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ACCEPTED MANUSCRIPT

The study of nitrogen inclusion in carbon nanotubes obtained by catalytic laser-induced chemical vapour deposition (C-LCVD)

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HIGHLIGHTS

- The C-LCVD is an efficient method to increase the amount of at low flows of N donor (in the present case ammonia) and low laser powers.
- Sample with the highest amount of dopant shows the best graphitization
- The relative amount of sp² C=C carbon has the same trend as I_{D4}/I_G
- The pyrrolic relative amount exhibits the same trend as $I_{2D4}/I_{G.}$

Abstract

Nitrogen doped carbon nanotubes were grown on Fe₂O₃ nanoparticles deposited on silicon substrates, by laserinduced chemical vapour deposition of acetylene/ammonia mixtures. The concentration of the nitrogen has been controlled in the range 1-6 atomic % by adjusting the flow rate of ammonia, pressure and laser power. XPS and Raman spectroscopy were used to quantitatively assess the compositional and structural properties of the nitrogen-doped carbon nanotubes (N-CNTs). First order Raman spectra were deconvoluted assuming five vibrational modes and the integrated peak intensity ratio I_D/I_G and I_{2D}/I_G of all samples are displayed. We demonstrate that the relative amount of sp^2 C=C carbon has the same trend as I_{D4}/I_G and the pyrrolic relative amount exhibits the same trend as I_{2D4}/I_G . The high resolution TEM images are consistent with the Raman and XPS results, revealing that the surface of the N-CNTs outer walls becomes more distorted at the highest content of N while the inner walls of the nanotube preserve a high crystallinity, corresponding to the lowest I_D/I_G ratio.

Keywords: Nitrogen-doped carbon nanotubes; catalyst nanoparticles; Laser-induced Chemical Vapour Deposition; localized growth

1. Introduction

The incorporation of nitrogen changes the overall microstructure of carbon nanotubes (CNTs) and subsequently alters their physical, chemical, and electrical properties. In other words, the physicochemical properties of nitrogen-doped CNTs strongly depend on the nitrogen doping level, crystallinity, and nanotube characteristics such as diameter and wall thickness [1]. Many factors including reaction temperatures, types of

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