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Micron-sized presolar carbon-spheres condensed before the origin of our solar system, in the atmosphere of asymptotic-giant-branch stars from carbon atoms dredged up following nucleosynthesis in the stellar interior, have been extracted by dissolution from primitive carbonaceous chondrite meteorites, identified by noble-gas and major-element isotopic analysis, and available to electron microscopists on earth for over two decades [1-4]. Some of these spheres appear to consist of graphite condensed on carbide grains, but others instead show “graphite-onion” rims that surround a spherical-core of unlayered-graphene i.e. carbon that in diffraction shows graphite (hk0) spacings only. The high spatial-frequency tail of these graphene lines [5] suggests a graphene-sheet coherence-width in the 4-nm size range [1], consistent with electron-phase-contrast images of single (sometimes segmented) edge-on sheets [2,4].

Three key questions are: What process in a low-pressure atmosphere can form condensed spheres of graphene with densities approaching that of graphite, but with graphitic-layering absent? How might we synthesize such material in the laboratory? What interesting properties might it have as a result?

Our group has been focusing on three aspects of this problem: (i) Measurements of core-material density (relative to the graphite rim) in ultra-microtomed spheres using zero-loss/deflection analysis [6] via energy-filtered transmission electron microscopy or TEM, (ii) modeling of graphene-sheet nucleation and growth from a carbon melt to get atom-positions for TEM image simulation and estimate the likelihood of pentagonal-loop nucleation as explanation of apparent edge-on facets [4] using the Tersoff potential [7], and (iii) the laboratory synthesis of unlayered-graphene via slow-cooling of carbon vapor.

Carbon plasmon-peak energies [1] suggest an unlayered graphene-density near the 2.3[g/cc] of graphitic carbon. Zero-loss/deflection analysis [6] shows promise for inelastic mean-free-path mapping in a wide range of disordered materials, but specular reflection by diffraction can complicate life (e.g. via Pendullosung or thickness-fringe contrast) by putting electrons into “Bragg storage” if the crystals are thick enough for multiple scattering within the same crystal. Although our first analyses suggest a density closer to 1.8[g/cc] for presolar unlayered-graphene, this may not be confirmed without further calibration studies on the presolar specimens themselves.

Condensation of particles in a low-pressure stellar-atmosphere will require local density increases [1] e.g. associated with jets of material around sunspots, and might take place one C atom at a time, by clustering of polycyclic aromatic hydrocarbons, or by condensation of droplets from a carbon vapor. The latter has the advantage that it will give us the spherical core-shape and density. Although thermodynamics predicts that below pressures of 10.8[MPa], carbon on warming sublimes to a vapor at around 3915K, containerless metals on cooling generally supercool in the liquid state until as much as 30% below the liquid condensation temperature [8], depending on the amount of rearrangement needed between liquid and solid state configurations.
A liquid condensation temperature for carbon around 4600K, in this context, along with the major rearrangement between 12-neighbor coordination in an fcc liquid and the graphene-sheet structure, thus suggest that supercooling to temperatures as low as 3220K (2950C) might be expected in the absence of condensation seeds (like the carbide grains seen at the center of other particles). This synthesis path might also explain the possible TEM evidence [2,4] that the graphene sheets are flat, except where they are seen edge-on to connect at sharp bends whose geometry is suggestive of nucleation on pentagonal (rather than hexagonal) carbon-loops. So far, we are assembling graphene-sheet and graphene-pentacone structures for TEM image simulation (Fig. 1), but only beginning to look at simulated annealing models of pentagonal versus hexagonal loop nucleation.

Finally, slow-cooled carbon-vapor synthesis in the inside of a resistively-evaporating carbon-cylinder now looks possible. However we are only beginning to examine carbon spheres created as a result.

References:
[9] Thanks to Roy Lewis at U. Chicago and T. J. Bernatowicz for the presolar specimens used here.

![Figure 1](image_url)

**Figure 1.** On-line strong-phase-object simulation of 15 faceted-nanocones in an 8-nm sphere with 26,846 “non-touching” C atoms. The projected-potential power-spectrum at right shows graphene {100} and {110} rings, as well as rel-rods associated with some of the edge-on graphene-sheets.