博 士 学 位 論 文 の 要 約

Efficient separation of large diameter semiconducting single-wall carbon nanotubes and its application to thin-film transistors

（半導体特性を持つ太い単層カーボンナノチューブの分離精製とその薄膜トランジスタへの応用）

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Single-wall carbon nanotubes (SWCNTs) are promising materials for electronic device applications owing to their inherently excellent properties, i.e., high carrier mobility, flexibility and solution processability. However conventionally synthesized SWCNTs has broad variety of structures which greatly affect their properties, thus affecting the device performances. For example, SWCNTs can have either metallic (m-) or semiconducting (s-) properties based on their chirality, which is seen as their rolling pattern. The presence of m-SWCNTs in semiconducting devices therefore causes shortcuts which renders the device unusable. Meanwhile, diameter and length of SWCNTs affect their carrier mobility and conductivity which increase by increasing diameter or length. Purification of large diameter, long, and high purity s-SWCNTs are therefore necessary to achieve the high performances of SWCNTs.

High resolution single-stage gel filtration

Gel filtration is among the most sophisticated purification method of high purity s-SWCNTs. In gel filtration, s-SWCNTs are adsorbed on a gel column while most m-SWCNTs pass through. Surfactant solution (i.e., sodium cholate/SC) is then used to obtain the adsorbed s-SWCNTs. However, a small amount of m-SWCNTs often remain, reducing overall purity of the s-SWCNTs.

Here, a controlled low-gradient elution (CLGE) system as shown in Fig. 1 is developed. The CLGE provides gentle change of surfactant composition used to obtain adsorbed SWCNTs, which selectively desorbs the remaining m-SWCNTs ahead of the s-SWCNTs. Hence, high purity s-SWCNTs is obtained in a single-stage gel filtration utilizing CLGE, as opposed to a recycling process (multi-stage) in conventional method. In addition, the CLGE also allows fractioned collection of s-SWCNTs with various diameter.

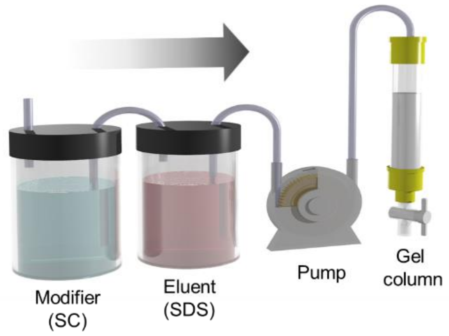

Figure 1 Schematic of CLGE setup where two surfactant solutions: SC and sodium dodecyl sulfate (SDS) are used as eluent. The grey arrow indicates flow direction.

Purification of large diameter semiconducting carbon nanotubes

Large diameter s-SWCNTs are intriguing materials for electronic applications for their higher carrier mobility than the small diameter ones. However, their strong interaction with surfactant lowers their purification selectivity and thus high purity s-SWCNTs obtained so far are limited to those with diameter less than 1.5 nm. Here, we developed a novel method to separate high purity large diameter s-SWCNTs by using combination of temperature-controlled gel filtration and CLGE method. By increasing the column temperature, large diameter s-SWCNTs are adsorbed on the gel due to entropy-driven weakening of interaction between surfactant and large diameter s-SWCNTs. Subsequently, the CLGE selectively collect the large diameter s-SWCNTs from the rest of the adsorbed SWCNTs. As shown in Fig. 2, s-SWCNTs with average diameter of 1.9 nm is obtained by the combinatorial method. Optical absorption and Raman measurements confirm the high purity s-SWCNTs contents of the 1.9-nm-diameter sample. Thin film transistors (TFTs) have been fabricated from the 1.9-nm-diameter s-SWCNTs, which exhibit superior carrier mobility (ca. 1.6 times) of those fabricated from 1.5-nm-diameter s-SWCNTs.

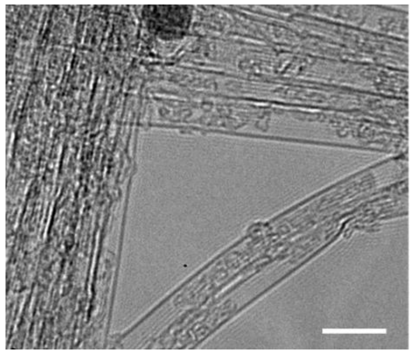

Figure 2 TEM image of the s-SWCNTs purified by using CLGE in a temperature-controlled gel filtration. The scale bar is 5 nm.