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Highly nitrogen and boron doped nanotubes: a route to synthesis and study of their properties by spatially resolved EELS

ANNICK LOISEAU, LEM, Cnrs-Onera, Chatillon, France

Doping C-nanotubes with B and/or N is expected to be a particular interesting way for tuning electronic and mechanical properties. BN nanotubes are predicted to behave as insulators whereas B(N) doped C-nanotubes are expected to be metallic, independent of their structure. In this framework, we have developped, both at Onera and GDPC, original routes to the synthesis of BN singlewall nanotubes (BN-SWNTs) and to highly doped nitrogen multi wall nanotubes (CN_x-MWNTs). CN_x-MWNTs were produced by a CVD method, using an aerosol injector which sprays in the reactor, heated at 950 ˚C, a liquid mixture of organic compounds with a controlled N/C ratio and suitable metal complexes as the catalyst precursors[1]. This procedure leads to dense amounts of MWNTs with controlled N/C ratios which can exceed 15-20% in average. Upon doping, tubes get a characteristic compartimentalized structure with a reduced number of layers identified in transmission electron microscopy. Using spatially resolved electron energy loss spectroscopy (EELS), N is found to be preferentially localized in inner layers and in the compartiments where the concentration can exceed 40 at.%. Structure of core losses in EELS reveals a high dependance of the N environment to the local concentration :chemical bonding of N can be graphitic, pyridinic or pyrrolic, this latter case being found for highest N concentrations. Relationships between these structural properties and formation mechanism will be discussed[2]. BN-SWNTs are issued from the vaporization of a BN target by a continuous CO_2 laser under a N_2 atmosphere[3]. We present here the first investigation on their electronic properties by two ways: first, analysis of the dielectric response of low loss EELS recorded on individual tubes provides the first identification of plasmons and of interband transitions in these tubes and the first measure of their gap found to be close to 5.8eV[4]. Second, optical absorption spectra measured on macroscopic samples strongly suggest the existence of a Frenkel exciton with a binding energy in the 1eV range[5]. 1-M. Glerup et al Chem. Commun 2542 (2003). 2-M. Castignolles et al submitted to Phys. Rev B (2005) 3- R. Lee et al, Phys. Rev. B Rapid Comm 64, 121405-1 (2001) 4-R. Arenal et al, submitted to Phys. Rev. Lett. (2005) 5-J.S. Lauret et al, Phys. Rev. Lett. (2005) in press. Coauthors: M. Castignolles[1,2], R. Arenal[1], O. Stéphan[3], M. Glerup[2,4],[1]LEM, CNRS-ONERA, Châtillon, France, [2]GDPC, Université Montpellier II, France, [3]LPS, Université Paris-Sud, Orsay, France, [4]GHMFL, MPI-CNRS, Grenoble, France.