EXTREME Si-29 AND Si-30 ENRICHMENTS FOUND IN RARE MURCHISON SiC-CONTAINING
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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 29Si and 30Si
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
\(\mu\)m) of the Murchison meteorite [5]. These graphites were deposited from suspension onto a glass slide,
embedded in resin, and then sliced into \(\leq 100\) nm sections with an ultramicrotome. The slices were
examined in a JEOL 2000FX analytical TEM equipped with a NORAN Energy Dispersive X-ray Spectrometer
(LEDXS), 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 \(\mu\)m onion with a large SiC (∼227 nm) in two adjacent
chunks at its periphery (figs 1a and 1b). The SiC appeared to be embedded in 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 -11] 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 1.0\(\mu\)m 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

<table>
<thead>
<tr>
<th>Graphite #</th>
<th># SiCs</th>
<th>SiC size (nm)</th>
<th>Polytype</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>23</td>
<td>35 (13-83)</td>
<td>3C &amp; 1D disordered</td>
<td>2</td>
</tr>
<tr>
<td>2</td>
<td>5</td>
<td>39 (15-61)</td>
<td>3C &amp; 3C/2H intergrowth</td>
<td>1, 3</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>26</td>
<td>3C</td>
<td>3</td>
</tr>
<tr>
<td>4</td>
<td>5</td>
<td>53 (30-75)</td>
<td>3C</td>
<td>3</td>
</tr>
<tr>
<td>5</td>
<td>1</td>
<td>227</td>
<td>3C &amp; 3C/2H intergrowth</td>
<td>this work</td>
</tr>
<tr>
<td>6</td>
<td>1</td>
<td>104</td>
<td>3C</td>
<td>this work</td>
</tr>
</tbody>
</table>

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 26Al 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 \(\delta^{28}\)Si/\(^{29}\)Si and \(\delta^{28}\)Si/\(^{30}\)Si ratios (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 \(^{28}\)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 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 $\rightarrow$ SiC), whereas two others contain central or interior SiCs (SiC $\rightarrow$ 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 $\delta^{30}$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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