Synthesis of Carbon Nanotubes from Mesophase Pitch under Arc-jet Plasma Conditions

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Abstract

Multi-walled carbon nanotubes (MWCNTs) were successfully synthesized from mesophase pitch using arc-jet plasma. The obtained MWCNTs were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). The SEM study reveals that the purity of as-produced carbon nanotubes with a length over several microns is quite high. The TEM study shows that all the nanotubes are bamboo-like, and have a diameter in a range of 50-150 nm. Our results demonstrate that mesophase pitch is an ideal carbon source for the mass production of CNTs with arc-jet method.

Keywords: carbon nanotubes, arc-jet plasma, mesophase pitch.

1. Introduction

Since the discovery of carbon nanotubes (CNTs) by Iijima in 1991 [1], CNTs has drawn much attention all over the world because of their peculiar characteristics such as electronic and mechanical properties [2-4]. In order to obtain high quality and low cost CNTs in high yield, various preparation methods have been developed, including arc-discharge, laser evaporation, chemical vapor deposition (CVD), etc. Up to now, the arc-discharge method has been widely used to synthesize high quality CNTs due to its simplicity and convenience of operation. However, it is evident that the conventional DC arc discharge method could not be easily scaled up and the synthesis rate of CNTs is limited by the evaporation of anode. In principle, all carbon-containing materials such as graphite, coal, hydrocarbons (CH_4, C_2H_4, C_2H_2, C_6H_6, etc.) and CO can be used as carbon sources for the synthesis of CNTs [5-8], but mesophase pitch, to the best of our knowledge, has never been used before as carbon source to synthesize CNTs.

Here we report that carbon nanotubes can be produced by pyrolysis of mesophase pitch with arc-jet plasma instead of arcing graphite or conductive coke. This novel approach is easy to operate and simple to scale up, which might be of potential as an efficient way for making high quality CNTs.

2. Experimental

The schematic of arc jet plasma set-up is shown in Fig. 1, which mainly consists an arc plasma generator and a reactor. The preparation experiments were carried out at atmospheric pressure rather than under vacuum conditions that is essential for conventional arc discharge method. The experimental parameters of arc-jet plasma used in the present study are shown in Table 1.

Table 1 Experimental parameters

Carrier gas (1) m^3/h	Carrier gas (2) m^3/h	Current A	Voltage V	Reaction time s
4	5	220-240	140-150	120

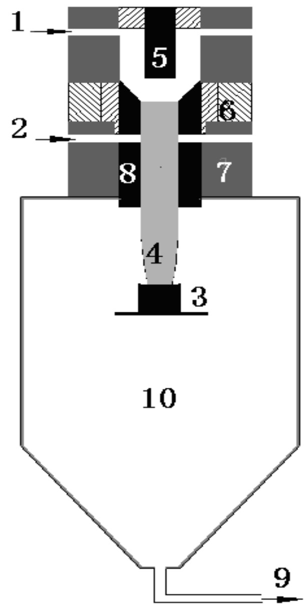

Fig. 1 Schematic drawing of the setup for making carbon nanotubes from pitch under arc jet plasma conditions; (1) cathode working gas, (2) anode working gas, (3) graphite boat, (4) arc plasma torch, (5) cathode, (6) insulator, (7) cooling water, (8) anode, (9) vent, (10) reactor.

In the present work, the feasibility of producing CNTs from mesophase pitch is explored. The chunk of pitch was ground into powder in mortar (<150 mesh), and then mixed finely with iron power in 1:1 mass ratio. The iron-pitch mixture was packed into a graphite boat that was put into the reactor under the nozzle of plasma jet. The distance between the torch plasma nozzle and the graphite boat was *ca.* 40 mm. The plasma torch was ignited by RF and for each run, the experiment lasted about 120 seconds.

The obtained products were characterized using scanning electron microscopy (SEM, JSM-5600LV), XRD and transmission electron microscopy (TEM, Philips Tecnai G^2 20). JEM-2010F equipped with energy-dispersive X-ray (EDX, Oxford instrument model 6587) was used to analyze the chemical composition of the CNTs.

3. Results and Discussion

The typical XRD pattern of carbon nanotubes (CNTs) is shown in Fig. 2. It can be clearly seen from Fig. 2 that the d_{002} peak of CNTs shifted to 25.24, and the interlayer spacing of CNTs is close to that of pure graphite. The XRD analysis also reveals that α-Fe and iron carbide are present in the CNTs products. These two species may play an important role in the formation of CNTs, which is evidenced by the fact that some CNTs are found to grow on these particles, as can be seen in Fig. 4a.

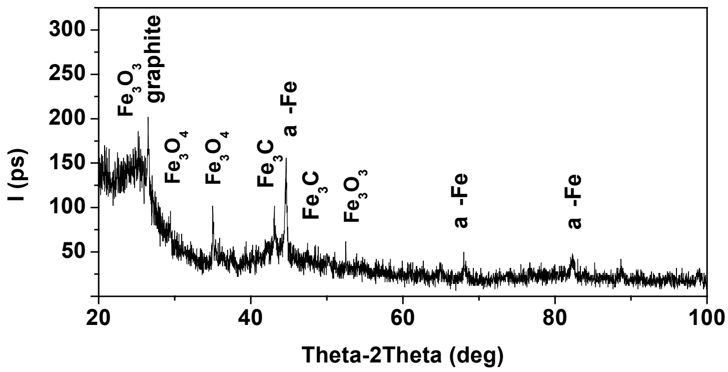

Fig. 2 Typical XRD pattern of as-synthesized CNTs

Fig. 3 shows one typical SEM image of the as-synthesized CNTs, from which it can be seen that the as-synthesized CNTs have fiber-like morphology and a quite good purity, in which very little impurities can be seen. It is estimated that the length of CNTs is over ten micrometers.

Fig. 4 shows the TEM images of CNTs, from which it can be seen that the diameters of the tubes are not uniform, which are approximately in a range of 50~150 nm, some big CNTs with a diameter over 180 nm can be observed occasionally. It is interesting to note that most of the tubes have bamboo-like morphology (as can be seen in Fig. 4b), the central cavities of CNTs are divided into a number of isolated small caves or departments along the length of the tubes. The formation of bamboo-like structures may be related to the special C fragment species and the presence of catalyst Fe.

It has been found that under conditions adopted in the present work, the synthesis time is a crucial factor for the formation of CNTs. When the operation time was shorter than 1 min or longer than 4 min, little CNTs were obtained. The detailed reasons for this phenomenon are not clear right now. The optimum time for the growth of CNTs under the experimental conditions is about 120-150 s.

Our preliminary results presented here show that mesophase pitch might become a suitable carbon source for producing CNTs in large scale, however, the mechanism involved in the process is still unclear at the moment. Whatever the mechanism is, the formation of CNTs must be closely related to the chemical composition of pitch. For future work, the influence of the chemical composition of mesophase pitch on the formation of CNTs must be taken into consideration.

Our previous work [5] has demonstrated that bamboo-like CNTs can be produced from coal with the aid of Fe by traditional arc-discharge, of which the formation mechanism is different from that of graphite. In the present work, a large amount of carbon species would be quickly released from pitch under the experimental arcing conditions because of the high temperature and peculiar chemical composition of pitch, which may be the reason for the rapid growth and wide diameter distribution of pitch-derived CNTs. Large active carbon species or primary fragments consisting of several benzene rings that are released from the decomposition or vaporization of pitch may be directly incorporated into the CNTs as building blocks. In this regard, more work is needed to clarify the formation mechanism of bamboo-like CNTs derived from pitch.

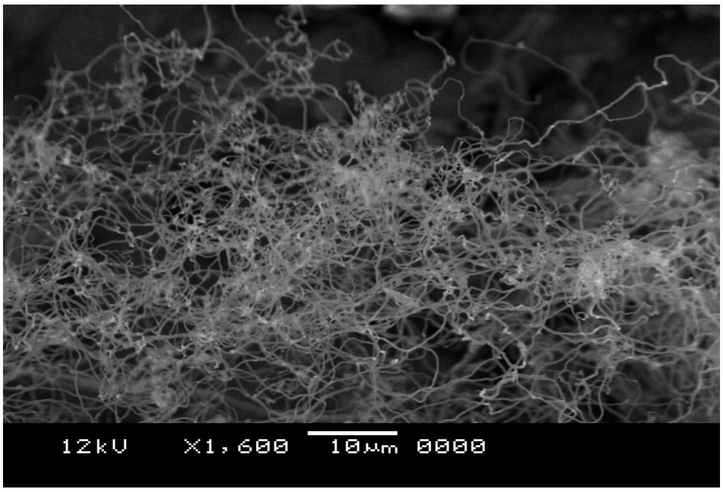

Fig. 3 Typical SEM image of CNTs from mesophase pitch under arc-jet plasma conditions.

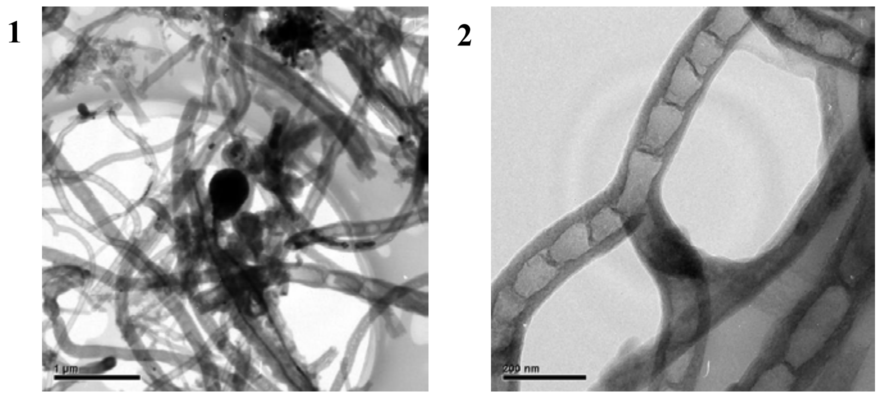

Fig. 4 TEM images of CNTs; (1) TEM image of as-synthesized CNTs; (2) a typical TEM image of CNTs, showing bamboo-like characteristics of the tubes.

4. Conclusions

Carbon nanotubes (CNTs) from pitch have been successfully synthesized with non-transferred arc-jet plasma. This approach might become a promising method to synthesize low cost CNTs in large scale with pitch-like carbons as precursor. The work is still ongoing in our lab to further optimize the experimental conditions for making high quality CNTs from pitch in high yields.

Acknowledgments

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