## Formation of C<sub>60</sub>- Encapsulated Double-Walled Carbon Nanotubes with Novel Electrical Transport Properties Based on Plasma Technology

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The encapsulation of  $C_{60}$  molecules inside double-walled carbon nanotubes (DWNTs) is realized by means of a plasma ion-irradiation method. Transmission electron microscopy observations confirm that DWNTs have been filled with  $C_{60}$  fullerenes having amorphous morphology. More importantly, electrical transport measurements indicate that  $C_{60}$ -encapsulated metallic DWNTs exhibit a novel negative differential resistance behavior.

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Single-walled carbon nanotubes (SWNTs) and double-walled carbon nanotubes (DWNTs) can be doped or intercalated with electron donors or acceptors, and the resulting materials show many interesting properties, including enhanced electrical conductivity. The controlled doping of the carbon nanotubes has been performed in a number of ways. Normally, the doping reactions have been carried out in a vapor or liquid phase. In contrast, plasma technology can provide another interesting way to modify or control the electronic properties of carbon nanotubes. Over the past few years, we have developed a different-polarity plasma ion technology which can be used to insert various alkali-metals (Li, Na, K, and Cs) or fullerene molecules into SWNTs [1-3]. Our recent results demonstrate that air-stable SWNTs with n-type conductivity due to the charge transfer effect can be created by Cs-encapsulation [4]. In addition, the growth of individual SWNTs with controlled structures has been realized by means of a reactive plasma method described in our previous work [5], suggesting that SWNTs with different electronic properties can be synthesized selectively by plasma technology. In this rapid communication, we demonstrate the formation of C<sub>60</sub>-filled DWNTs (C<sub>60</sub>@DWNTs) using a plasma ion-irradiation method. Electrical transport measurements indicate that metallic C<sub>60</sub>@DWNTs exhibit a novel negative differential resistance (NDR) behavior available for nanoelectronics devices.

DWNTs with inner diameters of approximately  $4 \sim 5$  nm were prepared using an arc discharge method employing Fe as a catalyst. DWNTs were dispersed by means of a supersonic treatment in ethanol. Droplets of this suspension were then dripped on a stainless steel substrate

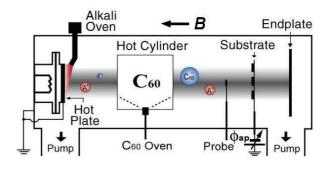


Fig. 1 Schematic of experimental apparatus for plasma-ion irradiation.

(15×15 mm). This DWNT-substrate was installed in a magnetized alkali (metals)-fullerene plasma column (B = 0.2 T), as schematically shown in Fig. 1 (details regarding the production of the alkali-fullerene plasmas used here are described elsewhere [3]). During the experimental process, DWNTs are filled with C<sub>60</sub> molecules by applying a positive bias of 20 V. The sample is characterized primarily through the use of field emission transmission electron microscopy (FE-TEM, Hitachi HF-2000) operated at 200 kV. Figure 2 shows typical TEM images for an empty DWNT (a) and DWNTs filled with numerous C<sub>60</sub> molecules (b). Owing to the large inner diameter of DWNTs, only the amorphous phase of C<sub>60</sub> molecules inside the DWNTs is observed.

The electrical transport properties of  $C_{60}$ @DWNTs are investigated by fabricating them as channels of field-

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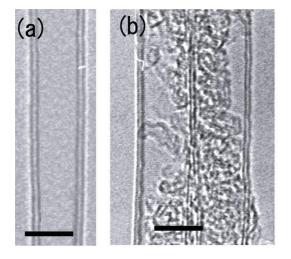


Fig. 2 TEM images of DWNTs: (a) empty DWNT and (b)  $C_{60}$ -filled DWNTs (scale bar: 5 nm).

effect transistor (FET) devices (the fabrication process of these devices is described in [6]). Figure 3 presents a current versus source-drain voltage ( $I_{\rm DS} - V_{\rm DS}$ ) curve obtained from a metallic C<sub>60</sub>@DWNTs transistor measured at room temperature. Interestingly, the measured curve (at a gate voltage of  $V_{\rm G} = 0$  V) exhibits unique NDR characteristics at high bias voltages of about 4.5 V and -5.5 V; namely, when the voltage is progressively increased, the current's initial rise is followed by a sharp decrease instead of the linear increase based on Ohm's law, indicating a typical feature of resonance tunneling electronic devices.

In conclusion, DWNTs have been successfully filled

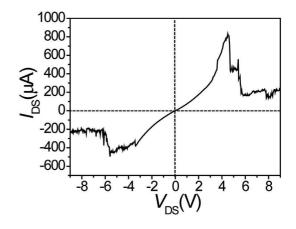


Fig. 3 A  $I_{DS} - V_{DS}$  curve obtained from a C<sub>60</sub>-filled DWNT transistor measured at room temperature, demonstrating that the device exhibits NDR behavior.

with  $C_{60}$  molecules using a plasma irradiation method. The electrical transport properties of metallic  $C_{60}$  @ DWNTs show a novel NDR behavior, suggesting that new functional DWNTs can be created by means of plasma technology.

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