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主　論　文　の　要　旨

論文題目　Diameter-Controlled Chemical Vapor Deposition Growth of Single-Wall Carbon Nanotubes using the Mist Flow Method
（直径のコントロール可能の単層カーボンナノチューブを合成する霧化流動化学気相成長法）

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論　文　内　容　の　要　旨

A novel floating catalyst CVD synthesis of SWCNTs using a continuous flow of catalyst-ethanol mist to synthesize SWCNTs has been developed with the removal of the influence of the substrate. One kind of mist generator that can generate fine ferrocene-ethanol droplet with average diameter 3 μm has been used. In the case of selecting the ferrocene as the catalyst, the influence of various parameters (CVD reaction temperature, total flow rate and concentration of ferrocene in ethanol) on controlling the mean diameter and diameter distribution of as-grown SWCNTs have been investigated, which has shown that increasing CVD reaction temperature leads to the formation of larger catalyst particles because of the higher aggregation rate, therefore forming larger diameter of SWCNTs and the flow rate can change either the stability of the gas flow or carbon supply kinetics in the reactor, which has played a significant role in controlling the diameter of as-grown SWCNTs. In the end, through digging out the growth mechanism with the help of HRTEM images of as-grown SWCNTs, there is no direct relationship between the size of iron particles and diameter of as-grown SWCNTs with mist flow CVD. In addition, under the different combination of various parameters, the mean diameter of SWCNTs can distribute from 0.9 to 1.5 nm controllably to satisfy the demand of various applications. And this can prove that the mist flow method is feasible and efficient for the controllable growth of SWCNTs.

Subsequently, one-step catalyst-free mist flow CVD growth of SWCNTs with C_{60} fullerenes has been carried out. No need to do any pretreatments prior to CVD to open the cap-structures, just to synthesize SWCNTs directly (one-step) from C_{60} fullerenes with mist flow CVD can not only simplify the experiment but also avoid the loss of numerous opened C_{60} caps in the air because it seems that C_{60} fullerenes are considered as “the seed” during the formation of SWCNTs; cap opening, nucleation and growing SWCNTs could occur simultaneously in the CVD chamber. Due to

the extremely low solubility of C_{60} in ethanol, an improvement for fabricating a kind of aqueous colloidal C_{60} solution has also been developed. Using modified C_{60} solution mixed with ethanol, CVD occurred under 950℃ and total flow rate of 300 sccm choosing Ar/H_2 (3%) as the carrier gas. As a result, the diameter distribution of as-grown SWCNTs is narrow (from 1.0 to 1.5 nm) and the mean diameter is 1.28 nm, which is rather close to the diameter of an individual C_{60} molecule. Moreover, the water looks like playing a significant role in synthesizing and even controlling the diameter of as-grown SWCNTs in case of the different amount, which would have pointed out an inspirational direction for the one-step controllable growth of SWCNTs in a floating CVD system in the future.

Meanwhile, the third kind of catalyst precursor – metal cluster (Fe_{18}) is also in process. According to the different carbon source (such as MeCN and methanol), different walls of CNTs (single-wall, double-wall and multi-wall) can be synthesized. In addition, choosing the MeCN as the carbon source can lead to the growth of thin diameter SWCNTs and mean diameter of as-grown SWCNTs (0.81 nm) considerably closed to the size of individual Fe_{18} cluster (0.65 nm). In addition, through the comparison with the catalysts-supported CVD (CCVD) under the same reaction condition, the as-grown products synthesized by CCVD consist of MWCNTs and SWCNTs because of the serious aggregation during the pretreatment; whereas the as-grown products synthesized by mist flow CVD consist of completely SWCNTs due to the short residence time which is able to weaken the aggregation of Fe_{18} clusters in the CVD chamber. Anyway, the mist generator can be applied for various kinds of liquid precursors. The wide applicability of the present method, that can generate various fine mists, can be an advantage for realizing selective growth of SWCNTs in the future.