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Morphology-directing transformation of carbon nanotubes under the irradiation of pulsed laser with different pulsed duration

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ABSTRACT

Carbon nanowires have many interesting properties in electronic, opto-electronic and nanoelectromechanical devices due to their high aspect ratios. Carbon nanotubes are ideal templates for the synthesis of one-dimensional carbon nanowires. T-carbon nanowires have been recently produced by the pseudo-topotactic conversion of multi-walled carbon nanotubes (MWCNTs) under picosecond laser irradiation. There is no data about the relationship between the carbon product morphology and laser pulse duration reported so far. Pulsed laser induced reaction is a fast and far from equilibrium process. Carbon nanostructures with different morphology have been produced from carbon nanotube suspension in methanol under laser irradiation with different pulse duration. Carbon nanoparticles, porous carbon nanorods, and even diamond nanobundles have been obtained under nanosecond laser irradiation. The one-dimensional template growth from MWCNTs to carbon nanowires is not successful under nanosecond laser irradiation. However, different carbon nanowires with diameters similar to shortened carbon nanotubes have been easily produced from carbon nanotube suspension under the irradiation of picosecond and femtosecond pulsed laser. The morphology-directing transformations of carbon nanotubes have also explained using reported mechanisms.

1. Introduction

The properties of carbon nanomaterials are dramatically affected by their morphologies. Electron of carbon nanowires are quantum confined laterally, resulting in special properties for nanoscale devices. Top-down and bottom-up are two basic approaches to synthesize carbon nanowires. Top-down approach is limited to size and shape resolution. Bottom-up approach is adopted for most synthesis techniques. Morphology-directing synthesis of different carbon allotrope is a big challenge for chemists. Template-growth is a feasible path to morphology-directing synthesis. Carbon nanotubes are ideal one-dimensional templates for the synthesis of carbon nanowires. Carbon nanotubes (CNTs) have been adopted as one-dimensional templates to synthesize one-dimensional nanostructures as nanoreactors [1–9]. However, the investigation of direct transformations of CNTs to other carbon allotrope nanowires is limited [10,11]. Pulsed-laser-induced liquid-solid interface reaction is a fast and far from equilibrium process. Instantaneous high temperature and high pressure are produced in the laser-material interfaces and then immediately quenched. The morphology of the starting materials can be preserved if the laser-material interaction process is short enough to inhibit nucleation and growth processes [12,13]. Diamond nanodots [14–17] or onion carbon nanoparticles [18] instead of nanowires have been produced from different hybridized carbon nanostructures under pulsed laser irradiation. The synthesis of morphology-directing nanostructures is still a great challenge. Recently, T-carbon nanowires have been successfully synthesized by pseudo-topotactic conversion of carbon nanotubes in methanol under the irradiation of picosecond laser [19]. The morphology-directing conversion of CNTs into carbon nanowires has not yet been explored under the irradiation of nanosecond or femtosecond pulsed laser in solution [10,17,20–30].

Herein, we report the dependence of product morphology on the laser pulse duration and its corresponding transformation mechanism. Nanoparticles, porous nanorods, diamond nanowires, and various carbon nanostructures have been obtained from the multi-walled carbon nanotubes (MWCNTs) suspension under the

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irradiation of nanosecond pulsed laser. Carbon nanowires with diameters similar to shortened MWCNTs have been readily produced from MWCNT templates under the irradiation of picosecond and femtosecond pulsed laser. The melting, nucleation, and growth mechanism has been adopted to explain the morphology of carbon products from nanosecond laser irradiation. Photon excitation, compression, and shear mechanism has been adopted to explain the morphology of carbon products from picosecond and femtosecond laser irradiation.

2. Materials and methods

2.1. Material preparation

The MWCNTs with diameters of 10–20 nm were synthesized by a catalytic chemical vapor deposition (CCVD) method [31–32] and were cut from millimeters long to 100–200 nm [33] (Fig. S1). The shortened MWCNTs (0.5 mg) were well dispersed in absolute methanol (40 mL). The suspension was then transferred into a self-designed quartz container with an optical path length of 40 mm. The sample was irradiated by laser in a nitrogen atmosphere and kept stirring with a magnetic stir bar. The laser was focused at 0.5 cm away from the front wall of the self-designed container with a beam size of 0.5 mm inside the liquid. The irradiation time of nanosecond and picosecond pulsed-laser is one hour. The reaction time of femtosecond pulsed-laser is half hour.

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Pulsed laser with three different pulse duration, nanosecond, picosecond and femtosecond were used for irradiation reactions. A Q-switched Nd:YAG laser with a wavelength of 532 nm, pulse duration of 10 ns, repetition rate of 100 Hz, and pulse energy of 200–600 mJ was applied as nanosecond pulsed laser. A high-Q-switched Nd:VAN laser was used with a wavelength of 532 nm, pulse duration of 10 ps, repetition rate of 1000 Hz, and pulse energy of 65–95 μJ was applied as picosecond pulsed laser. The Ti:sapphire laser with a wavelength of 800 nm, pulse duration of 120 fs, repetition rate of 1000 Hz, and pulse energy of 0.2–1.5 mJ was used as femtosecond pulsed laser.

2.2. Material characterization

HRTEM images were acquired by JEOL JEM-2100F transmission electron microscopy (acceleration voltage: 200 kV).

3. Results and discussion

MWCNTs were prepared by a CVD method subsequently shortened by a sonication method to improve their dispersion [19]. Carbon nanostructures with different morphology have been obtained from MWCNT suspensions under the irradiation of different pulsed laser (Fig. 1).

Crystalline carbon nanoparticles instead of nanowires were observed in the products of MWCNT suspension after irradiation of nanosecond pulsed laser with pulse energy of 200–600 mJ for one hour (Fig. 1a). Crystalline carbon nanoparticles with different shapes wrapped by amorphous carbons were produced from MWCNT suspensions in methanol under nanosecond laser irradiation. No carbon nanowires were observed from the products under the irradiation of nanosecond laser. However, carbon nanowires with diameters similar to those of shortened MWCNTs were easily produced under the irradiation of picosecond and femtosecond lasers. The carbon nanowires were also surrounded by amorphous structures (Fig. 1b & c). The carbon nanowires produced from the picosecond laser irradiation have been assigned to be T-carbon nanowires [19] (Fig. 1b). The carbon nanowires from femtosecond laser were observed to have much larger interplanar crystal spacing (2.85 Å, Fig. 1c) than that of T-carbon nanowires (1.95 Å, Fig. 1b). The length and diameter distribution of carbon nanowires from femtosecond laser is similar to those from picosecond laser (Fig. 2). Most of the nanowires have length about 100–150 nm (Fig. 2a). The diameters of the carbon nanowires from MWCNT suspensions under the irradiation of picosecond and femtosecond laser are mainly at 10–20 nm (Fig. 2b), which is analogous to the diameters from shortened MWCNTs. The shape of the pristine shortened MWCNTs with diameters around 10–20 nm has been maintained by the as-produced nanowires. The pseudo-topotactic conversion of MWCNTs to carbon nanowires were observed under the irradiation of picosecond pulsed laser with pulse energy of 65–95 μJ and femtosecond pulsed laser with pulse energy of 0.2–1.5 mJ.

Strong carbon and weak oxygen signals were detected from the EDS spectrum of the samples after irradiation by different laser (Fig. 3), where the oxygen is from the amorphous structure (Fig. S2) and the copper is from copper grid. The EDS results confirmed that the nanoparticles and nanowires from nanosecond and femtosecond laser are carbon nanostructures. The EDS results are well consistent with elemental analysis of T-carbon nanowires under the irradiation of picosecond laser, which has been reported recently [19].

Different powers were also tried by nanosecond pulsed laser. Only amorphous structures with no specific shapes were obtained when the pulse energy is under 100 mJ (Fig. 4c), which might also due to the reaction of methanol solvent (Fig. S3). Porous carbon rods with shape and size similar to that of MWCNTs were obtained when the pulse energy is more than 700 mJ (Fig. 4d). The rod crystals are composed of numerous small crystals. Few diamond nanobundles with morphology similar to MWCNT nanobundles were observed sometimes in the products under nanosecond laser (~300 mJ) irradiation (Fig. 4a). The interplanar crystal spacing of 2.05 Å corresponding to (1 1 1) lattice plane of diamond with cubic structure was clearly observed in the diamond nanowires (Fig. 4a). The SAED pattern is consistent well with that of diamond (Fig. 4b). However, the diamond nanowires are mix with carbon nanoparticles as shown in Fig. 1a. Further characterization is limited since the production yield of the diamond NWs is not high and the repeatability is very low.

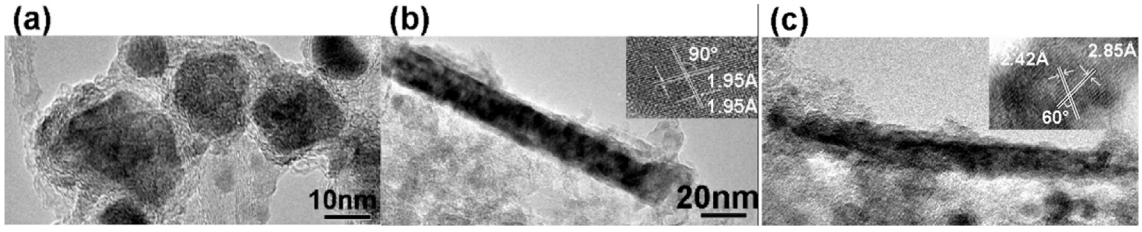

Fig. 1. HRTEM images of (a) carbon nanoparticles produced from nanosecond laser irradiation with pulse energy of 200–600 mJ, (b) carbon nanowire produced from picosecond laser irradiation with pulse energy of 65–95 μJ, (c) carbon NWs produced from femtosecond laser irradiation with pulse energy of 0.2–1.5 mJ.