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Transformation of multi walled carbon nanotubes irradiated by swift heavy ions

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ABSTRACT

The radiation stability of multiwalled carbon nanotube (MWCNT) buckypaper (BP) has been studied under extreme conditions. Samples of thick mat of MWCNT, called buckypaper, have been prepared by vacuum filtration method and were irradiated by 120 MeV Au ions with fluences ranging from 3×10^{11} ions/cm^2 to 5×10^{13} ions/cm^2. The samples were characterized by Field Emission Scanning Electron Microscopy (FESEM), High Resolution Transmission Electron Microscopy (HRTEM) and Raman spectroscopy. The surface imaging studies indicate the decrease in the average diameter of nanotubes under ion irradiation due to the sputtering of atoms from the CNT surface. Raman studies demonstrate initial healing at lower fluence, defect production at higher fluences and amorphization at highest fluence.

1. Introduction

Carbon nanotubes (CNTs) have attracted increasing attention since their discovery in 1991 by Iijima [1] and have demonstrated promising applications in various fields [2–4]. However to make them suitable for use in various applications a thin film or sheet is required, which would retain most of the properties of individual nanotubes. The fabrication of a thin membrane of randomly entangled carbon nanotubes network, called Buckypaper (BP), is a good method to provide a high concentration of nanotubes over a surface. CNT BP has applications in various fields such as in field emission devices [5], sensors [6,7], electronic devices [8] and nanotube reinforced composites [9].

Ion irradiation has proven to be a useful method to create defects on CNTs in a controlled manner and to modify the material for the required applications. CNTs have potential of applications in high radiation environment such as in space and in nuclear reactor, where these materials are exposed to various kinds of energetic ions. From the recent studies it has been found that by ion irradiation of metal-CNT composite, CNTs evolve into different other structures. This improved radiation tolerance of the composite material [10]. Many studies related to the effects of ion beam on CNTs have been reported [11–15]. Ion irradiation improves the phonon transport in CNTs leading to the enhancement of thermal conductivity of CNT mats [16]. Irradiation of CNT buckypaper with 140 keV H^+ and 3 MeV He^+ ions indicate radiation induced damage and defect removal by post irradiation annealing [17]. The theoretical work reported by Krasheninnikov et al. [18–21] provided a deep insight into irradiation induced defect production and its relation to beam characteristics. All these irradiation studies correspond to low energy (a few tens of keV to a few MeV), where nuclear energy loss (S_n) dominates. In nuclear energy loss (S_n), the incident ion loses its energy predominately by elastic collisions with the nuclei of solid. The individual nuclear collisions lead to deviation of the projectile's flight direction and give rise to a spatially extended damage distribution. However in swift heavy ions (SHIs; heavy ions with energy of the order of a few tens of MeV and higher), the incident energy is imparted dominantly to electronic sub system and is referred to as electronic energy loss (S_e). This energy is transmitted to the atomic system by electron-phonon interaction which generates a localized heating of the atomic system in the ion track of nanometric dimension. This leads to the transient thermal spike lasting for duration of the order of picoseconds. The temperature has a radial gradient from the ion path and it cools down away from the track [22]. Thus it is interesting to see the effect of SHIs on MWCNT BP. Only few studies for the MWCNT BP under SHIs are available. Earlier studies on single and multi walled CNT thin films indicate healing of the irradiation induced point defects at lower fluences and structural transformations under heavy ions [23–26].

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In this work samples of multiwalled carbon nanotube (MWCNT) BP have been prepared by vacuum filtration method. The structural modifications in MWCNT network, irradiated by Au ion beam of energy 120 MeV at different fluences, have been investigated.

2. Experimental

2.1. Sample preparation

Vacuum filtration technique was employed to prepare BP. Commercially available MWCNTs (Nanostructure and Amorphous materials, USA, diameter 20–50 nm and length 0.5–2 μm) were dispersed in 1% solution of surfactant (sodium dodecyl sulfate, SDS). The resulting suspension was then centrifuged and filtered over membrane filter under vacuum pressure. The MWCNT film thus obtained was washed with excess of deionized water and dried. Once dried; the film was carefully peeled-off from the surface of the membrane filter. The sample was then cut into pieces of 1×1 cm^2 and placed on silicon substrate for irradiation.

2.2. Irradiation and characterization

The irradiation experiment was carried out with Au ions of energy 120 MeV, using 15 UD Pelletron accelerator, at Inter University Accelerator Centre (IUAC) New Delhi. During irradiation the pressure of vacuum chamber was of the order of 10^{-6} mbar and average ion current was 0.5 Particle per nanoampere; pnA (1 pnA = 6.25×10^9 ions/s). The range of the ion fluence is from 3×10^{11} ions/cm^2 to 5×10^{13} ions/cm^2. The electronic energy loss (S_e), nuclear energy loss (S_n) and projectile range of ions was calculated by using 'stopping and range of ions in matter' (SRIM) simulation code which yields the S_e and S_n values as 1073 eV/Å and 12.7 eV/Å respectively [27].

The structural modifications in MWCNTs under ion irradiation were investigated by using field emission scanning electron microscopy (FESEM), High resolution transmission electron microscopy (HR-TEM) and Raman Spectroscopy. The surface morphology was studied using FESEM (MIRA II LMH from TESCAN) and HRTEM (FEI Technai G2 S-TWIN TEM). Raman spectra were recorded by using Renishaw inVia micro Raman set up. Raman measurements for pristine and irradiated samples were carried out at room temperature with Ar ion laser, with excitation wavelength of 514 nm, over a range 100–3200 cm^{-1}.

3. Results and discussion

3.1. Field emission scanning electron microscopy

The surface morphology of the samples before and after irradiation, recorded by FESEM, is shown the Fig. 1. A dense network of

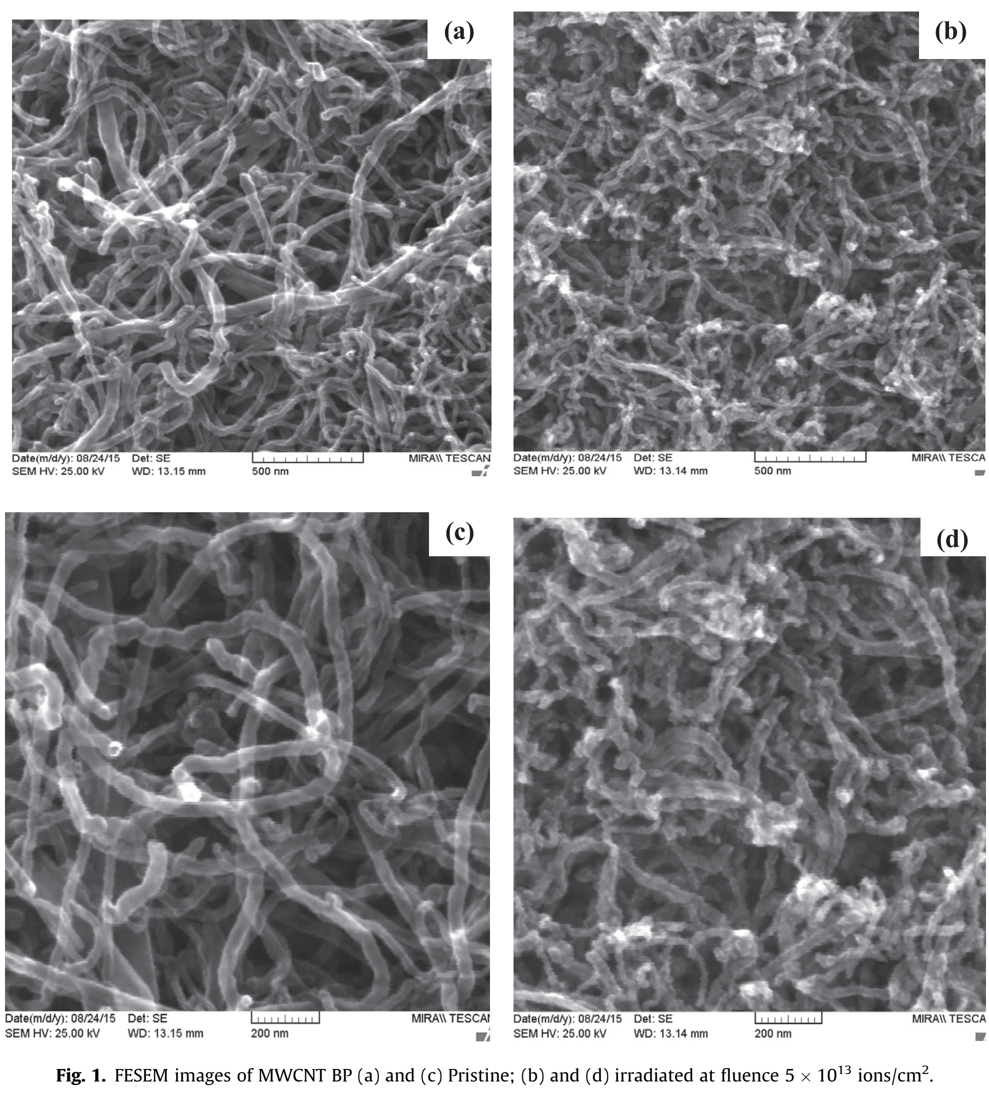

Fig. 1. FESEM images of MWCNT BP (a) and (c) Pristine; (b) and (d) irradiated at fluence 5×10^{13} ions/cm^2.