Phonon Confinement Model for Nanodiamond Powders: Challenges in Crystal Size Measurement

Osswald S.[a], Havel M.[a,b], Mochalin V.[a], Yushin G.[a,c], Gogotsi Y.[a,#]

[a]*Department of Materials Science and Engineering and A.J. Drexel Nanotechnology Institute, Drexel University, Philadelphia, Pennsylvania 19104, USA*
[b]*Current address: Arkema, Inc., King of Prussia, Pennsylvania, USA*
[c]*Current address: Department of Materials Science and Engineering, Georgia Institute of Technology, 771 Ferst Drive, N.W., Atlanta, Georgia 30332-0245, USA*
[#] *Corresponding author: gogotsi@drexel.edu*

Today, Raman spectroscopy is a standard tool for the characterization of nanostructured materials. However, since most of their properties are size-dependent, the interpretation of Raman data becomes more difficult as one approaches the lower end of the nanoscale (10 nm or less). Several models have been developed in order to account for size effects and explain the observed changes in the Raman spectrum of confined structures such as quantum dots, nanocrystals or carbon nanotubes. The phonon confinement model (PCM) was successfully used to explain finite size effects in the Raman spectra of various materials such as GaAs, Si, and BN. However, for nanodiamond, the agreement between the theory and experimental data remains unsatisfactory and the Raman spectra are still not fully understood. The assumptions of current models oversimplify the complex structure of nanodiamond crystals and neglect contributions from, for example, the surface and lattice defects. In this work we introduce a modified PCM that accounts for lattice defects, multiple phonon dispersion relations, surface effects and changes in phonon lifetime. Moreover, we provide a simple model that allows a fast analysis of the crystal size in nanodiamond powders. Oxidation of detonation nanodiamond [1] provided by NanoBlox, Inc., was used to produce diamond powders with different crystal size [2] and the developed model was tested on those powders. The results of Raman spectroscopy measurements were compared with X-ray diffraction and transmission electron microscopy measurements. While all techniques show the same trend, somewhat different absolute values of the average crystal size were measured using different techniques. The reasons for these discrepancies will be discussed.

[1] S. Osswald, G. Yushin, V. Mochalin, S. Kucheyev, Y. Gogotsi, Control of sp^2/sp^3 carbon ratio and surface chemistry of nanodiamond powders by selective oxidation in air, *J. American Chemical Society* **128**, 11635-11642 (2006)

[2] S. Osswald, M. Havel, V. Mochalin, G. Yushin, Y. Gogotsi, Increase of Nanodiamond Crystal Size by Selective Oxidation, *Diamond and Related Materials*, (2008) in press, DOI: 10.1016/j.diamond.2008.01.102