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ScienceDirect
NUCLEAR SCIENCE AND TECHNIQUES
Nuclear Science and Techniques, Vol.18, No.3 (2007) 137–140

Intensive irradiation of carbon nanotubes by Si ion beam

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Abstract Multi-walled carbon nanotubes were irradiated with 40 keV Si ion beam to a dose of 1×10^{17} cm^{-2}. The multiple-way carbon nanowire junctions and the Si doping in carbon nanowires were realized. Moreover, the formation processes of carbon nanowire junctions and the corresponding mechanism were studied.

Key words Carbon nanotube, Carbon nanowire, Junction, Ion beam, Doping

CLC numbers TB383, TN304.1$^+$8, TN305.3

1 Introduction

The miniaturization of components for making electronic and optoelectronic devices is an essential requirement of modern technology development. The nanoscale interconnections among/inside the building blocks are required for achieving further miniaturization towards nanoscale devices. One-dimensional carbon nanostructures, such as nanotubes and nanowires, have become important nano-materials due to their unique mechanical and electronic properties[1]. Some nanoscale electronic and optoelectronic devices can be fabricated now through simple interconnection using metallic or semiconductor nanowires[2,3], but there are still difficulties in producing complicated devices. One of the most dominating difficulties is the generation of various multiple-way junctions. Moreover, it is also an open question to dope some elements in the junctions and nanowires to tune their electronic or optoelectronic characteristics.

Recent studies indicate that multiple-way junctions of carbon nanotubes (CNTs) and carbon nanowires (CNWs) can be synthesized using arc discharge[4], chemical vapor deposition[5-8], and ion beam irradiation[9]. Wang et al. [9] reported the fabrication of CNW junctions (CNJs) produced by C ion beam irradiation. They confirmed the formation of CNJs through TEM observation. As well known, the implantation of the energetic ion beams can be used for not only the controlled metastable phase formation but also the material modification by doping[10,11]. The atom doping will possibly tune properties of the CNWs, including the conductance and mechanical properties.

In our previous work [12], we realized the fabrication of CNW networks through Si ion beam irradiation. In this paper, we focus on the formation of CNJs and the doping of CNWs induced by Si ion beam irradiation. The formation mechanism of CNJs is proposed.

2 Experimental details

Multi-walled CNTs were synthesized by chemical vapor deposition methods on catalyst iron particles. Purified CNTs were dispersed on silicon substrates or holey carbon micro-grids, and were irradiated with 40 keV Si ion beam to a dose of 1×10^{17}cm^{-2} on an electromagnetic isotope separator (EMIS), which kept at about ~10^{-3} Pa during the irradiation. And the substrate temperature is at room temperature. Morphology and

Supported by the National Natural Science Foundation of China (Grant No.10375085) and the Chinese Academy of Sciences Knowledge Innovation Program (Grant No. KJCX3.SYW.N10).

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Received date: 2006-03-05

structure of the CNTs before and after irradiation were examined by scanning electron microscopy (SEM, LEO 1530VP) and transmission electron microscopy (TEM) (JEOL 2010 and Tecnai F30).

3 Results and discussion

Typical morphology and structure of as-grown CNTs are shown in Fig.1. Diameters of the as-grown CNTs (Fig.1a) range from 18 nm to 35 nm. TEM images revealed that the as-grown CNTs were of hollow structure (Fig.1b) and the formation of junctions was not observed. The high-resolution TEM (HRTEM) images in Fig.1c revealed well-ordered graphitic sheets in [002] orientation (average plane spacing ~0.34 nm) with some disordered graphitic lattice in the outer walls.

During the ion bombardments, collision cascades generated large quantities of vacancies and interstitials in the CNTs walls and between the walls. Consequently, CNTs were rendered into solid CNWs at sufficiently high doses. As confirmed by TEM and HRTEM observations, CNWs were formed after irradiating the CNTs with 1×10^{17} cm^{-2} 40 keV Si ions (Fig.2 b). The HRTEM image in Fig.2b (the upper left insert) showed that the CNWs were of amorphous phase. And the selective area electron diffraction pattern (SAD) inserted in the lower left of Fig.2b showed typical amorphous halo rings, which confirmed the formation of solid amorphous CNWs. At the same time, the CNJs with different shapes were induced as shown in Fig.2.

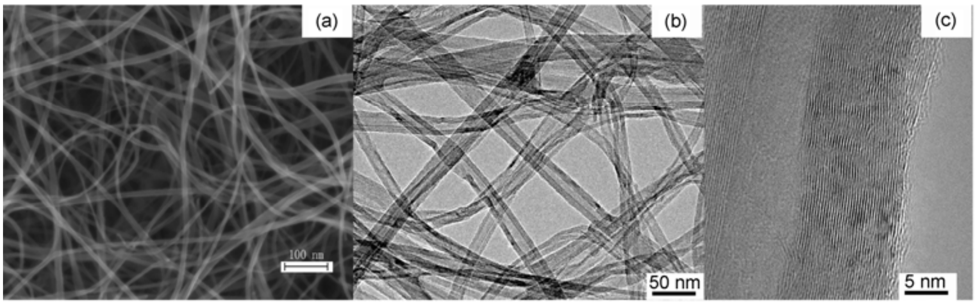

Fig.1 Typical morphology and structure of the as-grown CNTs.
(a) SEM image, (b) TEM image, (c) HRTEM image.

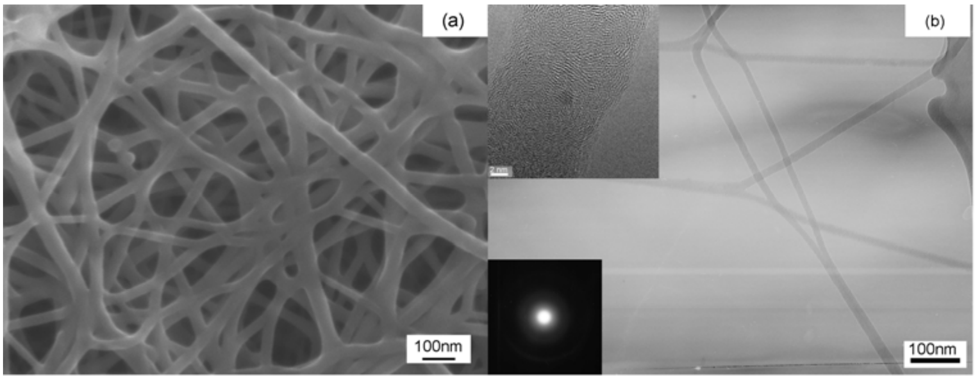

Fig.2 Typical SEM and TEM images of the CNTs irradiated with 40 keV Si ions to a dose of 1×10^{17} cm^{-2}.
(a) SEM image, (b) TEM image. The insets are HRTEM (upper left) and SAD (low left) images.

Detailed HRTEM observations of the CNJs (Fig.3) show that junctions in “Y”, compressed “Y” or “K” shape, were fabricated by the Si ion bombardment. The CNJs consist of amorphous phase. During formation of the CNWs, some carbon atoms recoiled under the Si ion bombardment on the CNTs, bonded together with vacancies in surface of the CNTs or CNWs, and finally formed an interconnected junction between adjacent crossed CNTs or CNWs. The junction shapes depend on style of adjacent crossed CNTs or CNWs. Except for the formation of CNJs of different shapes, CNWs networks were also fabricated, as reported in our previous paper [12].

To understand chemical composition and element distribution in the CNJs, the specimens were studied with electron energy loss spectroscopy (EELS). Typical bright-field TEM image and EELS spectra of the CNW (Fig.4) show that a small quantity of Si atoms was incorporated into CNJs. Besides, energy dispersive X-ray (EDX) analysis has confirmed that the Si atoms are about 2 wt% implanted in the CNTs specimens.

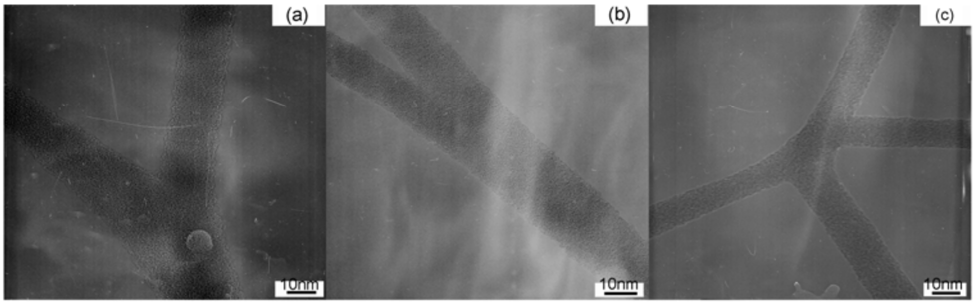

Fig.3 HRTEM images of CNJs in “Y” (a), compressed “Y” (b), or “K” (c) shape.

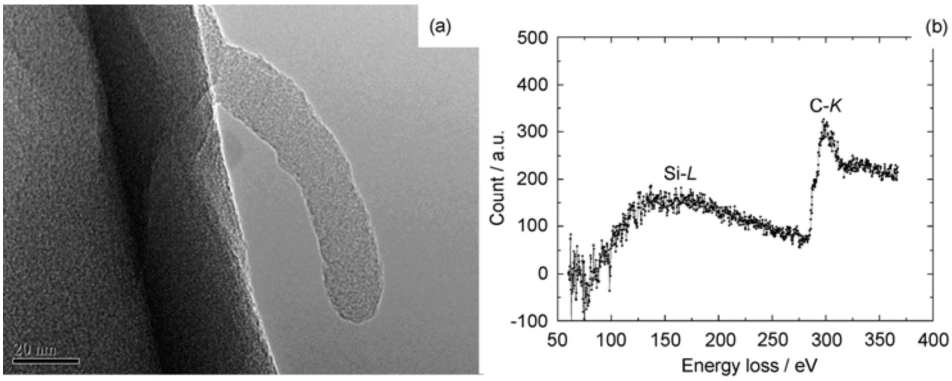

Fig.4 Bright-field TEM image (a) and typical EELS spectra (b) of the CNW.

Many experimental and theoretical studies have been done on synthesis and fabrication of carbon nanotube junctions and CNJs [4-8, 13-17]. Some groups fabricated carbon nanotube junctions using electron beams, [16,17] while other groups focused on theoretical calculations of ion beam induced carbon nanotube junctions by molecular dynamics[13,15]. However, to the authors’ knowledge so far, large-scale fabrication of carbon nanotube junctions can hardly be achieved through electron irradiation, and limited efforts were made in experimental fabrication of carbon nanotube junctions and CNJs using ion beams, while little has been known about mechanisms of CNJs formation induced by ion beam bombardment.

Based on our experimental results, we proposed a two-step process for the formation of CNJs as schematically demonstrated in Fig.5. Firstly, the ion irradiation leads to a gradual amorphization of the nanotube (Fig.5 b). During irradiation, the energetic Si ions transfer energy to atoms in the uppermost shell of CNTs and create several primary carbon atom recoils and single- or multiple vacancies. The carbon atom recoils produce more recoil atoms as they collide with carbon atoms in other shells. Large quantities of vacancies and interstitials are generated by the recoiling carbon atoms and impinging Si ions. This makes nanotubes in a highly unstable structure and induces rearrangements of carbon atoms within graphitic structures around vacancies to reduce surface free energy. As a result, an amorphous region can be formed. Secondly, irradiating two crossed nanotubes to certain doses induces the formation of bonds to bridge the CNWs and successively to the formation of CNJs with different shapes (Fig. 5c). Experiments and theoretical simulations have demonstrated that ion bombardment can create various junctions between crossed CNTs[9,15]. Molecular dynamics simulation [15] shows that single-walled CNTs welding is mediated by dangling bond saturation and carbon network reconstruction near the radiation-induced vacancies in the junction area. Being much heavier than carbon atoms, the Si ions can cause more forward recoils of the carbon atoms, which generate more carbon atom recoils in the inner part of loosely distributed CNTs films. Some of the carbon recoils may bond with the vacancies in surface of the CNTs or CNWs and finally form an interconnected junction between adjacent CNWs. A quantitative understanding of the mechanism, however, requires knowledge of size effects of the as-grown CNTs, the energy and mass of incident ions, and the substrate temperature, etc. Theoretical simulations of the interactions between energetic particles and CNTs with molecular dynamics are under way.

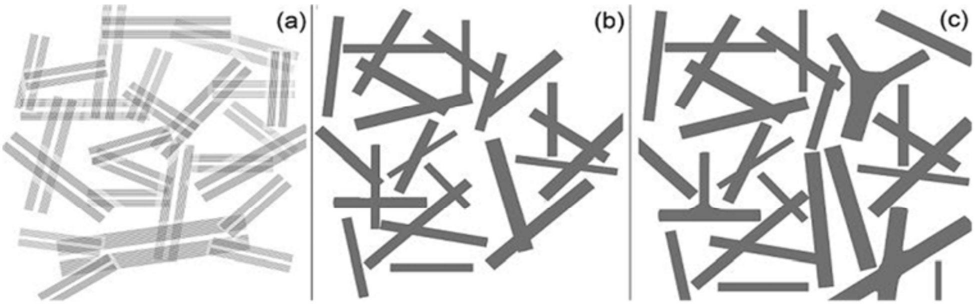

Fig.5 Schematic processes of the CNJs synthesis. (a) As-grown CNTs, (b) Defect generation to form amorphous region, (c) Formation of CNJs with different shapes.

4 Conclusions

In summary, we realized the fabrication of CNJs and the doping of CNWs by Si ion beam irradiation. Based on the interactions between energetic particles and CNTs, we present a model to explain the formation of CNJs and corresponding mechanism. Our results provide insights into the structural change of CNTs and formation of CNJs by ion beam irradiation, and help us understand the interaction between energetic particles with the solids and even achieve controllable devices fabrication in nanoelectronics. Combined the ion irradiation technology with current nanotechnology, it is of great significance for miniaturization and large-scale fabrication of electronic and optoelectronic devices.

Acknowledgments

The authors would like to thank Mr. HUANG Jianming and YE Bonian for their assistance in specimen treatments.

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