

SPATIALLY RESOLVED SP² AND SP³ CARBON IN NANODIAMOND RESIDUES FROM THE ALLENDE AND MURCHISON METEORITES

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Introduction: A critical question for the interpretation of the bulk chemical and isotopic analyses of meteoritic nanodiamond residues, and ultimately the origin of the nanodiamonds, is whether the residues contain any additional species of C. Electron energy loss spectroscopy (EELS) [1-3] of nanodiamond residues have consistently shown the presence of varying levels of sp² C in addition to the sp³ diamond. Because up to 25% of the C atoms in a 2 nm particle are at the surface, this sp² C has been attributed to deviations in the surface bonded carbon from the bulk diamond character. This interpretation is supported by prior transmission electron microscope (TEM) imaging studies that have not revealed a significant fraction of other C components in the residues [4].

In light of the recent advances in electron microscopy instrumentation that have enabled direct imaging of low-Z monolayers [5], we have revisited the possibility that a second C component is present in the residues. We present here a comparison of results from conventional TEM and aberration-corrected scanning TEM (STEM) studies of the Allende DM nanodiamond separate and a new Murchison nanodiamond separate.

Methods: The new Murchison residue was prepared at the Carnegie Institution of Washington according to established CsF-HClO₄ dissolution methods. The residues were each deposited as suspensions in high purity water onto two types of TEM support grids: lacey C with and without a ~3 nm C backing. TEM studies, including bright field imaging, electron diffraction and high-resolution imaging were carried out at the Naval Research Lab with a JEOL 2200FS, field-emission microscope operated at 200kV. Aberration-corrected STEM studies were conducted at Oak Ridge National Laboratory, with a Nion UltraSTEM 100. Annular dark-field imaging and EELS studies were performed at an operating voltage of 60 kV with a probe size of < 1.5 Å.

Results and Discussion: Conventional TEM studies of the residues on lacey C supports are consistent with prior studies: the diamond aggregates suspended over large holes in the film appear to be densely packed with little intergranular material. The same residues supported on the ultrathin C formed more well-dispersed monolayers, with disordered sheet-like C between the nanodiamonds readily visible in high resolution images. Aberration-corrected STEM and dark-field STEM imaging confirmed the presence of a distinct sheet-like C phase, as well as abundant surface impurity atoms. EELS low loss and C K-edge core loss spectra demonstrate that the sheet-like C, which is spatially distinct from the nanodiamond surfaces, is sp² bonded. This second C component may be carrier of some of the isotope anomalies observed in these residues.

References: [1] Blake D. F., et al. 1987. *Nature* 332:611-613. [2] Bernatowicz T. J., Gibbons P. C. and Lewis R. S. 1990 *ApJ* 359:246-255. [3] Garvie L. A. J. 2006 *MAPS* 41:667-672. [4] Daulton T. L. et al. 1996. *GCA* 60:4853-4872. [5] Krivanek et al. 2010. *Nature* 464:571-574.