ONE-STEP PREPARATION OF HIGHLY DISPERSED METAL-SUPPORTED CATALYSTS BY FLUIDIZED-BED MOCVD FOR CARBON NANOTUBE SYNTHESIS

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Catalysts play a pivotal role in modern industries, especially in the petrochemical industry, such as in the processes for production of advanced materials such as carbon nanotubes. Since the discovery by Iijima in 1991, carbon nanotube has received increasing interest worldwide owing to their unique structural, electronic and mechanical properties [1], and their promising applications in molecular electronics [2] and hydrogen storage [3], etc. Chemical vapor deposition (CVD) of hydrocarbons proves to be an efficient method to synthesize single-walled and multi-walled carbon nanotubes [4].

In this study, a novel technique, fluidized-bed metal-organic chemical vapor deposition (FB-MOCVD), has been developed as a one-step method to prepare highly dispersed metal-supported catalysts for carbon nanotube synthesis. Using ultrafine powder of gamma-alumina (~100 nm in mean diameter) as the support and iron-pentacarbonyl ($Fe(CO)_5$) and molybdenum hexacarbonyl ($Mo(CO)_6$) as the metal precursors, Fe/Al_2O_3, Mo/Al_2O_3 and $Fe\text{-}Mo/Al_2O_3$ catalysts have been synthesized in a fluidized bed, as shown in Figure 1. Compared with the conventional methods of catalyst synthesis using solutions, such as impregnation, ion-exchange or co-crystallization, the one-step FB-MOCVD technique is advantageous in many aspects such as eliminating the solid-liquid separation and the subsequent operations of drying and high-temperature calcination, therefore minimizing the agglomeration and grain-size growing problems [5]. The metal-supported catalysts obtained by FB-MOCVD were characterized with inductively coupled plasma atomic absorption spectroscopy (ICP-AES), scanning electron microscopy-energy dispersive X-ray (SEM-EDX), X-ray diffraction (XRD), and nitrogen isothermal adsorption, etc. Figure 2 illustrates the results of EDX, ICP-AES and X-ray mapping for the resulting Fe/Al_2O_3 catalyst from the FB-MOCVD process at various temperature between 200 and 600^{o}C. The amount of metal deposition can be controlled by temperature. The activities of the catalysts have been tested for carbon nanotube synthesis by fluidized-bed CVD of 10% acetylene (C_2H_2) in high purity H_2 at 650^{o}C. The resulting carbon nanotubes were characterized with SEM and thermogravimetric analysis (TGA), and Figure 3 shows the SEM image of the carbon deposits on the Fe/Al_2O_3 catalyst. The fluidized bed CVD method has a great potential for large-scale production of carbon nanotubes.

References:

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Figures:

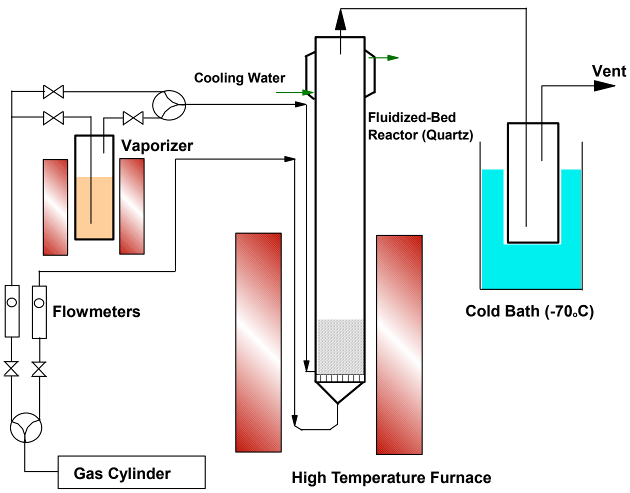

Figure 1. The fluidized-bed MOCVD setup

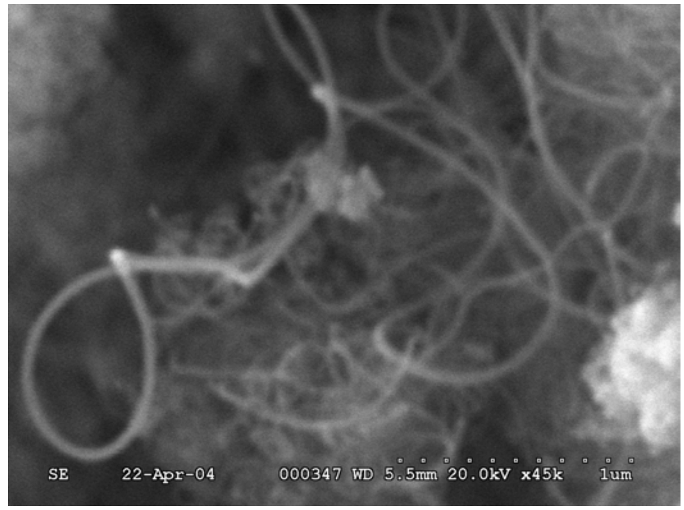

Figure 3. SEM image of carbon nanatubes synthesized by FB-CVD of C_2H_2 at 650°C with Fe/Al_2O_3 catalyst

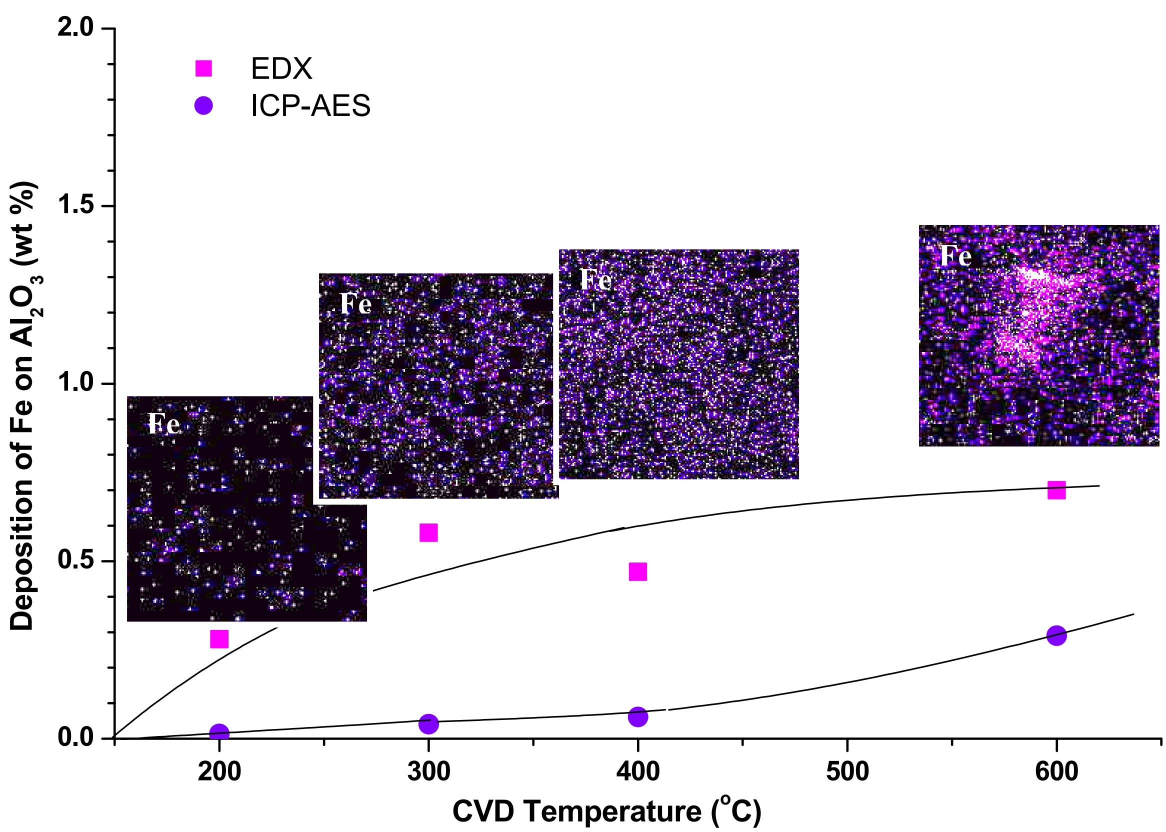

Figure 2. Deposition of metal (Fe) on gamma-Al_2O_3 particles by the FB-MOCVD process