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Do nanodiamonds look like diamonds? A TEM and EELS study of meteoritic nanodiamonds

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Nanodiamonds (NDs) are an intriguing, widespread constituent of primitive meteorites. They display a relatively uniform size distribution, with mean diameter between 2 and 3 nm. NDs in primitive unmetamorphosed chondrites occur with concentrations on the order of 0.15 wt% of the C, which is equivalent to an estimated 3 x 1017 diamonds per gram of meteorite matrix. An intriguing question is why have they not yet been identified outside our Solar System, given their abundance in our primitive pre-solar nebula? To answer this question, high-resolution transmission electron microscopy (HRTEM) and electron energy-loss spectroscopy (EELS) were used to reveal the structure and bonding of NDs from the Orgueil carbonaceous chondrite. The sample studied consists of a nanodiamond concentrate prepared by acid dissolution of the meteorite. HRTEM imaging with the aberration-corrected JEOL JEM ARM200F scanning TEM (STEM) microscope operating at 80 kV accelerating voltage, shows that the bulk of the residue is composed of ∼2-nm-sized diamond particles, with lesser amounts of an unidentified C-rich material. The C K-edge EELS spectra of the NDs acquired at ∼ 1 eV resolution are consistent in shape with that of diamond, although with broadened spectral features. In addition, the C K edge exhibits a sharp low-intensity prepeak to the main diamond spectrum; this prepeak is not present for bulk diamond. Sub-eV EELS show the prepeak to be composed of three components at 282.5, 284.7, and 286.4 eV. The low-loss spectrum of the nanodiamonds is broad with a flat-topped maximum between ∼20 to 30 eV. The maximum near 20 eV, which is also present for bulk diamond, corresponds to the surface plasmon mode. However, the bulk plasmon peak of diamond at 32 eV is not present as a distinct maximum for the NDs; instead the nanodiamond low-loss spectrum exhibits a gradual decrease in spectral intensity above ∼25 eV. In contrast to bulk diamond, the maximum corresponding to the surface plasmon is more intense than the spectral region corresponding to the bulk plasmon. The C K edge prepeak features below 288 eV, which are not present for the bulk diamond, are attributed to transitions from the C 1s surface core level to unoccupied surface states. In particular, the peak at 286.4 eV is attributed to transition to unoccupied pi* antibonding states of surface pi-bonded dimers. The importance of surface states is further revealed in the low-loss spectrum where surface-loss peak is more intense than the bulk plasmon peak. The EELS and HRTEM data are consistent with a ND particle consisting of a diamond core covered by regions of sp2-bonded carbon, possibly with surface reconstruction to fullerene-like structure, C-H dangling bonds, and a range of functional groups. While the core of the diamond does not absorb visible light, the complex surface structure will impart optical properties not present for diamond. Given the high surface-to-volume ratio of the NDs, it is expected that the surface states will contribute significantly to their optical properties. Hence, nanodiamonds in interstellar or circumstellar regions may not look like diamond. |End Text|