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Effect of Mo in Co-Mo/MgO catalysts on the synthesis yield and structure of carbon nanotubes

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Abstract

Carbon nanotubes were synthesized at 600-800˚C by a typical CVD method using Co/MgO and Co-Mo/MgO, and ethylene as feedstock. The effects of the composition of the catalyst, synthesis temperature on the yield and structure of carbon nanotubes were investigated. The experimental results showed that small amount of Mo addition in Co could improve the yield of carbon nanotubes by 8 times. The carbon nanotubes synthesized using Co-Mo/MgO showed broader diameter distribution and “cup-type” structure.

Introduction

Carbon nanotubes are expected to have a large application in composite materials, batteries and many other fields because they have excellent properties like high aspect ratio, high mechanical strength and electric conductivity, etc. The technology to fabricate CNTs with homogeneous structure and size in a low cost is still a big challenge for researchers. Chemical vapor deposition (CVD) in the presence of catalysts such as Co, Ni and Fe is a promising method for large-scale synthesis of CNTs. The yield and structure of CNTs depend on many parameters, especially on temperature, catalyst composition and catalyst particle size [1,2].

In this study, we investigated the effect of Mo addition in Co/MgO catalyst on the yield and structure of CNTs.

Experimental

The catalysts were obtained by mixing $Co(Ac)_2$, $Mg(Ac)_2$ and $(NH_3)_6Mo_7O_{24}$ and calcined at 700ºC for 5h. Syntheses of CNTs were performed at 600ºC, 700ºC and 800ºC using C_2H_4 as carbon source for 30-120 min. The yield of CNTs was calculated according to (amount of products-amount of catalyst)/(amount of CoO and MoO_3 in catalyst). The morphology and structure of CNTs were examined using FESEM and TEM. The catalysts before and after reduction were also examined using XRD.

Results and Discussions

By using Co/MgO as catalyst, synthesis at 700ºC showed much higher carbon nanotube yield then those at 600ºC and 800ºC. It is found that addition of Mo in the Co-Mo/MgO catalyst improved the carbon nanotube yields significantly. For Co/MgO catalyst, the yield of carbon nanotubes was 426% for 30 min synthesis, and 497% for 120 min. It indicates that most of the catalyst particles deactivated after 30 min. On the other hand, the carbon yield for Co-Mo/MgO was 1269% for 30 min, and 4582% for 120 min, suggesting the catalyst particles still maintained the activity to grow carbon nanotubes.

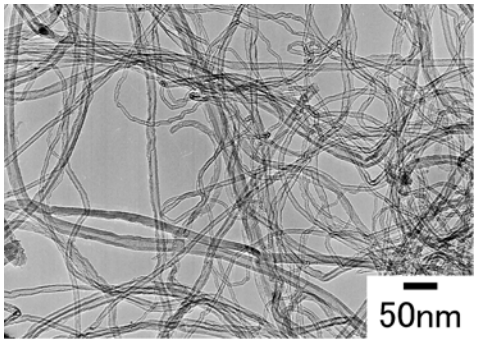

Fig. 1. TEM image of as-grown CNT synthesized at 700ºC from Co/MgO.

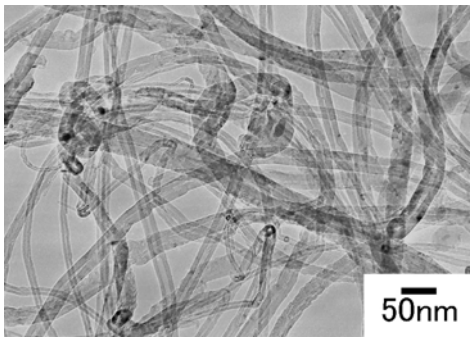

Fig. 2. TEM image of as-grown CNT synthesized at 700ºC from Co-Mo/MgO.

Fig.1 and 2 showed the morphology of CNTs. The carbon nanotubes from Co/MgO showed diameter ranging from 8 to 29 nm with an average of 12.7 nm, while the CNTs from Co-Mo/MgO showed diameter ranging from 6 to 47 nm with an average of 17.2 nm. Observation of HRTEM showed that CNTs from Co/MgO showed a typical MWNT structure, but the CNTs from Co-Mo/MgO showed a cup-stacked structure.

The above results suggested that Mo addition in the catalyst increased the catalyst size distribution and made the catalyst activity last longer. XRD analysis of catalyst showed existence of Co_3O_4, MoO_3 and CoO-MgO solid solution in Co-Mo/MgO catalyst before reduction, which was one reason for the broad distribution of catalyst size. It is thought that formation of MoC_2 in catalyst during growth of CNTs played a role in the stable growth of CNTs.

Conclusions

Mo addition in Co/MgO improved significantly the carbon yield due to longer activity of catalyst, but the diameter distribution became broader. It is still necessary to control the catalyst size distribution.

References

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