MICROSTRUCTURAL DIFFERENCES AMONG THE ISOTOPIC GROUPS OF LOW-DENSITY ORGUEIL GRAPHITES. 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: Although the overall isotopic distributions of presolar graphites from the Orgueil (ORG) and Murchison (MUR) meteorites are similar [1], grain sizes are larger in the high-density ORG fraction and morphological differences are seen between the ORG and MUR low-density (LD) fractions. Furthermore, the first three ORG graphites studied with TEM and NanoSIMS showed types and combinations of internal grains not found previously among 13 MUR graphites [2, 3], and thus it is possible that there is a fundamental difference between the ORG and MUR populations. Here, we present TEM results from four more ORG LD graphites, one from the ^{13}C-rich group, one with a nearly solar isotopic composition, and two that show isotopic anomalies consistent with a supernova (SN) origin.

Experimental: Graphites from the ORG1d density and size separate (1.75-1.92 g cm^{-3}, >1 μm) of the ORG meteorite [1] 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 ~70 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). Graphites from this mount (m3) showed indications of contamination that likely dilute any minor element isotopic anomalies (e.g. in N, O, Si).

Results: Table 1 shows isotopic ratios from ORG graphites measured with NanoSIMS prior to ultramicrotomy. Graphite d9 is typical of the ~20% of LD graphites that are ^{13}C-rich, with a large ^{13}C-enrichment but little or no anomalies in N, O or Si. Graphite d11 is similar in size and morphology to other presolar LD graphites, but shows only relatively small C anomalies (~33% of LD graphites are in this nearly solar isotopic group). Graphites d20 and d25 are typical supernova (SN) graphites (~40% of LD graphites in this group), with large ^{18}O and slight ^{28}Si enrichments. Their slight ^{28}Si enrichments in Table 1 are likely diluted somewhat because of mount contamination. D20 also shows a slight ^{15}N enrichment. Both SN graphites are slightly larger than typical MUR LD graphites (at 10 and 18 microns, respectively).

^{13}C-rich graphite d9. Along with their C isotopic composition, ^{13}C-rich graphites are distinct from SN graphites in that they lack refractory carbides. As was

Table 1. Orgueil graphite isotopic ratios

#	$^{12}C/^{13}C$	$^{14}N/^{15}N$ [1]	$^{16}O/^{18}O$	$\delta^{29}Si$	$\delta^{30}Si$
m3-d9	13.7 +/-0.1	253 +/-15	524 +/-9	15 +/-7	11 +/-8
m3-d11	92.4 +/-0.5	276 +/- 17	527 +/- 9	-10 +/- 5	-8 +/-6
m3-d20	72.9 +/-0.4	241 +/- 15	299 +/- 5	-40 +/- 10	-55 +/-5
m3-d25	99.8 +/-0.6	255 +/- 15	461 +/- 8	-30 +/- 6	-28 +/- 7

1. N ratios corrected for contamination problem.

the case for two previous ^{13}C-rich graphites studied in TEM [2, 3], d9 (Fig. 1a.) contained no internal TiCs. D9 has an upper limit of 50 ppm on carbide abundance, in contrast to SN graphites which have ubiquitous TiCs with 25-2400 ppm abundances [3]. A kamacite grain ($Fe_{89}Ni_{11}$; 2.9+/-0.1Å FCC) and numerous chromites were found along the periphery of graphite d9. However, previous NanoSIMS isotopic measurements of chromites showing normal O ratios suggest that chromites are either adjacent material from the meteorite matrix or perhaps form via alteration of pre-existing iron grains during laboratory processing (e.g. oxidation by dichromate solution during extraction from meteorite). The peripheral kamacite also might not be a primary presolar condensate. However, such iron grains and refractory metal nuggets occur with a disproportionately high frequency among ^{13}C-rich graphites. Graphite d9 also contained a 125 nm diameter Si-rich inclusion (Fig. 1b). EDXS of this grain showed a 3x higher O than is seen from internal SiCs, indicating a possible silicate. Although some facets do appear, no evidence of crystallinity was observed while tilting the grain.

Nearly solar graphite d11. As with the ^{13}C-rich group above, d11 did not contain TiCs (upper limit on TiC abundance of ~30ppm). Only peripheral oxides and chromites were seen. Furthermore, although d11 selected area diffraction patterns did show {002}, {100} and {110} graphitic rings, its microtexture is quite different and lacks the turbostratic layering that is evident in other ORG and MUR LD graphites (Fig. 2).

SN graphites d25 and d20: Graphite d25 shows the advantage of serial ultramicrotomy in that 55 slices were visible on the TEM grids (only a small fraction of which have been searched) and further material is

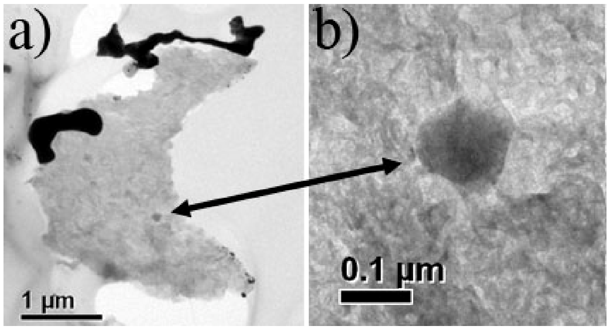

Fig.1. Bright-field (BF) TEM images of a) d9 graphite (showing deposited gold at edge and sputtering damage from SIMS) and b) internal SiO-rich grain.

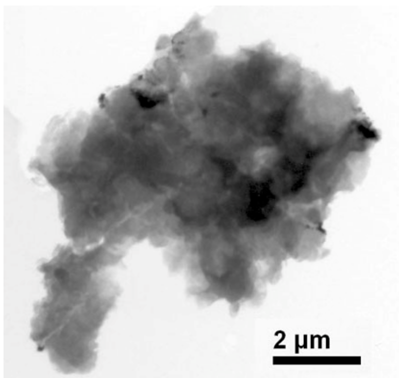

Fig.2. BF image of d11 nearly solar composition graphite.

available on grid bars for NanoSIMS analysis. Copious TiCs are found, with a TiC abundance of ~300 ppm. Figure 3a shows a typical graphite section with the position of internal grains (mostly TiCs) indicated around its perimeter. The carbides were mostly euhedral with 75 nm average diameter, and with little evidence of the partially amorphous rims present on many MUR SN carbides (e.g. Fig. 3c). In terms of their chemical composition, the carbides were similar to those from MUR SN graphites, with an avg V/Ti = 0.073 (60% of solar ratio) and lacking enrichment in Mo and other s-process elements (typical upper limit on Mo/Ti < 10x solar). Graphite d25 also contains a 40 nm troilite grain (FeS; hexagonal, a = 3.45, c = 5.75), which unlike most oxides and sulfides was found towards the center of the graphite (Fig. 3b). Graphite d20 has a far lower TiC abundance than d25 (only 2 small TiCs found; ~5 ppm abundance). Further, it has abnormally high Si content, with Si/C count ratios of ~0.2 (10x higher than in typical graphites). No Si-rich inclusions were found, and thus the Si appears to be trapped within the turbostratic graphite itself.

Discussion: With the absence of TiC and presence of iron grains, graphite d9 is similar to other ^{13}C-rich graphites. Although internal SiCs have been found in this isotopic group [2], the apparently non-crystalline Si-rich (and O-rich) inclusion in d9 is unique. Despite

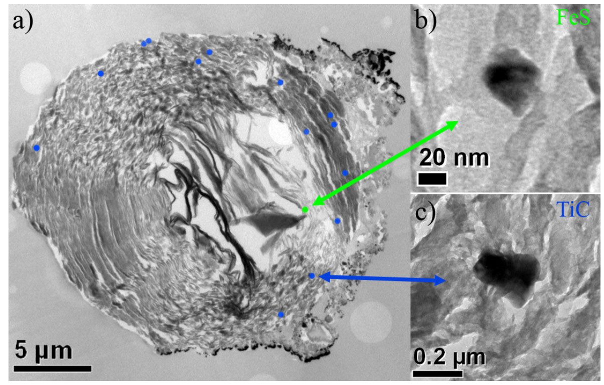

Fig.3. a) SN graphite d25 with positions of internal grains (mostly carbides) indicated, along with b) internal troilite grain and c) ~225 nm TiC grain.

this finding, the properties of several ^{13}C-rich graphites observed in TEM are consistent with a born-again AGB origin [2, 4].

Graphite d11 (showing only a minor ^{12}C enrichment) has an atypical morphology and lacks refractory carbides and other common internal grains. Although some near-solar graphites probably will show the typical indications of presolar graphite, these results suggest a different origin for many of the 1/3 of graphites with nearly solar isotopic composition.

Out of 5 ORG SN graphites studied to date [2, this work], d25 is most similar to the MUR SN graphites. This similarity shows that the fundamental properties of SN graphites are not greatly altered by the different processing they experienced on the ORG and MUR parent bodies. With the material available from this large graphite, chemical and isotopic trends in the internal grains as a function of distance from the graphite center can be studied, and this will reveal the detailed evolution of conditions in the gas as this graphite condensed. That unique LD graphites, such as d20 with its unusually high Si content, continue to be found again demonstrates the diverse behavior of SN graphites, and each such graphite provides a particular insight into grain condensation around SNe. NanoSIMS measurements of TEM cross-sections are planned, which will allow accurate determinations of Si, Ti and other isotopic ratios, while avoiding the surface contamination problems encountered with this particular mount. We will also measure O isotopes from peripheral chromites and oxides to determine if any are intrinsic to the graphites themselves.

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References: [1] Jadhav M. et al. (2006) *New Astron. Rev. 50,* 591. [2] Croat T.K. et al. (2009) *LPS XL*, Abstract # 2175. [3] Croat T.K. et al. (2003) *GCA*, *67*, 4705. [4] Jadhav M. et al. (2008) *ApJ, 682,* 1479.