Influence of Metal Catalysts on Synthesis of Low-Diameter Single-Walled Carbon Nanotubes by DC Thermal Plasma Process

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Abstract: Single-walled carbon nanotubes (SWCNTs) synthesis using DC thermal plasma process with CH_4 and H_2 gases mixture and various metal catalysts. Measuring the average diameter of synthesized SWCNTs to be about 1 nm. The nano-sized metal catalyst has the smallest average particle size at 5.2 nm. Nickel and iron contributed as effective catalysts in the synthesis of SWCNTs.

1. Introduction

Despite a promising outlook, conventional SWCNT synthesis methods face significant challenges in meeting industrial demand, including high production costs, elevated defect levels, purity concerns, low yields, and difficulties in controlling nanotube diameter [1,2]. These obstacles have hindered the widespread adoption of SWCNTs across various industries, to overcome these challenges, researchers are exploring alternative synthesis methods, such as the thermal plasma process for SWCNT production [3]. This approach offers several advantages, including effective carbon source decomposition in high-temperature environments (up to 20,000 K), rapid quenching rates (10^5−10^6 K/s) facilitating nanoscale material synthesis, and potential for mass production due to high reaction rates [1,3]. This study was performed to synthesize SWCNTs by varying the selection of catalysts and feeding rates in DC thermal plasma process. In the synthesis of carbon nanotubes (CNTs), transition metals play a crucial role by increasing the activation energy, thereby facilitating the growth of CNTs. The thermal plasma process does not require a substrate, as the CNTs grow directly on the surface of the catalyst particles. This catalyst-assisted growth mechanism in the gas phase is a distinctive feature of the thermal plasma process for SWCNT synthesis.

2. Methods

The process using methane (CH_4) and hydrogen (H_2) mixture gases were used as reactive gases. The various micro-sized metal catalyst powders (Ni, Co & Fe mixture, Ni & Fe mixture) were injected with nitrogen carrier gas. The numerical analysis was performed to calculate the thermal plasma jet using magnetohydrodynamic and computational fluid dynamics codes. Transmission electron microscopy (TEM) was mostly used to analyze the shape and diameter of synthesized SWCNTs, measuring the average diameter of observed tubes.

3. Results and discussion

Most synthesized nanomaterials were collected at the reactor walls. The heat insulation was inserted at the upper part of reactor. TEM analysis confirmed SWCNT synthesized image, with an average diameter of around 1 nm. The highest number of observable SWCNTs when nickel & iron mixed catalyst was used. At this point, the size of the nano-sized metal catalyst was the smallest (average particle size: 5.2 nm), which was discussed as a favorable condition for the synthesis of low-diameter SWCNTs. The numerical analysis revealed that injecting reaction gas into the upper reactor acted as a cooling gas, reducing the high-temperature region of the plasma jet. It increased flow rate and caused plasma acceleration. Injection into lower reactor resulted in a longer residence time in the SWCNT growth temperature range (1,000−1,300 K).

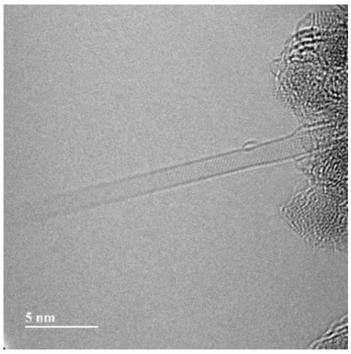

Fig. 1. HR-TEM images of SWCNT synthesized by DC thermal plasma. It was synthesized using a nickel & iron mixed catalyst.

4. Summary

SWCNT was synthesized by DC thermal plasma. The growth region of SWCNTs changed depending on the injection location of the reactive gases. SWCNTs with diameters around 1 nm were successfully synthesized. The average size of the nano-sized catalyst particles was the smallest at 5.2 nm. Furthermore, it was discussed that nickel has an activation temperature (900−1,300 K) similar to the growth temperature of SWCNTs, making it favorable for synthesis. In the case of iron, its high carbon solubility was noted as advantageous for the synthesis process. These characteristics nickel and iron contribute to their effectiveness as catalysts in SWCNT production.

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References

[1] F. Daneshver *et al.*, Nanoscale Adv., **3**, 942−962 (2021).
[2] D. Yang *et al.*, Nat. Commun., **14**, 2491 (2023).
[3] R. Rao *et al.*, ACS Nano, **12**, 11756−11784 (2018).