Change of Carbon Nanotube Structure by Ion Irradiation

Mariya M. Brzhezinskaya¹, Eugen M. Baitinger¹, Vladimir V. Shnitov² and Aleksey B. Smirnov²

¹Chelyabinsk State Pedagogical University, Department of Physics 69 Lenin Ave., Chelyabinsk 454080, Russia

Phone: +7-351-2-65-09-26 E-mail: brzhezinskaya@fromru.com

² Ioffe Physico-Technical Institute of the Russian Academy of Sciences,

26 Polytekhnicheskaya St., St. Petersburg 194021, Russia

1. Introduction

Carbon nanotubes (CNTs) hold much promise for nanotechnology. Ion irradiation is a possible way of welding of CNTs.

In this paper, the results of experimental study of change of single-walled and multi-walled carbon nanotube (SWNT and MWNT) structure by ion irradiation are presented. They were obtained by X-ray photoelectron spectroscopy (XPS), reflection energy loss spectroscopy (EELS) and Auger electron spectroscopy.

2. Experimental

The samples of SWNTs and MWNTs were periodically irradiated by argon ions (Ar^+) *in situ* in the spectrometer chamber. The Ar^+ energy was 1 keV. The maximum dose of Ar^+ irradiation (*Q*) was 360 μ C/cm². The samples were degassed in ultra-high vacuum better than 10⁻⁹ Torr for 70 hours.

Each irradiation of a sample was followed by Auger spectra measurement in the mode of constant absolute energy resolution $\Delta E = 0.6$ eV in order to determine the concentration of argon absorbed in the near surface region of the sample.

The XPS measurements were carried out using ultra-high vacuum electron spectrometer PHI-5500 (produced by Perkin-Ermler) using Mg K α line. Absolute energy resolution (ΔE = Const) was 0.2 eV, when the transmission energy was 10 - 25 eV. Influence of ion irradiation on the energy of C 1s peak and on its full width at half maximum (FWHM) was determined.

Energy loss spectroscopy of reflected electrons was used: the incident angle of the primary electron beam and the registration angle were 45°, the diameter of the beam on the sample was approximately 1 mm, the analyzer aperture was 12°. Absolute energy resolution (ΔE = Const) was 0.2 eV when the transmission energy was 10 - 25 eV.

3. Results

It was found, that concentration of absorbed Ar increases with the increase of the dose of ion irradiation. The concentration of absorbed Ar reaches the maximum at 4-5 atomic percents. The process of Ar absorption by SWNTs and MWNTs reveals nonlinear character. CNTs accumulate Ar more quickly at small values of Q (Fig. 1).

The C 1s peak energy increases and the plasma peak broadens slightly with the increase of the dose Q of

Ar⁺ irradiation. The satellite photoelectron peak located at higher binding energies (the binding energies from 284.5 eV to about 330 eV) changes essentially under Ar⁺ irradiation. It was found out that the energy of π -plasmon peak decreases and it broadens essentially under Ar⁺ irradiation; the π + σ -plasmon peak has a doublet shape and changes essentially under Ar⁺ irradiation, also.

The influence of ion irradiation on the π -plasmon energy E_{π} and the full width at half maximum of the plasma peak δE_{π} were studied by EELS. The E_{π} value decreases significantly ($\Delta E_{\pi} = 0.9$ eV for SWNTs and ΔE_{π} = 1.2 eV for MWNTs) with increasing dose of Ar^+ irradiation (Fig. 2). The π -plasmon peak broadening is accompanied by its intensity decrease. The full width at half maximum δE_{π} increases from 2.7 to 3.35 eV (approximately by 24%) for SWNTs and from 2.38 to 2.8 eV (approximately by 17%) for MWNTs (Fig. 3). The determined dependence of the π -plasmon energy E_{π} on the dose Q of ion irradiation can be approximated by hyperbola, although the hyperbola coefficients are different for SWNTs and MWNTs (Fig. 4). Furthermore, it was found that the hyperbola coefficients are different at small values of Q ($Q < Q_0 = 30 \ \mu\text{C/cm}^2$) and at large values of Q ($Q > Q_0$) for SWNTs and MWNTs: accumulation of defects is slower at the beginning of irradiation.

4. Conclusions

In this presentation, possible causes of the observed effects are discussed. Phenomenologically, it is possible to use the model of the damped harmonic oscillator for quasi-one-dimensional system of carbon nanotubes. The microscopic model is proposed. The observed effects have been explained by narrowing of π -bands caused by field of charged defect and by decrease of energy of interband transitions in carbon nanotubes under ion irradiation.

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Figure 1. Dependence of argon concentration on the surface of samples on the dose of ion irradiation Q for MWNTs (triangles) and SWNTs (squares).



Figure 2. The π -plasmon energy dependence on the dose of Ar ion irradiation Q for SWNTs (a) and for MWNTs (b). The inset shows the π -plasmon energy dependence on the dose of ion irradiation Q for SWNTs in the double logarithmic scale.



Figure 3. The relation between concentration of interstitial Argon atom for MWNTs and SWNTs and the full width at half maximum (FWHM) of the plasma peak δE_{π} and. (1) – MWNTs; (2) - SWNTs.



Figure 4. The π -plasmon energy dependence on the inverse dose of ion irradiation 1/Q for SWNTs (*a*) μ for MWNTs (*b*).