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# TOWARD COMPLETE ISOTOPIC ANALYSIS OF INDIVIDUAL PRESOLAR SILICON CARBIDE GRAINS: C, N, Si, Sr, Zr, Mo, AND Ba IN SINGLE GRAINS OF TYPE X*


M. J. Pellin[1], W. F. Calaway[1], A. M. Davis[2,3], R. S. Lewis[2], S. Amari[4], and R. N. Clayton[2,3,5]

[1]Materials Science and Chemistry Divisions, Argonne National Laboratory, Argonne, IL 60439
[2]Enrico Fermi Institute, The University of Chicago, Chicago, IL 60637
[3]Department of the Geophysical Sciences, The University of Chicago, Chicago, IL 60637
[4]McDonnell Center for the Space Sciences, Washington University, St. Louis, MO 63130
[5]Department of Chemistry, The University of Chicago, Chicago, IL 60637,



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**TOWARD COMPLETE ISOTOPIC ANALYSIS OF INDIVIDUAL SILICON CARBIDE GRAINS: Ba, C, Mo, N, Si, Sr, AND Zr IN SINGLE GRAINS OF TYPE X.** M. J. Pellin[1], W. F. Calaway[1], A. M. Davis[2,3], R. S. Lewis[2], S. Amari[5] and R. N. Clayton[2,3,4], [1]Materials Science and Chemistry Divisions, Argonne National Laboratory, Argonne, IL 60439, [2]Enrico Fermi Institute, [3]Department of the Geophysical Sciences, [4]Department of Chemistry, University of Chicago, Chicago, IL 60637, [5]McDonnell Center for the Space Sciences, Washington University, St. Louis, MO 63130 (pellin@anl.gov; a-davis@uchicago.edu; royl@rainbow.uchicago.edu; sa@howdy.wustl.edu; r-clayton@uchicago.edu)

**Introduction:** Presolar silicon carbide grains form in a variety of types of stars, including asymptotic giant branch red giant stars and supernovae. The dominant mechanisms of heavy element nucleosynthesis, the *s*-process and *r*-process, are thought to occur mainly in AGB stars and supernovae, respectively [1]. We have previously reported that mainstream SiC grains have strong enrichments in the *s*-process isotopes of Sr, Zr and Mo [2–4] and initial results for X-type SiC grains showing enrichments in the *r*-process isotopes of Mo[5]. We report here the first measurements of Zr, Sr, and Ba isotopes along with expanded studies of Mo in individual X-type SiC grains, which have previously been identified as having formed from supernova ejecta.

**Experimental methods:** Approximately 2000 grains of Murchison SiC grain size separate KJG [6] were dispersed onto a soft gold mount. 11 X-grains were identified by ion imaging [7] with the Washington University ion microprobe. Si, C and N were measured by ion microprobe in these grains using a $Cs^+$ primary ion beam. The mount was then characterized by electron imaging and x-ray mapping with an SEM at the University of Chicago, identifying all grains on the mount.

The isotopic compositions of Mo and Zr in a number of grains were measured by laser ablation laser resonant ionization mass spectrometry using the CHARISMA instrument at Argonne National Laboratory. The analytical methods were similar to those used previously [4]. Ba was measured for the first time in single SiC grains using 18060.3 cm$^{-1}$ and 24798.1 cm$^{-1}$ beams to resonantly ionize the atom ($^1S_0 \rightarrow {}^1P_1^O \rightarrow$ionization continuum). The addition of a third laser at 21698.5 cm$^{-1}$ allowed the simultaneous photoionization and measurement of Sr ($^1S_0 \rightarrow {}^1P_1^O \rightarrow$ionization continuum) minimizing sample consumption and allowing relative concentration measurements to be made.

**Results:** We report here the heavy element isotopic compositions of seven SiC grains of type X. In all of these grains the C, N, and Si isotopic compositions along with at least one heavy element isotopic composition have been determined. Remarkably, two of the grains were sufficiently large to allow determination of all four heavy elements (Ba, Mo, Sr, and Zr).

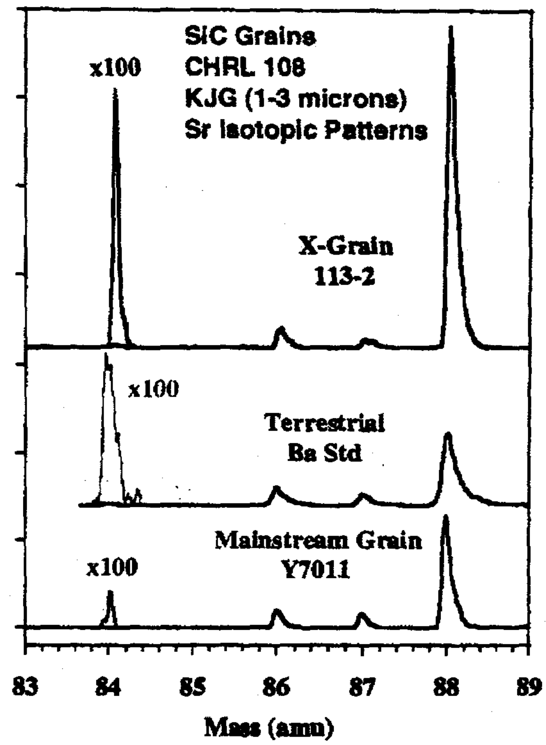


*Fig. 1. Ba Isotopic Mass Spectra of an X-Grain, a Terrestrial Standard and a Mainstream Grain. Spectra are normalized to the height of $^{136}$Ba. Note a large noise peak due to $^{133}$Cs has been removed for clarity.*

Mass spectra of a terrestrial standard, one of these remarkable X-grains and a mainstream grain are compared in Fig. 1 and 2 for their Sr and Ba isotopic compositions. Unfortunately, in the Ba spectra, a large $^{133}Cs^+$ implanted during the C, N and Si isotopic measurements, makes $^{134}$Ba determintion problematic. It is immediately apparent that significant differences exist between the terrestrial, mainstream and x-grain patterns for both Ba and Sr. The most significant difference is the large excesses in $^{88}$Sr and $^{138}$Ba for the X-grain relative either to terrestrial or to mainstream. There are also increases in the *p*-process isotopes $^{84}$Sr and $^{132}$Ba. While these represent very few total counts, they are the first significant increases in *p*-process isotopes that we have measured. Unfortunately, overlap from an implanted $^{133}Cs^+$ peak makes analysis of $^{134}$Ba problematic.

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**Discussion:** *Zr.* Table 1 demonstrates that the X-Grains measured demonstrate significant enhancement in $^{96}$Z relative to $^{94}$Z (the most predominantly s-process isotope of Zr). The origin of $^{96}$Zr-enriched isotopic patterns is not clear. $^{96}$Zr excesses are to be expected from the *r*-process, but they can also be produced in the *s*-process if the neutron density is high enough to drive through the *s*-process branch at $^{95}$Zr ($T_{1/2}$ = 65 d).

*Sr.* Enrichments measured in $^{88}$Sr relative to $^{86}$Sr (a pure *s*-process isotope) is indicative of a neutron density high enough to drive through the *s*-process branch at $^{73}$Kr ($T_{1/2}$ = 10.73 a).

*Ba.* Enrichments measured in $^{138}$Ba relative to $^{136}$Ba (a pure *s*-process isotope) is indicative of a neutron density high enough to drive through the several *s*-process branches above $^{133}$Cs, the most difficuly 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).

*Mo.* Expanded Mo isotopic measurements of X-grains generally demonstrates that, as determined previously [5], enrichment relative to $^{96}$Mo (a pure *s*-process isotope) in all isotopes that can be produced by the *r*-process ($^{95}$Mo, $^{97}$Mo, $^{98}$Mo and in some cases $^{100}$Mo). However, the largest enrichments are measured for $^{95}$Mo and the enrichment at $^{100}$Mo is significantly smaller if present at all. For these grains then the neutron density was not or only marginally high enough to drive through the *s*-process branch at $^{99}$Mo ($T_{1/2}$ = 2.75 d).

**Conclusion:** The X-Grains measured here experienced a neutron density higher than produced in normal AGB stars, but lower than might be expected in the center of a typical supernovae.

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**References:** [1] Wallerstein G. et al. (1997) *Rev. Modern Phys., 69,* 995–1084. [2] Nicolussi G. K. et al. (1997) *Science, 277,* 1281–1283. [3] Nicolussi G. K. et al. (1998) *GCA, 62,* 109–1104. [4] Nicolussi G. K. et al. (1998) *Phys. Rev. Lett., 81,* 3583–3586. [5] Pellin, M. J. et al. (1999) *LPSC, XXX,* 1969-1970. [6] Amari S. et al. (1994) *GCA, 58,* 459–470. [7] Nittler L. R. et al. (1995) *Ap. J., 453,* L25–L28. [8] Amari S. et al. (1999) *Ap. J.,* submitted. [9] Amari S. et al. (1992) *Ap. J., 394,* L43–L46. [10] Hoppe P. et al. (1996) *Science, 272,* 1314–1316. [11] Nittler L. R. et al. (1996) *Ap. J., 462,* L31–L34. [12] Clayton D. D. et al. (1997) *Ap. J., 486,* 824–834. [13] Zinner E. (1998) *Ann. Rev. Earth Planet. Sci. 26,* 147–188.

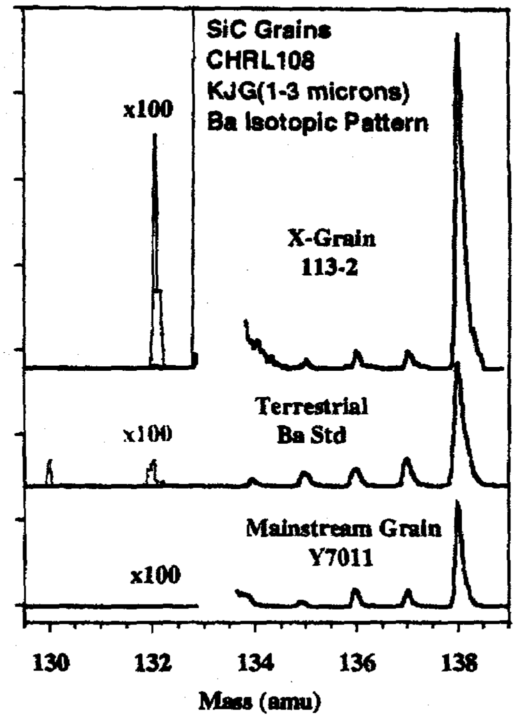


*Fig. 2. Ba Isotopic Mass Spectra of an X-Grain, a Terrestrial Standard and a Mainstream Grain. Spectra are normalized to the height of $^{136}$Ba. Note a large noise peak due to $^{133}$Cs has been removed for clarity.*

Table 1. C, N, Si, Mo, Zr, Sr, and Ba isotopes in SiC grains of type X in CHRL108.

| Grain | $\delta^{13}$C | $\delta^{15}$N | $\delta^{29}$Si | $\delta^{30}$Si | $\delta^{92}$Mo |
|---|---|---|---|---|---|
| 113-2 | -421±10 | 3734±82 | -223±4 | -378±6 | -10±120 |
| 113-3 | -856±7 | 2877±132 | -320±5 | -445±7 | -87±119 |
| 209-1 | -708±11 | 2570±243 | -309±5 | -565±5 | -271±90 |
| 100-2 | -542±15 | 4440±103 | -263±7 | -407±8 | -196±136 |
| 153-8 | -765±16 | 4023±106 | -355±8 | -585±7 | 63±218 |
| 196-5 | -678±19 | 3881±139 | -246±8 | -403±9 | |

| Grain | $\delta^{94}$Mo | $\delta^{95}$Mo | $\delta^{97}$Mo | $\delta^{98}$Mo | $\delta^{100}$Mo |
|---|---|---|---|---|---|
| 113-2 | -110±127 | 331±152 | 253±161 | 194±130 | 34±141 |
| 113-3 | -250±118 | 622±190 | 507±197 | 236±141 | 276±175 |
| 209-1 | -273±103 | 832±181 | 703±182 | 320±130 | -4±125 |
| 100-2 | -78±168 | 731±241 | 513±241 | 298±181 | 99±187 |
| 153-8 | -62±225 | -65±198 | -104±217 | 50±202 | -382±170 |

| Grain | $\delta^{90}$Zr | $\delta^{91}$Zr | $\delta^{92}$Zr | $\delta^{96}$Zr |
|---|---|---|---|---|
| 113-2 | -134±192 | 494±382 | 243±315 | 4510±144 |
| 113-3 | -373±125 | -96±231 | -17±226 | 2223±855 |
| 196-5 | 325±428 | 95±510 | 290±512 | 723±1334 |

| Grain | $\delta^{84}$Sr | $\delta^{87}$Sr | $\delta^{88}$Sr | $\delta^{130}$Ba |
|---|---|---|---|---|
| 113-2 | 106±256 | -146±96 | 2669±254 | -996±4122 |
| 113-3 | 4698±2363 | -1±370 | 1914±696 | -991±9029 |

| Grain | $\delta^{132}$Ba | $\delta^{135}$Ba | $\delta^{137}$Ba | $\delta^{138}$Ba |
|---|---|---|---|---|
| 113-2 | 7243±5033 | -495±140 | -241±155 | 1890±446 |
| 113-3 | 8029±7272 | -418±225 | 843±473 | 3808±1075 |