

HEAVY METAL ISOTOPIC ANOMALIES IN SUPERNOVAE PRESOLAR GRAINS. M. J. Pellin^{1,2}, M. R. Savina^{1,2}, W. F. Calaway^{1,2}, C. E. Tripa¹, J. G. Barzyk^{1,2,3}, A. M. Davis^{2,3,4}, F. Gyngard⁵, S. Amari⁵, E. Zinner⁵, R. S. Lewis^{2,4}, R. N. Clayton^{2,3,4,6}. ¹Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, ²Chicago Center for Cosmochemistry, Chicago, IL 60637, ³Department of the Geophysical Sciences, University of Chicago, Chicago, IL 60637, ⁴Enrico Fermi Institute, University of Chicago, Chicago, IL 60637, ⁵Laboratory for Space Sciences, Washington University, St. Louis, MO 63130, ⁶Department of Chemistry, University of Chicago, Chicago, IL 60637

Introduction: Presolar SiC grains, isolated from acid residues of primitive meteorites, are ejecta of stars that contributed to the protosolar nebula. Due in part to SiC's refractory, chemically inert nature, these grains have survived solar system formation to provide an isotopic record of the nuclear processing in their parent stars. Among these grains are a rare fraction (~1%), called type X, which are believed, based on a concordance between grain isotopic abundances and type II supernova (SNII) model predictions [1-3], to have formed in the stellar outflows of SNII explosions. X-grains are characterized by enrichments (relative to solar isotopic abundances) in ¹²C (most grains), ¹⁵N, and ²⁸Si. Many of these grains have isotopic overabundances in ²⁶Mg [4-6], ⁴⁴Ca [7], and ⁴⁹Ti [8, 9] that are likely to have come from the radioactive decay of ²⁶Al ($t_{1/2}$ ~0.7 Myr), ⁴⁴Ti ($t_{1/2}$ ~60 yr), and ⁴⁹V ($t_{1/2}$ ~330 d) after grain formation. Here, the isotopic compositions of C, N, Si, Fe, Sr, Zr, Mo, Ru, and Ba are determined in individual X-grains. Surprisingly, the isotopic patterns measured are not consistent a canonical *r*-process nucleosynthesis, but rather correspond to models producing a rapid, but *limited* neutron dose [10, 11].

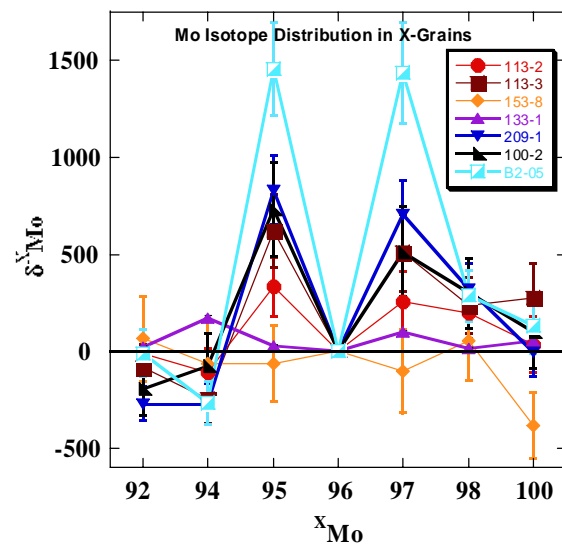
Experimental: Two separate SiC grain mounts from the Murchison meteorite (size separate KJG and KJH) were prepared. Secondary ion mass spectrometry (SIMS) analysis was used to identify the grains of type X on each mount [12]. Heavy element isotopic compositions in each were measured by resonant ionization mass spectrometry (RIMS) with CHARISMA [13]. Standards were terrestrial Mo, Zr, Sr, Ru metal and BaTiO₃. C, N and Si isotopic compositions in each grain were measured by SIMS at Washington University. Standards were synthetic SiC and Si₃N₄ grains.

Results: Twelve SiC grains were identified as type X. The results of isotopic analyses are displayed in Table 1. Consider in turn the results for Mo, Zr, and Ba.

Molybdenum. Molybdenum isotopic ratios were measured in 7 presolar grains of type X (an eighth has been included because of its similar isotopic ratio, though light element isotopic analysis was not possible). Additional Mo isotopic measurements of X-grains demonstrate that the unusual Mo isotopic pattern noted previously [14], enrichment relative to ⁹⁶Mo (a pure s-process

isotope) in ⁹⁵Mo, ⁹⁷Mo, a smaller enrichment ⁹⁸Mo. Surprisingly, no clear signature of ¹⁰⁰Mo enhancement has been seen in X-grains. Figure 1 displays the situation. These results are consistent with a proposed neutron burst nucleosynthesis [10, 11] for these isotopes. The signature is also evident in the supernovae models of Rauscher et al. [1] at the outer edge of the oxygen shell. Moreover the Mo abundance in this shell is sufficient to account for the observed isotopic patterns.

Figure 1: Mo isotopic pattern for 7 SiC grains of type X.

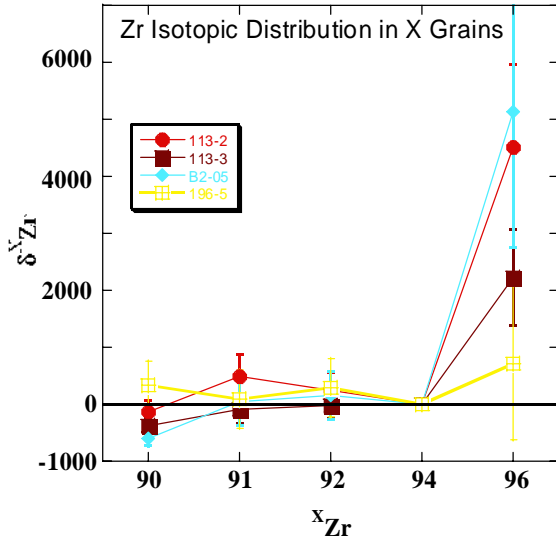


The observed variations in the magnitude of the isotopic anomalies may have several sources. Extensive mixing of the supernova ejecta must be present in order to account for the formation of SiC grains and for their isotope ratios. The heterogeneity of the isotopic production of Mo in different shells of the supernovae combined with incomplete mixing could well yield the variation observed. It is also possible that Mo within the grains has been contaminated with solar Mo as has been suggested for AGB grains at this conference.

Zirconium. The X-grains measured are significantly enhanced in ⁹⁶Zr relative to ⁹⁴Zr while ⁹⁰Zr is depleted. The enhancement in ⁹⁶Zr is consistent with the neutron burst model (the ⁹⁵Mo and ⁹⁷Mo enhancement originate from ⁹⁵Zr and ⁹⁷Zr enhancements trapped as the neutron dose ends). While supernovae models also see this isotope pattern at the outer edge of the oxygen shell, the situation here is more

complicated since the abundance of Zr in this shell is not sufficient to alter the whole star isotopic abundance patterns. The variation in Zr isotopic enrichments may be a result of the small statistics. It seems less likely that contamination is responsible based on results presented at this conference.

Figure 2: Zr isotopic pattern for 4 SiC grains of type X.



Barium. Enrichments measured in ^{138}Ba relative to ^{136}Ba (a pure s-process isotope) are indicative of a neutron density high enough to drive through the several s-process branches above ^{133}Cs , the most difficult of which is at ^{136}Cs ($T_{1/2} = 13.16$ d). This interpretation is strengthened by depletions in ^{135}Ba that demonstrate flow through ^{134}Cs ($T_{1/2} = 2.065$ a) and ^{135}Cs ($T_{1/2} = 2.3$ Ma). In contrast, the r-process is expected to give enhanced $\delta^{135}\text{Ba}$, $\delta^{137}\text{Ba}$ and $\delta^{138}\text{Ba}$ values.

Ruthenium and Fe. Two grains have been examined for Ru. One, 322-1, is a demonstrated grain of type X. The other is included based on its isotopic similarity to 322-1. Both grains show a ^{100}Ru deficit. This pattern is not consistent with current supernova models. For Fe three grains have been examined two of them show an ^{58}Fe excess.

Conclusions: We are continuing our study of isotopic enhancements in SiC grains of type X. The results continue to be consistent with a neutron burst signature. This same signature has been inferred in whole rock meteorite studies [15-17].

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Table 1. Heavy metal isotopic abundances for SiC grains of type X (2 σ error).

Grain	$\delta^{13}\text{C}$	$\delta^{15}\text{N}$	$\delta^{29}\text{Si}$	$\delta^{30}\text{Si}$	$\delta^{\text{E}}\text{E} = \left(\frac{[X_{\text{E}}]/[E_{\text{E}}]}{[X_{\text{E}}]/[E_{\text{E}}]} - 1 \right)$ for C Y=12; for N Y=14 for Si Y=28; for Sr Y=86 for Zr Y=94; for Mo Y=96 for Ru Y=100 for Ba Y=136; for Fe Y=56	
113-2	-421 \pm 10	3734 \pm 82	-223 \pm 4	-378 \pm 6		
113-3	-856 \pm 7	2877 \pm 132	-320 \pm 5	-445 \pm 7		
153-8	-765 \pm 16	4023 \pm 106	-355 \pm 8	-585 \pm 7		
196-5	-678 \pm 19	3881 \pm 139	-246 \pm 8	-403 \pm 9		
133-1	-798 \pm 7	7479 \pm 154	-299 \pm 5	-416 \pm 7		
209-1	-708 \pm 11	2570 \pm 243	-309 \pm 5	-565 \pm 5		
100-2	-542 \pm 15	4440 \pm 103	-263 \pm 7	-407 \pm 8		
322-1	-508 \pm 23	3001 \pm 180	-211 \pm 10	-338 \pm 11		
327-5	-484 \pm 19	3015 \pm 98	-366 \pm 6	-440 \pm 7		
032-2	-852 \pm 5	7248 \pm 428	-55 \pm 6	-89 \pm 10		
B2-05	83 \pm 1	63 \pm 0	-188 \pm 6	-331 \pm 5		
Grain	$\delta^{87}\text{Sr}$	$\delta^{88}\text{Sr}$	$\delta^{90}\text{Zr}$	$\delta^{91}\text{Zr}$	$\delta^{92}\text{Zr}$	$\delta^{96}\text{Zr}$
113-2	-157 \pm -15	2652 \pm 91	-134 \pm 192	494 \pm 382	243 \pm 315	4510 \pm 1447
113-3	200 \pm 65	2456 \pm 242	-373 \pm 125	-96 \pm 231	-17 \pm 226	2223 \pm 855
196-5			325 \pm 428	95 \pm 510	290 \pm 512	723 \pm 1334
B2-05			-591 \pm 152	44 \pm 412	153 \pm 428	5140 \pm 2382
Grain	$\delta^{92}\text{Mo}$	$\delta^{94}\text{Mo}$	$\delta^{95}\text{Mo}$	$\delta^{97}\text{Mo}$	$\delta^{98}\text{Mo}$	$\delta^{100}\text{Mo}$
113-2	-10 \pm 120	-110 \pm 127	331 \pm 152	253 \pm 161	194 \pm 130	34 \pm 141
113-3	-87 \pm 119	-250 \pm 118	622 \pm 190	507 \pm 197	236 \pm 141	276 \pm 175
153-8	63 \pm 218	-62 \pm 225	-65 \pm 198	-104 \pm 217	50 \pm 202	-382 \pm 170
133-1	23 \pm 91	172 \pm 121	29 \pm 94	98 \pm 112	17 \pm 86	53 \pm 110
209-1	-271 \pm 90	-273 \pm 103	832 \pm 181	703 \pm 182	320 \pm 130	-4 \pm 125
100-2	-196 \pm 136	-78 \pm 168	731 \pm 241	513 \pm 232	298 \pm 181	99 \pm 187
B2-05	-9 \pm 120	-265 \pm 111	1459 \pm 242	1437 \pm 261	285 \pm 131	131 \pm 144
E2-10	-350 \pm 116	-229 \pm 151	1437 \pm 315	4031 \pm 632	-662 \pm 64	1528 \pm 354
Grain	$\delta^{96}\text{Ru}$	$\delta^{98}\text{Ru}$	$\delta^{99}\text{Ru}$	$\delta^{101}\text{Ru}$	$\delta^{102}\text{Ru}$	$\delta^{104}\text{Ru}$
322-1	5275 \pm 1670	2491 \pm 1345	2344 \pm 855	1188 \pm 568	2240 \pm 788	4727 \pm 1385
H-2	900 \pm 228	695 \pm 303	156 \pm 124	154 \pm 117	220 \pm 116	161 \pm 119
Grain	$\delta^{135}\text{Ba}$	$\delta^{137}\text{Ba}$	$\delta^{138}\text{Ba}$	$\delta^{54}\text{Fe}$	$\delta^{57}\text{Fe}$	$\delta^{58}\text{Fe}$
113-2	-495 \pm 280	-241 \pm 310	1890 \pm 893	-128 \pm 48	185 \pm 94	454 \pm 281
113-3	-418 \pm 451	843 \pm 945	3808 \pm 2150	-229 \pm 45	455 \pm 105	806 \pm 314
153-8	452 \pm 474	34 \pm 324	-141 \pm 219	187 \pm 99	218 \pm 154	321 \pm 424
133-1	-204 \pm 70	-27 \pm 71	306 \pm 75			
327-5	162 \pm 146	-27 \pm 112	-49 \pm 88			
032-2	-534 \pm 147	-108 \pm 198	-198 \pm 141			

t == terrestrial isotopic abundance
g == isotopic abundance for a particular grain

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