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Continuous process of carbon nanotubes synthesis by decomposition of methane using an arc-jet plasma

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Abstract

We present a method of producing carbon nanotubes by means of the thermal plasma decomposition of methane in an arc-jet plasma of high temperature (5000–20,000 K). Carbon nanotubes are produced under a floating condition by introducing methane and a mixture of Ni–Y powders into the arc-jet plasma flame generated by a non-transferred plasma torch. Material evaluations of the synthesized product by transmission electron microscopy (TEM) and scanning electron microscopy (SEM) reveal that the growth rate of carbon nanotubes is very high, and that the multi-walled carbon nanotubes of high purity are mainly produced. Since this process is continuously operable and easily scalable, it is expected to be a promising technique for large-scale commercial production of carbon nanotubes.

Keywords: Carbon nanotubes; Arc-jet plasma; Thermal plasma process; Methane decomposition

1. Introduction

Because of their unique properties, carbon nanotubes (CNTs) have a great potential of practical applications to field emission display [1], reinforcing component [2], hydrogen storage medium [3,4], and so on. However, in order to realize their industrial applications, a continuous process for large-scale production of CNTs is essentially required. To date, several methods such as arc discharge [5,6], laser ablation [7], chemical vapor deposition (CVD) [8] and catalytic pyrolysis [9,10] have been widely used for CNTs production.

It is known that the conventional arc discharge method using carbon rods as the electrodes can generate highly crystallized CNTs by a high temperature ambience (above 5000 K) of arc discharge. However, it has serious drawbacks of poor purity synthesis and non-continuous process for its commercial use. In this conventional process, the carbon electrode is a carbon source material for CNTs synthesis. The carbon electrodes in a reactor are consumed by arc erosion at a certain rate, and the reaction period is determined by the limited lifetime of the consumable electrodes. Furthermore, the product has to be collected around the reactor inner wall. In this case, the process is non-continuous because the product cannot be collected without a vacuum destruction in a limited vacuum capacity of the reactor.

Therefore, numerous researches have been conducted in order to improve the synthesis CNT purity and to achieve a continuous process in the arc discharge method. Ananzawa et al. [11] reported that purity could be improved from the stabilized arc by equipping a permanent magnet. Kanai et al. [12] stabilized the arc in a gravity-free state to get an increased purity, while Zhao and Liu [13] studied the effects of the chamber temperature on the improvement of purity. For making an attempt to get a continuous process, synthesis methods by arc discharge in water or liquid have been tried [14–16]. But, because the electrodes were used as a source material, the electrode lifetime is still limited. As another attempt for a continuous process, Tian et al. [17] developed a method of the coal derived CNTs synthesis by injecting coal and copper into the thermal plasma jet. However, CNTs were found in the carbon product only deposited inside the torch where the product cannot be continuously collected.

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Table 1
Comparison of the arc-jet plasma method with different arc plasma method

Method	Process continuity	Buffer material	Feedstock	Catalyst	Pressure (Torr)	Arc power (kw)	Crystallinity	Synthesis purity
Conventional method [5,24,25]	low	He, Ar	graphite	Ni, Y, Co, Fe	300–600	1.5–4.0	high	low
Arc method in liquid [14–16]	medium	water, liquid nitrogen	graphite	Y, Gd, Ni	760	1.0–3.4	high	low
Thermal plasma jet method [17]	medium	$Ar+H_2$	coal	Cu	760	25.2	low	low
Arc-jet plasma method	high	Ar+He	methane	Ni, Y	500	15	low	high

In this paper, we present a promising technique for the continuous large-scale production of CNTs by decomposition of hydrocarbons in an arc-jet plasma. The arc-jet plasma is generated by blowing a cold plasma forming gas into the current path of dc arc discharge between two metal electrodes. Since the arc-jet plasma has thermal plasma properties of high temperature (5000–20,000 K) and high velocity (100–1000 m/s) [18,26], it has been widely used for plasma spray coating [19,20], fine powder synthesis [21,22], and hazardous waste treatment by pyrolysis of organic materials and melting of solid wastes [23]. In our new method of arc-jet plasma, the metal electrodes are not used as a carbon source material, and the plasma jet is used just as a heat source. The lifetimes of the W cathode and the Cu nozzle anode are very long compared to the consumable carbon electrode used in the conventional arc method. And the carbon source of gas phase like methane and the catalyst source like Ni and Y are introduced into the thermal plasma jet independently from the injection elements. So the feedstock can be continuously supplied for the CNTs synthesis. In addition, because the synthesized products are moving along the plasma flow, we can easily collect the produced CNTs nanotubes in the gas filter through some modification of the exhaust system. In Table 1, the arc-jet plasma method is compared with different arc plasma method in detail.

In the synthesis of CNTs by the arc-jet plasma method, additional process features compared with a pyrolysis method are expected as follows: (i) It is possible to achieve the high rates of hydrocarbon decomposition and CNT growth resulted from the high temperature and enthalpy of thermal plasma; (ii) This technique itself is regarded as a process for the metal nano-particle synthesis [22] due to evaporation of the metal powder of large size in the thermal

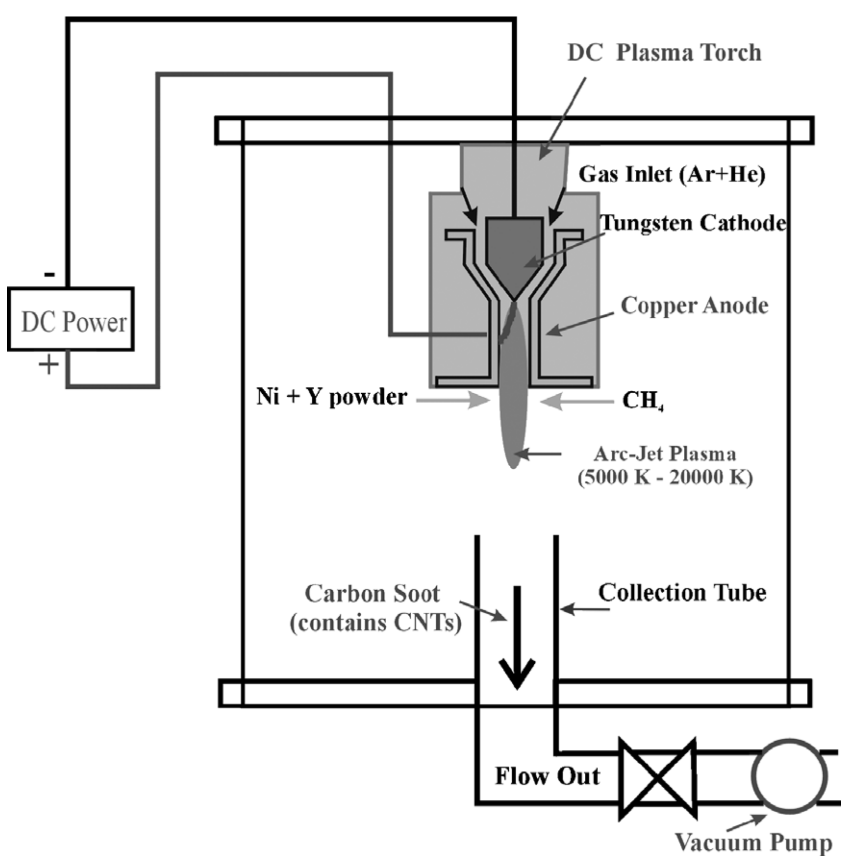

Fig. 1. Schematic diagram of an arc-jet plasma reactor for CNTs synthesis by decomposition of CH_4.