Indarch SiC by TIMS, RIMS, and NanoSIMS. C. L. Jennings[1], M. R. Savina[2], S. Messenger[3], S. Amari[3], R. H. Nichols, Jr.[1], M. J. Pellin[2], and F. A. Podosek[1], [1]Department of Earth and Planetary Sciences, Washington University, St. Louis, MO, 63130, [2]Materials Science Division, Argonne National Laboratory, Argonne, IL 60439, [3]McDonnell Center for Space Sciences, Washington University, St. Louis, MO, 63130.

Introduction: Silicon carbide is among the most abundant types of presolar grains in meteorites, reaching, for example, 5.8 ppm in Indarch (EH4) [1]. Isotopic studies have shown that these grains are from a variety of stellar sources. Mainstream grains (~95% of presolar SiC) exhibit enhancements in s-process isotopes and there is abundant evidence that they formed around low mass AGB stars [2]. The less well known A and B type grains (~3-4%) are defined as having $^{12}C/^{13}C < 10$, with B grains in particular having $^{12}C/^{13}C > 3.5$ [3]. Here we report isotopic measurements of SiC grains from Indarch by mass spectrometry: thermal ionization (TIMS), resonant ionization (RIMS), and secondary ion (SIMS) mass spectrometry.

Experimental: The samples were prepared following the method used to produce the KJ series from Murchison [4], with the exceptions that CS_2 was used instead of NaOH to dissolve sulfur, and sulfuric acid was not used to dissolve spinel because of its low abundance. The residue was separated by centrifugation into five grain-size fractions. The largest grain-size fraction was examined by SEM and found to be about half SiC and half Si_3N_4 by grain counting. Si_3N_4 is much more abundant in these Indarch separates than in the Murchison KJ series because Indarch is rich in Si_3N_4 of solar system origin. However, a small fraction of Indarch Si_3N_4 grains have been shown to be presolar [5].

For TIMS measurements, aliquots of each grain-size fraction were direct-loaded with 1/2 µl boric acid on V-shaped Re filaments. No corrections were made for instrumental discrimination, since this requires a known ratio and all of the measured values were highly anomalous. Uncertainties due to lack of discrimination correction are much lower than the anomalies seen in the grain aggregates.

Eighteen single ~1-5 µm grains were handpicked from the largest grain-size fraction and pressed into a gold substrate for RIMS and SIMS analyses. The Ba isotopic compositions were determined by RIMS on the CHARISMA instrument, which is described in detail elsewhere [6]. The C and N isotopic compositions of eight grains that remained after RIMS analysis were subsequently measured on the new Washington University NanoSIMS in multicollection mode using procedures similar to those previously described [7].

Results and discussion: The results are comparable to the previous measurements of SiC aggregates [2,8,9]. The Ba isotopic composition of the Indarch grain-size fractions were consistent with s-process nucleosynthesis. The isotopes made only by p-process, ^{130}Ba and ^{132}Ba, were depleted relative to normal composition while the others were enhanced to varying degrees. This is to be expected for SiC separates dominated by mainstream SiC grains, which are believed to originate in AGB stars [2,10].

The s-process composition was calculated by the same method as in [9]. In brief, a three-isotope plot of $^{132}Ba/^{136}Ba$ vs. $^{135}Ba/^{136}Ba$ ratios was used to find the composition of $^{135}Ba/^{136}Ba$ when $^{132}Ba/^{136}Ba$ is forced to zero. This yields the composition of $^{135}Ba/^{136}Ba$ ratio when the p-only isotope ^{132}Ba is removed from the mixture. This composition was used to calculate all the other s-process compositions by the same method. The results (Table 1) are in agreement with the previous work, especially for the most fine-grained sample, which is expected to be the most pure separate of Indarch SiC, and also the most enhanced in s-process isotopes [3,9].

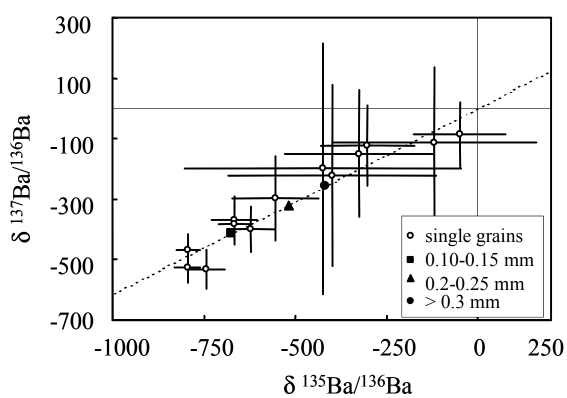

Fig. 1. Ba isotopic compositions (in permil deviations from normal) in presolar SiC from Indarch: single grains and bulk grain-size fractions.

The RIMS data are shown in Fig. 1, along with the TIMS data described above, and in Table 2 along with the NanoSIMS data. Two Si_3N_4 grains were also subjected to RIMS analysis but did not contain measurable Ba. The Ba isotopic compositions of individual SiC grains plot on the same line as the grain-size fractions for each three-isotope system, but show a far larger range of δ-values. Since each grain

Table 1. Ba isotopic compositions of Indarch grain-size fractions measured by TIMS and calculated s-process compositions

Sample	^{130}Ba/^{136}Ba	dev[c]	^{132}Ba/^{136}Ba	dev	^{134}Ba/^{136}Ba	dev	^{135}Ba/^{136}Ba	dev	^{137}Ba/^{136}Ba	dev	^{138}Ba/^{136}Ba	dev
Normal[a]	0.01346		0.01290		0.308		0.839		1.429		9.129	
Observed Mean Compositions												
KJ (bulk)[b]	0.00294	-782	0.00278	-784	0.334	84	0.287	-658	0.844	-409	6.849	-250
0.10-0.15 μm	0.00269	-800	0.00251	-805	0.330	73	0.273	-674	0.838	-413	7.059	-227
0.15-0.20 μm	0.00523	-612	0.00495	-616	0.325	57	0.405	-518	0.970	-321	7.452	-184
0.20-0.25 μm	0.00433	-678	0.00414	-679	0.329	70	0.360	-572	0.923	-354	7.168	-215
0.25-0.30 μm	0.00346	-743	0.00330	-744	0.334	85	0.313	-627	0.870	-391	6.917	-242
> 0.3 μm	0.00664	-506	0.00629	-512	0.323	51	0.489	-417	1.061	-257	7.559	-172
Calculated s-process composition												
KJ (bulk)[b]	0.00005		≡ 0		0.341		0.135		0.684		6.220	
0.10-0.15 μm	-0.00002		≡ 0		0.337		0.134		0.682		6.491	
0.15-0.20 μm	0.00020		≡ 0		0.339		0.127		0.669		6.306	
0.20-0.25 μm	0.00010		≡ 0		0.342		0.128		0.673		6.223	
0.25-0.30 μm	-0.00229		≡ 0		0.352		0.129		0.529		5.526	
> 0.3 μm	-0.00240		≡ 0		0.349		0.145		0.528		5.190	

[a] Normal composition from Lewis et al. 1983; [b] Prombo et al. 1993; [c] Deviations from normal composition in permil

represents an individual star, the range of isotope distributions obtainable by s-process nucleosynthesis in AGB stars is much larger than that indicated by aggregate measurements. Measurements of Ba isotopes in individual SiC grains from the Murchison meteorite are indistinguishable from individual SiC grains from Indarch [11]. It was not possible to extrapolate to a pure s-process component for single grains because the only purely non-s-process isotopes, ^{130}Ba and ^{132}Ba, were too scarce to measure in individual grains.

Of the eight individual grains measured by both RIMS and NanoSIMS, seven were mainstream grains and one (grain #6) was a B grain. The Ba isotope distribution for grain #6 was indistinguishable from solar, as was another B grain culled from the Murchison meteorite [11]. Previous single-grain measurements of B-type SiC grains show near-normal isotope distributions for Mo, and Zr, with the notable exception of a large enhancement in ^{96}Zr [12]. One of the sources of A+B grains has been proposed to be J stars. Thus far, single-grain RIMS measurements exist for seven A+B grains, none of which show s-process signatures. This is consistent with previous results [13] that A+B grains measured so far have little or no s-process contribution. Though ^{96}Zr is primarily an r-process isotope, it can also be made via the s-process if the neutron flux is high enough to overcome the s-process branch at ^{95}Zr, ($t_{1/2}$ = 65d). It is interesting to note that no corresponding enhancement was seen in ^{100}Mo which, like ^{96}Zr, is also primarily r-process, but can be made by the s-process by crossing the branch point at ^{99}Mo, ($t_{1/2}$ = 2.7d). Thus, ^{96}Zr in B-grains is truly anomalous, since neither an r-process nor an s-process scenario can account for all of the isotopic compositions in all of the grains studied to date. Additional single-grain analyses are thus required to determine the stellar sources of B-grains.

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Table 2. Ba isotopic compositions[a] of individual SiC grains from Indarch measured by RIMS, and C and N isotopic compositions on the same grains measured by NanoSims.

Grain	δ^{134}Ba/^{136}Ba	δ^{135}Ba/^{136}Ba	δ^{137}Ba/^{136}Ba	δ^{138}Ba/^{136}Ba	^{12}C/^{13}C	^{14}N/^{15}N
#1	695 ± 251	-666 ± 62	-370 ± 80	-361 ± 60	69 ± 0.8	1161 ± 180
#2	82 ± 136	-792 ± 34	-467 ± 51	-252 ± 51	69 ± 1.1	851 ± 80
#4	94 ± 181	-621 ± 65	-399 ± 74	-264 ± 66	68 ± 2.4	330 ± 21
#6	-5 ± 179	-49 ± 126	-86 ± 108	-7 ± 93	3 ± 0.0	143 ± 6
#7	-54 ± 136	-795 ± 35	-525 ± 51	-469 ± 40	38 ± 0.5	1708 ± 211
#15	-128 ± 702	-424 ± 378	-200 ± 415	489 ± 586	70 ± 3.4	274 ± 24
#17	75 ± 188	-744 ± 52	-531 ± 65	-367 ± 61	73 ± 4.2	273 ± 17

[a]In permil deviations from measured standard; all reported errors are 2 σ