LOW-DENSITY PRESOLAR GRAPHITE SPHERULES FROM THE ORGUEIL METEORITE.

T. K. Croat, M. Jadhav, E. Lebsack and T. J. Bernatowicz, Laboratory for Space Sciences and Department of Physics, Washington University, St. Louis, MO 63130, USA, tkc@wustl.edu.

Introduction: The vast majority of isotopic and TEM studies of presolar graphite spherules have been from the Murchison (CM2) meteorite, although more recently graphite has become available from the Orgueil (CI1) meteorite [1]. Although the graphites from both meteorites show similar types of isotopic anomalies, SEM studies have discovered curious differences, namely larger grain size in the Orgueil high-density fraction and different surface morphology in low-density fraction [1]. Here we present results from TEM studies of 3 Orgueil low-density graphites, 2 of which are supernova (SN) candidates and 1 from the little-studied ^{13}C-rich group. All of these graphites have subgrains with mineral compositions never before seen among 13 low-density Murchison graphites [2].

Experimental: Graphites from the ORG1d density and size separate (1.75-1.92 g cm^{-3}, >1 μm) of the Orgueil meteorite [1, 3] were selected based on NanoSIMS C, O, N and Si isotopic measurements. These graphites were picked from the mounts, embedded in resin, and then sliced into ~100 nm ultramicrotome sections. The slices were retrieved on holey carbon-coated copper TEM grids and examined in a JEOL 2000FX analytical TEM equipped with a NORAN Energy Dispersive X-ray Spectrometer (EDXS).

Results: Table 1 summarizes the isotopic ratios from these Orgueil graphites measured with NanoSIMS prior to ultramicrotomy [3]. Although they lack Si anomalies, the ^{18}O-rich compositions of d2 and d6 suggest a SN origin (they also show ^{15}N and slight ^{12}C enrichments). Further, contamination on this mount probably diluted any O and Si isotopic anomalies [3]. D7 shows a highly ^{13}C-rich composition and a ^{15}N enrichment (with approximately solar O and Si ratios). Carbon isotopic distributions from presolar graphite always show a pileup of grains at the ^{13}C-rich end [4], suggesting that these constitute a distinct isotopic group, and d7 is effectively the first of these studied in TEM.

SN graphites d2 and d6: Three internal SiC grains (20-50 nm) were found within the d2 graphite (Figs. 1a and b), a phase not found previously within low-density SN graphites [2]. All were indexed via electron diffraction patterns to the 3C-SiC polytype (FCC, 4.4Å), and 2 of 3 showed twinning on {111} planes. EDXS showed little Al (1-3 at. %) and no detectable Mg (<1 at. %), concentrations too low to determine whether an initial ^{26}Al excess, such as those seen in SiC-X grains [5], might be present. Graphite

Table 1. Orgueil graphite isotopic ratios

#	^{12}C/^{13}C	^{14}N/^{15}N	^{16}O/^{18}O	δ^{29}Si	δ^{30}Si
m3-d2	113 +/-1	182 +/-2	435 +/-8	4 +/-10	-33 +/-12
m3-d6	124 +/-1	218 +/- 2	437 +/- 8	-9 +/- 3	-6 +/-3
m3-d7	11.1 +/-0.1	236 +/- 2	537 +/- 10	-33 +/- 12	-6 +/-14

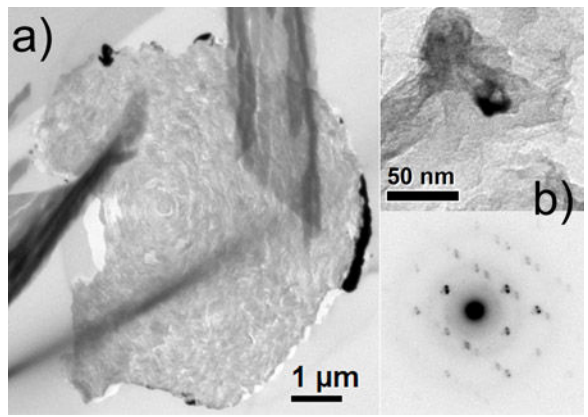

Fig.1. a) d2 graphite and b) internal SiC with associated [011] diffraction pattern showing twinning.

d2 also contains many Ti-rich grains (~200 ppm abundance; 20-35nm), which were shown in all cases to be TiC (patterns from 6 grains indexed to ~4.4Å FCC phase). The TiCs lacked s-process element enrichment (e.g. Zr, Mo) with upper limits of 3-10x solar in the Mo/Ti ratio, and thus are similar to the SN graphites but markedly different from many high-density KFC1 graphites of AGB origin [6], which typically show Mo enrichments of several hundred times the solar ratios.

The d6 graphite (Fig. 2a) shows a high number density of Ti-rich internal grains (~1200 ppm), comparable to that found in low-density Murchison supernova (SN) graphites [2]. The Al content of the grains is high and quite variable (0 < Al/Ti < 1.15; Mg not detected), often above the solubility limit for Al in TiC. Again, no s-process element enrichments were seen (upper limit of <10x solar in Mo/Ti). Of 11 Ti-rich grains from d6 investigated with electron diffraction, 5 were conclusively indexed to the TiC phase (FCC, ~4.4Å) whereas for the others the patterns are inconsistent with TiC. This is in contrast to Murchison SN graphites, where hundreds of TiCs are found unaccompanied by other Ti phases. The other Ti-rich grains are ~3x smaller than carbides, more Al-rich, and some have high Si content (up to ~30 at. %). Of the remaining grains, several index to a larger cubic phase with an 8.8-9.9Å lattice parameter, possibly a different carbide phase (TiC_2; 8.6Å). Several other grains are hexagonal, possibly the α-Ti metallic phase. However, identification of the non-TiC phases is tentative. Two grains appear to be intergrowths of the TiC phase and one of the TiAl-rich phases (Fig. 2b). However, since the relative orientations of the two intergrown phases are not yet known, it is possible that they are merely adjacent and not grown together.

^{13}C-rich graphite d7: D7 (Fig. 3a) differs greatly from the SN graphites in the types of internal grains found. From its internal grain chemical compositions, we infer a very s-process enriched stellar source. This enrichment is most evident in a ~20 nm RuOs-rich grain (Fig. 3b) of composition $Ru_{34}Os_{24}Mo_{23}Fe_{17}Ni_2$. Four major zones (including the [0001] and [-1 2 1 1]) of the RuOs-rich grain were indexed to a close-packed hexagonal (a=2.8, c=4.4) solid solution of metallic Ru and metallic Os. Two internal SiCs (e.g. Fig. 3b) were also found (~30nm and ~70 nm). Both index to the 3C-SiC polytype (FCC; a~4.5Å), and show extensive twinning on {111} planes. Both Os-rich grains and SiCs are very rare among Murchison graphites (2 Os-rich and 6 SiCs found in ~1500 high density graphite sections and none among 13 low-density graphites). In d7, 2 low-nickel (Ni < 6 at. %) kamacite grains (BCC; a~3.0Å) and several chromites were also found, although the chromites found at the graphite's periphery may just be attached grains from the meteorite matrix rather than true internal grains. No TiCs were found, although much of d7 has not yet been examined.

Discussion: Despite its absence in the Murchison low-density SN graphites, SiCs of the type found in Orgueil d2 have been found in rare instances within high-density Murchison graphites. Two of the SiC-containing high-density graphites showed very unusual Si anomalies in the internal SiCs, with extreme ^{29}Si and ^{30}Si compositions indicative of a massive star origin [7] but at the opposite extreme from the SiC-X SN grains (with large ^{28}Si enrichments). The Si anomalies seen in the internal SiCs were not measurable elsewhere in the host graphites, possibly due to low Si abundance and/or dilution. This has consequences for d2, in that the lack of a measured Si anomaly from the graphite does not necessarily constrain the SiC's isotopic composition, especially given the suspected contamination of this mount [3]. The Al-richness of internal grains within d6 makes it unlike any graphite yet studied, possibly representing contributions from compositionally different SN zones. Further, that both Orgueil SN graphites show different properties than the Murchison ones indicates that these overall grain populations may be different despite showing similar ranges of isotopic anomalies. The s-process enrichment in the d7 ^{13}C-rich grain agrees with previous data from high-density Murchison graphites. When s-process enriched phases, such as metallic RuFe, were found within high-density graphites, they predominately occurred in this ^{13}C-rich isotopic sub-group rather than in the more common ^{12}C-rich graphites. The large s-process enrichments in the internal grains indicate that CH stars or Sakurai's object are plausible stellar source types for the ^{13}C-rich graphites, as was the case for s-process enriched SiC A+B grains [8].

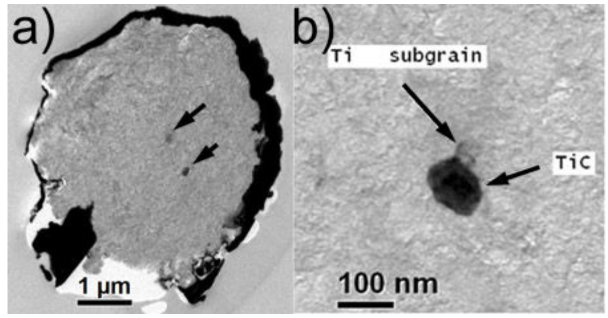

Fig.2. a) d6 graphite with internal Ti-rich subgrains (arrows) and b) d6 TiC with an attached TiAl-rich subgrain

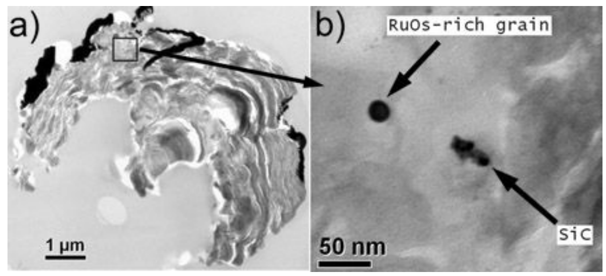

Fig.3. a) d7 graphite section and b) inset region from d7 showing a RuOs-rich grain and a twinned SiC.

Acknowledgments: We thank S. Amari and T. Maruoka for preparation of the Orgueil samples.

References: [1] Jadhav M. et al. (2005) *LPS* XXXVI*, Abstract # 1976*. [2] Croat T.K. et al. (2003) *GCA*, *67*, 4705. [3] Jadhav M. et al. (2008) *LPS* XXXIX*, Abstract # 1047*. [4] Hoppe P. et al. (1995) *GCA*, *59*, 4029. [5] Hynes et al. (2008) *LPS* XXXIX*, Abstract # 2076*. [6] Croat T.K. et al. (2005) *ApJ*, *631*, 976. [7] Croat T.K. et al. (2008) *LPS* XXXIX*, Abstract # 1739*. [8] Amari S. et al. (2001) *ApJ*, *559*, 463.