental error).

Depletion of zirconium is expected from equilibrium thermodynamic calculations of condensation and has experimental support. Zirconium is calculated to be one of the first elements to condense, as ZrC, under C/O > 1 conditions [7]. Once ZrC condensed, it could have been removed from the gas before SiC condensation by the two abundant grain phases, TiC and graphite, which, both have condensation temperatures (T_{cond}) ~200 K below the T_{cond} of ZrC and ~150 K above the T_{cond} of SiC [7]. Indeed, subgrains of ZrC, usually in solid solution with titanium, molybdenum and ruthenium carbides, were detected in graphite grains from carbon-rich AGB stars [11,12].

The problem with this scenario is that niobium, like zirconium, is a very refractory element. Equilibrium thermodynamic calculations predict that NbC will condense at a temperature ~40 K below the T_{cond} of ZrC (taking into account the *s*-process enrichment of zirconium) and ~150 K higher than the T_{cond} of TiC and graphite [7]. This means that, like ZrC, NbC might be expected to condense and be removed from the stellar gas by TiC and graphite. However, NbC subgrains have not been detected to date in graphite grains from AGB stars [11,12]. The reason may be that the kinetics of growth of NbC grains was not fast enough. Bernatowicz et al. [13] showed that the kinetics of growth of graphite, TiC and ZrC grains in spherically symmetric outflows from AGB stars are too slow to allow growth to the grain sizes measured in the lab [11,12] and hence the grains must have condensed in anisotropic outflows. In the case of niobium, it may be that due to its low abundance in the envelopes of these stars, down to 1/80 of the zirconium abundance (atomic), significant condensation of NbC is too slow and as a consequence niobium was not depleted in the gas.

Zirconium depletion from the gas, without niobium depletion, produces radiogenic arrays that are approximately parallel to the one in Fig. 1 and displaced above it to an extent that is proportional to the degree of depletion. Thus, condensation of SiC grains from stellar gas with varying degrees of zirconium depletion, corresponding to different stars and/or different stages in the AGB phase of the same star(s), generates the observed pattern of grains and accounts for the spread in their Nb/Zr ratios. The measured grain data could be explained by applying this scenario to AGB stars in the mass range of ~1.5−3 $M_\odot$, which agrees with the inferred mass range based on the isotopic composition of heavy trace elements in the grains [10]. An example of this scenario is shown in Fig. 2 for a 2 $M_\odot$ AGB star. Hypothetical radiogenic arrays are plotted for cases where 50%, 80%, 90% and 95% of zirconium was lost from the gas by the time SiC started to condense (the 0% zirconium-loss curve is identical to the radiogenic array in Fig. 1). As can be seen, the measured compositions of most grains could be explained by these curves. A combination of similar curves for stars of different masses (in the ~1−3$M_\odot$ range) could reproduce all of the measured grains' compositions.

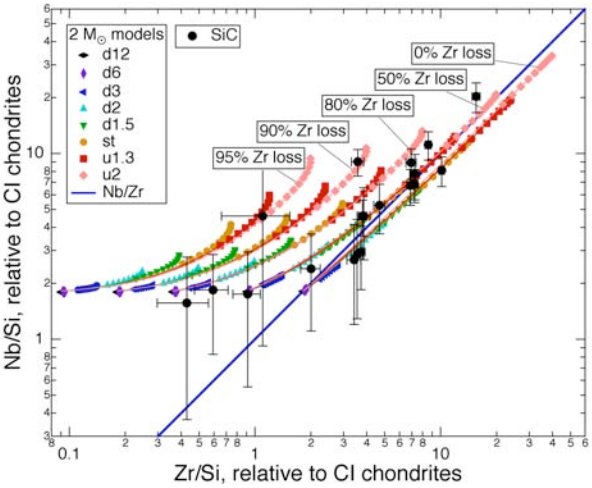

Figure 2. *Radiogenic arrays for SiC grains condensing from gas of a 2$M_\odot$ AGB star with varying degrees of loss of zirconium by prior ZrC condensation. Uncertainties are 2σ. Note that this is a log-log plot.*

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