EXTREME Si-29 AND Si-30 ENRICHMENTS FOUND IN RARE MURCHISON SiC-CONTAINING GRAPHITES. T. K. Croat and F. J. Stadermann, Laboratory for Space Sciences and Department of Physics, Washington University, St. Louis, MO 63130, USA, tkc@wustl.edu.

Introduction: Although rare (in <1% of graphites), silicon carbides are found as internal grains within Murchison KFC1 onion graphites, with four instances reported thus far [1-3]. These composite grains (e.g. SiCs found within graphite) are of particular interest, as they are a clear case in which both presolar phases form from the same star. Here we report on the microstructure as well as the chemical and isotopic compositions of two newly-discovered graphites containing relatively large internal SiCs (100-225 nm in size). NanoSIMS isotopic analyses of both SiC-containing graphites reveal large ^{29}Si and ^{30}Si enrichments indicative of a massive star origin, with one nearly as extreme as the unusual SiC from [4]. Until now, an anomaly of this type had been measured only once among tens of thousands of presolar SiCs.

Experimental: Graphites were obtained from the KFC1 density and size separate (2.15-2.20 g cm^{-3}, >1 μm) of the Murchison meteorite [5]. These graphites were deposited from suspension onto a glass slide, embedded in resin, and then sliced into ≤ 100 nm sections with an ultramicrotome. The slices were examined in a JEOL 2000FX analytical TEM equipped with a NORAN Energy Dispersive X-ray Spectrometer (EDXS), and EDXS quantitative analyses were done based on basaltic glass standards. The TEM grid was then mounted into a clamping holder and imaged in the NanoSIMS for $^{12}C^-$, $^{13}C^-$, $^{28}Si^-$, $^{29}Si^-$ and $^{30}Si^-$ and secondary electrons.

Results: The properties of previously discovered SiC-containing graphites and #5 and #6 from this work are summarized in Table 1. Graphite #5 is a 1.3 μm onion with a large SiC (~227 nm) in two adjacent chunks at its periphery (figs 1a and 1b). The SiC appeared to be embedded within the graphite at all stage tilt angles (rather than being a stuck-on foreign grain). Tilting studies also revealed more than a dozen separate crystalline domains with an average domain size of ~80 nm. No obvious preferred orientation relationships were seen between adjacent domains. Selected area diffraction (SAD) and/or microdiffraction patterns were indexed from 9 different domains, with 8 indexing to 3C-SiC (4.4 +/- 0.1Å, FCC). One domain indexed to 3C-SiC [011] at one orientation but also showed a [01 -1 1] hexagonal zone axis pattern (ZAP) at another orientation 34° away, indicating a 2H/3C intergrowth. At their [011] orientations some of the domains showed stacking faults, evident as streaking in diffraction patterns and alternating bright and dark domains in DF images (Fig 1b) whereas others did not. SiC-containing graphite #6 is a 1.0μm onion with a central SiC that is morphologically very different from #5, consisting of a single weakly-diffracting domain, presumably due to disorder. The diffracted intensity was too weak for SAD (normally feasible for grains of this size) so microdiffraction is shown (Fig 1c inset). ZAPs were indexed from the [011] and [111] FCC zones (found with expected intrazonal angle), yielding a lattice parameter of 4.27 +/- 0.05. The SiC's central position in the onion suggests it acted as a heterogeneous nucleation site for the graphite, but the disorder evident in the grain suggests it may have no longer been a stable phase in the graphite-forming environment.

Table 1. Properties of SiC-containing graphites

Graph-ite #	# SiCs	SiC size (nm)	Polytype	Ref
1	23	35 (13-83)[a]	3C & 1D disordered	2
2	5	39 (15-61)[a]	3C & 3C/2H intergrowth	1, 3
3	1	26	3C	3
4	5	53 (30-75)[a]	3C	3
5	1	227	3C & 3C/2H intergrowth	this work
6	1	104	3C	this work

a. indicates SiC size range

Both SiCs have relatively high Si/C count ratios and undetectable levels of Ti (<0.15 at. %), Zr (<0.2 at. %) and Mo (<0.3 at. %), and thus are clearly SiC and not other isostructural carbides. SiC #5 had a composition of $(Si_{99.3}Al_{0.7})C$ with <0.1 at. % Mg, from which we can place an upper limit on Mg/Al atomic ratio of ~0.15. This value is significantly lower than the measured Mg/Al ratios from SiC-X grains, among which 3 of 4 showed appreciable Mg presumably from ^{26}Al decay (with Mg/Al at. ratio ~0.6)[3]. The spectra from SiC #6 showed no detectable Al or Mg.

The SiC in graphite #5 has extreme $\delta^{29}Si/^{28}Si$ and $\delta^{30}Si/^{28}Si$ values (see Table 2) that are exceeded only by the unusual SiC from [4]. The $^{12}C/^{13}C$ ratios in #5 are 240 +/- 9 in the SiC and 253 +/-3 in the graphite (identical within 1σ errors), and less extreme than the unusual SiC from [4] (with $^{12}C/^{13}C$ = 844 +/- 34). The SiC in graphite #6 falls near the SiC-Z grains (see Fig. 2). The $^{12}C/^{13}C$ ratios in #6 are 113 +/- 1 in the graphite and 123 +/- 4 in the SiC (identical within 2σ).

Discussion: The large ^{29}Si and ^{30}Si enrichments in #5 necessitate a massive star origin, either a Type II supernova (SN) or Wolf-Rayet star [4]. Such Si enrichments are predicted to occur in the O/C and O/Ne SN zones. Both SN zones also show strong ^{12}C enrichments, and all SiC-containing graphites thus far (and the unusual SiC [4]) also show $^{12}C/^{13}C$ greater than solar. However, the SiC-graphites lack the ^{28}Si

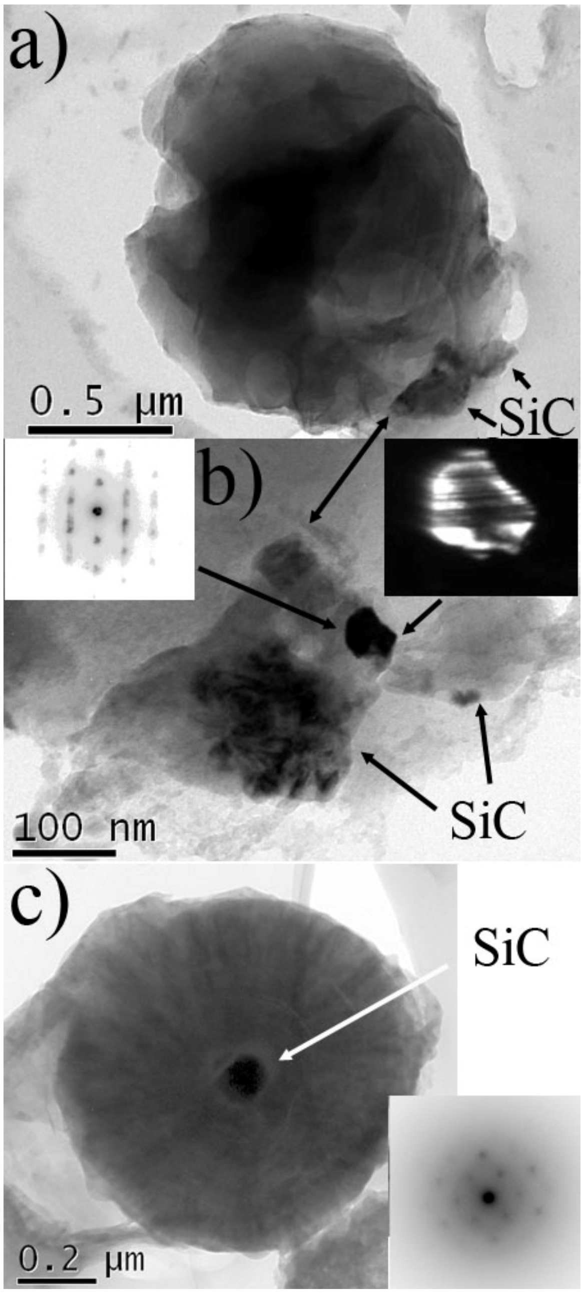

Fig. 1 a) TEM bright-field (BF) image of graphite #5 with internal SiC at periphery. b) close-up on SiC grain from graphite #5 showing multiple dark crystalline domains; inset microdiffraction pattern and dark-field (DF) image show stacking faults from the darkest SiC domain. c) BF image of graphite #6 with central disordered SiC and inset [011] FCC diffraction pattern.

Table 2. Isotopic results from SiCs within graphites for C and Si (in permil), both with 1 σ errors.

Graphite #	$^{12}C/^{13}C$	$\delta^{29}Si$	$\delta^{30}Si$
1	110+/-2	n/a	n/a
3	782+/-18	n/a	n/a
5	240+/- 9	1290+/-50	1040+/-50
6	123 +/- 4	120 +/- 30	380 +/- 45

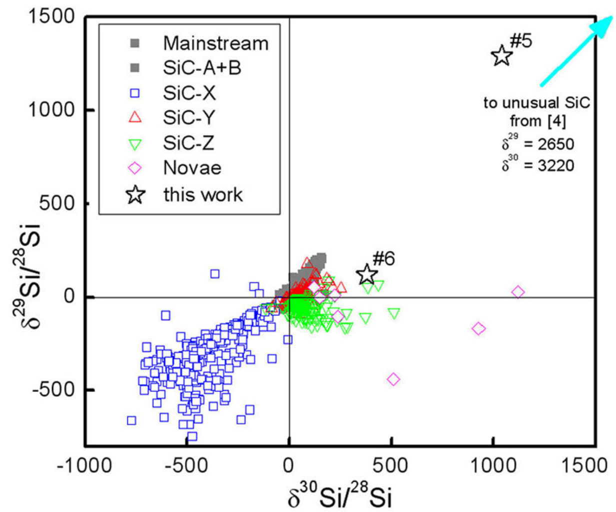

Fig. 2. Si isotopic plot of SiC-containing graphites #5 and #6 along with other presolar SiC types ([6] and references therein). Star symbols are roughly the same size as 1σ error bars.

enrichments and the evidence for ^{26}Al seen in SiC-X SN grains (which are dominated by Si from Si/S SN zone). The phase condensation sequence inferred from the SiC-graphites differs. Four have peripheral SiC formation (graphite → SiC), whereas two others contain central or interior SiCs (SiC → graphite). Despite its significant Si anomalies, SiC-graphite #6 could possibly originate in a low metallicity AGB star. However, it has higher $^{12}C/^{13}C$ and $\delta^{29}Si/^{28}Si$ values than all reported SiC-Z grains and is exceeded in $\delta^{30}Si/^{28}Si$ by only 3 of 127 Z grains. Overall, it seems most likely that the 2 grains here and that from [4] define their own distinct SiC group, until now mostly hidden within graphites, whose exact stellar origin requires further studies.

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