A TRANSMISSION ELECTRON MICROSCOPY STUDY OF ULTRAMICROTOMED SIC-X GRAINS.
K. M. Hynes, T. K. Croat, S. Amari, A. F. Mertz and T.J. Bernatowicz, Laboratory for Space Sciences and Department of Physics, Washington University, St. Louis, MO 63130, USA, khynes@hbar.wustl.edu.

Introduction: A small sub-population of presolar SiC grains from Murchison show isotopic signatures of a supernova origin [1]. These rare grains (named SiC-X) are often identified from their large ^{28}Si excesses, but also have significant carbon anomalies as well as radiogenic ^{26}Mg and ^{44}Ca [2, 3]. Extensive transmission electron microscopy (TEM) investigations have been done of Murchison KJB SiCs, but mostly of mainstream grains and without correlated knowledge of their isotopic compositions [4]. Microstructure and phase information has been obtained from only a few known SiC-X grains [5]. Here we present preliminary results from TEM studies of four SiC-X grains from the Murchison KJG fraction.

Experimental: SiC grains from the Murchison meteorite (KJG fraction, 3 µm observed average size [6]) were analyzed in the NanoSIMS for C and Si. Of those identified as X grains from their Si isotopic anomalies, four (with sizes from 2.5-4 µm) were selected for TEM studies. These grains were removed from the NanoSIMS mount, embedded in resin, and sliced into ≤100 nm sections with a diamond ultramicrotome. The slices were then studied in a JEOL 2000FX analytical TEM equipped with NORAN Energy Dispersive X-ray Spectrometer (EDXS). EDXS quantitative analysis of Mg, Al, and Si was done using a basaltic glass standard (USNM 113498).

Results: Isotopic data from the four selected KJG grains are summarized in Table 1, all with significant ^{28}Si excesses. All show lower δ^{30}Si than δ^{29}Si, falling near the slope=0.67 line defined by most SiC-X grains in a three-isotope plot [2]. The carbon anomalies also lie within the previously-observed range ($10 < {}^{12}C/{}^{13}C < 6800$). Figure 1 shows a typical ultramicrotomed SiC cross-section, of which several were available for each SiC. Although much of the material is preserved, the SiC is fragmented and shattered. Crystal domains (Fig. 2) range in size from ~70-200 nm (geometric mean diameter ~170 nm), similar in size to one of the previously-studied SiC-X grains, but much larger than the other (mean crystal size ~10 nm) [5]. The SiC-X crystal domains are smaller than KJB mainstream SiCs, in which most (66%) of the 320-700 nm grains consisted of a single, untwinned domain. Since these SiC-X domains are smaller than the fragments, their size can be observed independently of any fragmentation. Further, adjacent crystal domains are often found with well-defined orientation relationships with one another, showing that twinning (and not ultramicrotome damage) is responsible for the apparent small domain size.

Table 1. Isotopic results from KJG SiC-X grains for Si (in permil) and C (both with 1 σ errors).

Grain	δ^{29}Si	δ^{30}Si	^{12}C/^{13}C
KJG 31-1	-309 +/- 4	-436 +/- 5	250 +/- 4
KJG 129-1	-187 +/- 5	-329 +/- 6	111 +/- 1
KJG 253-2	-231 +/- 5	-396 +/- 6	162 +/- 2
KJG 411-2	-298 +/- 5	-422 +/- 6	216 +/- 3

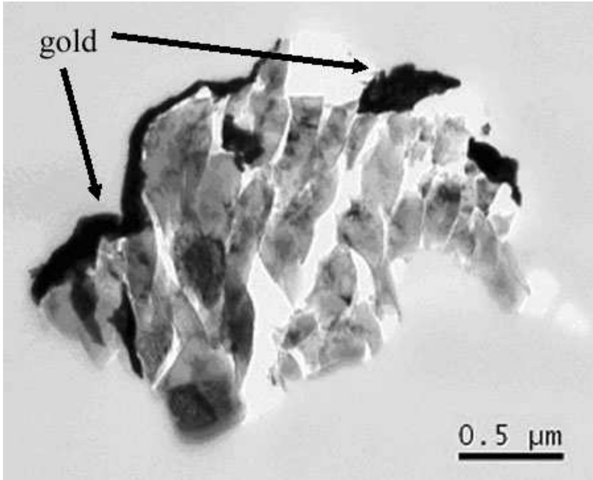

Figure 1. Bright-field (BF) image of an ultramicrotome slice of KJG SiC-X grain (31-1) showing its generally shattered condition after ultramicrotomy. The dark areas at top are gold that was sputter deposited during SIMS.

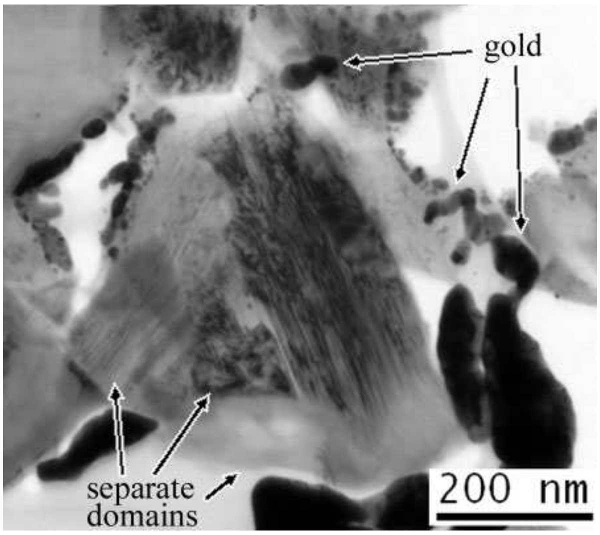

Figure 2. BF image of fragment from SiC-X 129-1 showing multiple crystalline domains. Intensity variation among domains is due to orientation-dependent diffraction contrast.

Twinning across close-packed planes at certain angles creates a low-energy boundary in FCC materials, and

thus is common in SiCs. The preponderance of twinning makes determination of the polytype challenging. Because of the complex multi-domained structure, it is also time-consuming to determine whether a SiC-X grain is a single multiply-twinned grain or is an aggregate of separate grains. Thus far, using convergent beam diffraction (CBED) patterns, we have determined the polytype from seven crystal domains. Six of these patterns (from SiC-X 129-1 and 253-2) were found consistent with the 3C-SiC polytype (cubic; a=4.35Å), mostly using <110> zones. This polytype was also commonly seen in mainstream SiCs (in ~80% of KJB grains [4]). Figure 3 shows a pair of twinned 3C-SiC domains from SiC-X 253-2 along with CBED patterns at <110> orientations. The two patterns are rotated 70.3° with respect to one another, indicating a common Σ=3 twin. <110> zone patterns from each domain were also found 60° away in tilt, giving conclusive evidence of a twinned 3C-SiC cubic structure. The only CBED pattern inconsistent with 3C-SiC was from SiC-X 31-1, appearing instead to be an intergrowth of 3C-SiC and another polytype. Further crystallographic analysis is required to resolve this question. Polytype determinations from SiC-X 411-2 have not been done.
Searches for internal subgrains of other phases have been done, using both EDXS and also observation at high magnification while the sample was systematically tilted. None have yet been found. However, the existence of radiogenic ^{44}Ca in ~20% of SiC-X grains [3] leads us to expect that internal TiCs will be present in some grains.

EDXS measurements of two of the SiC-X grains (411-2 and 129-1) showed significant Mg and Al

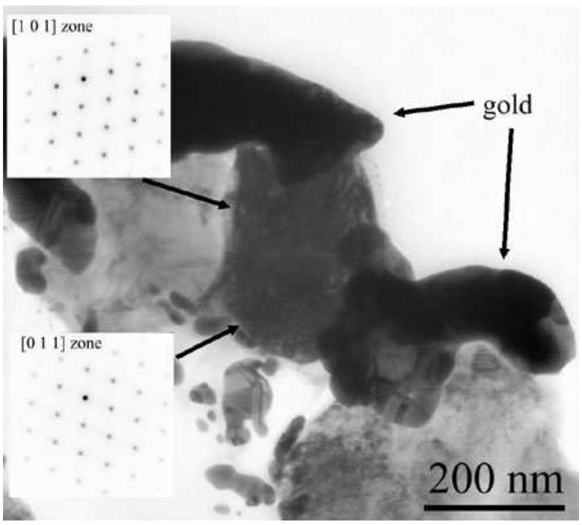

Figure 3. BF image of twinned (Σ=3) 3C-SiC crystals along with separate CBED [011] patterns from each twin domain. The (111) interface plane is approximately horizontal in the image.

content, both having approximate concentrations of $Si_{95}Al_3Mg_2$ (excluding carbon). SiC-X 253-2 showed lower, but still measurable, concentrations, with $Si_{98}Al_{1.4}Mg_{0.6}$. The fourth grain (31-1) did not show significant Mg or Al content, with upper limits near 0.2 and 0.7 at. %, respectively. Since an insignificant amount of Mg typically condenses within SiCs during their formation (Mg/Al<0.05 in mainstream SiCs [7]), the Mg is likely to be of radiogenic origin (from ^{26}Al decay). Mg/Al ratios of 0.60+/-0.04 (8%), 0.67+/-0.06 (9%), and 0.44+/-.04 (10%) were derived from 411-2, 129-1, and 253-2, respectively (reported with combined errors from 2σ counting statistics and k-factor determination). If the measured Mg is radiogenic, these ratios are analogous to inferred ^{26}Al/^{27}Al ratios, and are found to be consistent with the highest ^{26}Al/^{27}Al ratios inferred from SiC-X grains [2]. We do not yet know the spatial distribution of Mg and Al within the SiC-X grains.

Discussion: Preliminary determinations show that 3C-SiC is a common polytype in SiC-X grains, similar to results for mainstream SiCs. This result is interesting, since the higher gas pressures in SN ejecta could conceivably lead to formation of higher-order hexagonal or rhombohedral polytypes, which tend to condense at temperatures greater than those inferred for the formation of mainstream SiC (1470 K – 1720 K) [4]. We consider the question of whether the 3C polytype dominates the population of SiC-X (as is true for mainstream grains) to be an undecided issue in view of our limited data, and one to pursue in future studies. The finer domain sizes in SiC-X grains could be the result of more rapid formation. There is evidence (namely incorporation of considerable live ^{49}V [8]) that SiC-X grains condense more quickly, over a timescale of several months, as opposed to the several years available for grain formation in AGB outflows [9]. However, microstructural investigations of mainstream SiCs from the same size fraction (KJG) are needed to investigate possible effects of faster formation on crystal size.

Acknowledgements: We thank Roy Lewis (Enrico Fermi Inst./ Univ. of Chicago) for providing the Murchison KJG samples.

References: [1] Amari, S. et al. (1992) *ApJ. 394*, L43. [2] Nittler, L.R. et al. (1995) *ApJ 453*, L25. [3] Hoppe, P. et al. (2000) *Met. Planet. Sci. 35*, 1157. [4] Daulton, T. L. et al. (2003) *GCA 67*, 4743. [5] Stroud, R. M. et al (2004) *Met. & Planet. Sci. 39*, 5039. [6] Amari, S. et al. (1994) *GCA 58*, 459. [7] Amari, S. et al. (1995) *Meteoritics 30*, 679. [8] Hoppe, P. & Besmehn, (2002) *ApJ, 576,* L69. [9] Bernatowicz, T.J. et al (2005) *ApJ, 631*, 988.