NEW DIRECTIONS FOR DETECTIVE WORK ON PRESOLAR CORE\RIM CARBON ONIONS.

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Introduction: The subset of pre-solar grains extracted from meteorites, and made up of s-process isotopes, likely formed around thermally-pulsing asymptotic giant branch (AGB) stars "after third dredge-up" of newly made carbon atoms [1]. In particular the core\rim subset of micron-size carbon spheres in pr-solar collections e.g. from the meteorite Murchison contain unlayered graphene sheets in their core [2, 3], whose size and abundance likely provide information on the thermal history during formation.

Since 2018 we've reported results in LPSC abstracts on: (i) the laboratory synthesis of carbon onions with an unlayered-graphene composite core [4], albeit with a cooling rate much higher than that likely during ejection from an AGB star, (ii) back of envelope estimates of that latter cooling rate [5], and (iii) quantitative modeling of graphene sheet coherence width and abundance as a function of that cooling rate [6].

Here we introduce further components of this work, namely the nanoscale study of single atom impurities and graphene nucleation/growth in presolar and laboratory core\rim onions, as well as DFT study of early "pent-first" stages in the nucleation process itself [7]. The focus here is however not on these components alone, but on the larger picture emerging about prospects for further experimental work on presolar core\rim carbon spheres. In particular, we focus on two questions: First, what might an individual core tell us about its thermal and compositional history long ago, during formation and ejection from its parent AGB star. Secondly, what might continued study of these particles disclose about the solidification of supercooled liquid carbon at low pressure, as well as its possible application to the synthesis of diffusion barriers with unprecedented capability.

The history of individual presolar cores: What can a single presolar core\rim carbon onion tell us about its history, using models (a) presently in hand, and (b) worth developing in the future, using both computer simulations and condensation experiments here on earth?

Key experimental observations here might include electron microscope measures of rim & core diameter, as well as mass-density estimates e.g. from EELS plasmon edge structure [8] and zero-loss to brightfield intensity ratio [9], electron diffraction studies of graphene sheet coherence width and abundance, HAADF studies of heavy atom distribution and abundance in the cores, electron phase studies of contrast from faceted pentacones, and of course where possible nano-SIMS studies of isotopic ratios as well. All of these observations except for the size measurements, if we want to restrict them to data from the core, may have to be done on very thin microtome slices of individual onions, and/or on torn edges of sliced onions like that shown in Fig. 1.

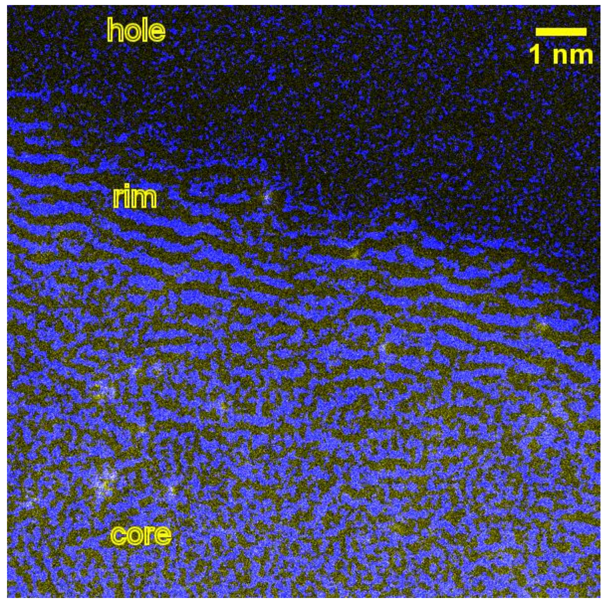

Figure 1: Composite brightfield (blue) and high angle annular darkfield (yellow) STEM image of single heavy atoms near the edge of a presolar core.

Rim and core sizes, as well as density, relate to the particle's cross-section for radiation pressure ejection from the stellar atmosphere. The ratio between core and rim diameters provide information on the integral rates of carbon deposition onto the particle before and after the solidification event. In core regions that are sufficiently thin, Z-contrast image analysis of single heavy atoms may allow atomic number determination. Given the likely diffusion barrier nature of this unlayered graphene composite, there might even be useful information contained in heavy atom compositional gradients from center to edge of the cores.

Data on graphene sheet sizes and number densities, available from core-only powder diffraction profiles as well as from electron phase-contrast images, connect to cooling rates during the likely short solidification process, as illustrated schematically in Fig. 2.

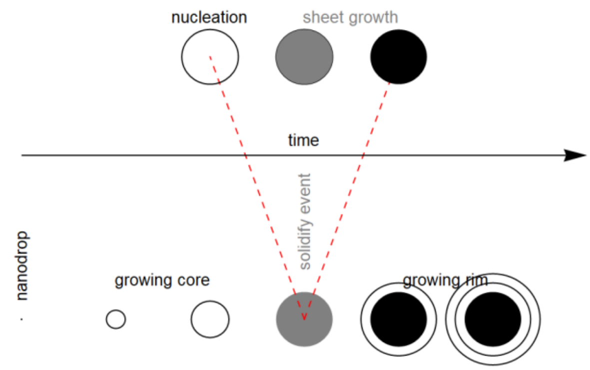

Figure 2: Schematic of the carbon vapor condensation process, showing vapor to liquid, liquid to solid, and finally vapor to solid (graphite rim) components.

So far, of course, we've only used observed sheet size and number density data to roughly calibrate the observed size and number densities in comparing presolar and laboratory formed spheres. This means that more controlled "slow cooled carbon vapor" condensation experiments in the laboratory are needed, but as we discuss below these are also relevant to fundamental studies of liquid carbon at low pressures as well.

Liquid carbon at low pressure: What can collective observations on these presolar core\rim onions further tell us about the behavior of supercooled liquid carbon droplets in containerless settings, including e.g. prospects for making unlayered graphene-composite diffusion barrier material to help with He loss from earth into space.

Although experimental evidence for carbon droplet formation following vaporization has turned up in the literature [10, 11], for us observation of presolar core\rim carbon onions has already been the catalyst for a serious look at the ways that supercooled liquid droplets emerge when carbon vapor is cooled, and in particular on how the three stages: vapor to liquid, liquid to solid, and vapor to solid manifest in the laboratory as well as in theory.

It has thus highlighted the prediction [12] that the carbon phase diagram looks quite different on the nanoscale (for both diamond as well as liquid carbon), raising the possibility that the first stages of condensation in a cooling carbon gas may involve stable (rather than metastable) liquid droplets. Laboratory as well as presolar observation of the formation of unlayered graphene sheets, further suggests that slow cooled carbon vapor condensation (at least in containerless form) through the 2000-3000K range might open the door to synthesis of graphene sheet diffusion barriers without graphite's weakness of van der Waals layering.

Since we are still perhaps 7 orders of magnitude away from being able to cool carbon vapor in containerless form as slowly as does an AGB star during carbon particle ejection, presolar core\rim carbon onions are an important resource for studying this part of carbon's phase diagram. Such particles, along with oven design experiments for laboratory slow-cooling the laboratory (perhaps in microgravity settings to reduce gravitational settling rates), may be key experimental resources to this end. These will be crucial for calibrating models of graphene sheet growth during solidification, for developing models of "saturation" as growing sheets begin to compete for fewer and fewer atoms in the liquid, and for modeling the effectiveness of graphene's hex-ring structure in slowing down the diffusion of both He atoms here on earth, as well as the heavy atoms incorporated into presolar cores from an AGB star during solidification.

The bottom line is, in both of these contexts, that more "high-density" s-process carbon separates from meteorites like Murchison might be quite valuable in the days ahead, for the study of (i) element distribution from AGB star atmospheres as well as of (ii) new uses for carbon condensation here on earth.

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