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Synthesis of carbon nanotubes by CVD using Fe /MgO and Fe-Mo/MgO system as catalysts

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Abstract – The addition of Mo in a chemical vapor deposition (CVD) synthesis of carbon nanotubes (CNTs) is known to have important effects in the characteristics and yield of these structures. CNTs have been synthesized by CVD using Fe/MgO and Fe-Mo/MgO systems. The Mo plays a role in the distribution of iron phases. Due to the strong interaction between the iron and the MgO, most of iron in Fe/MgO is not available for CNT growth. In Fe-Mo/MgO system, the Mo prevents the dissolution of iron, by forming Fe–Mo oxides, which leads to the formation of high concentration of active nanoparticles.

Among the various synthesis methods, the chemical vapor deposition (CVD) of hydrocarbons or carbon monoxide over ceramic-supported nanoparticles of transition metals such as Fe, Co, Ni has been considered the most promising method for the large-scale production of carbon nanotubes [1]. The addition of Mo to transition metals is known to have very important effects in the characteristics (number of walls, diameter distribution, presence or not of non-tubular nanocarbons) and yield production of carbon nanotubes [2]. Despite its importance, the mechanism by which Mo favors the formation of carbon nanostructures is not yet totally clarified [2,3]. In this work, carbon nanotubes have been synthesized by catalytic chemical vapor deposition of ethylene (C_2H_2) over MgO-supported Fe catalyst at 900 ºC with and without the addition of Mo, and the influence of Mo as an activator in this system has been studied in details.

The catalysts systems were produced by coprecipitation technique. A stoichiometric mixture of magnesium nitrate ($Mg(NO_3)_2.6H_2O$) and iron nitrate ($Fe(NO_3)_3.9H_2O$) were dissolved in deionized water. The mixture was heated to the boiling and pure ammonium hydroxide solution was added drop by drop. For Mo-containing catalysts, stoichiometric amount of molybdenum (II) acetate dimer ($[Mo(OCOCH_3)_2]_2$) was also added. The precipitates were collected by filtration, washed with distilled water several times and then dried overnight in a vacuum at 110ºC. Finally, the obtained dried powders were crushed and calcined at 500ºC for 1 h. The calcined powders were also annealed in the CVD reactor under argon atmosphere from room temperature to 900ºC to simulate the conditions of the materials just before starting CNT growing.

For CVD growth of carbon nanotubes, the catalyst (~100 mg) was put into an alumina boat inside a 2" quartz tube and heated to the reaction temperature (900°C) under Ar flow (1000 sccm). After temperature stabilization, 35 sccm of ethylene (C_2H_4) was added and the Ar flow was increased to 2000 sccm, resulting in a flow ratio ($[C_2H_4]/[Ar]$) of 1,75%. After the CNT had been allowed to grow for 25 min, the ethylene gas was turned off and the furnace was cooled to room temperature under an Ar atmosphere.

The analysis of the different catalysts and carbon samples shows that the Mo plays an important role in the distribution of the iron ions and iron phases in the bimetallic catalyst. Due to the strong interaction between the iron ions and the MgO matrix, most of iron in Fe/MgO system is not available for CNT growth. On the other hand, in Fe-Mo/MgO system, the Mo prevents the dissolution of iron in the MgO lattice by forming Fe–Mo oxides, which leads to the formation of high concentration of active nanoparticles for CNT growth. In the bimetallic catalyst, however, large particles are also produced resulting in the growth of other carbon species than carbon nanotubes. The phase composition of the catalysts and carbons was studied using different techniques (synchrotron X-ray diffraction, Mössbauer spectroscopy, SEM, TEM and EDS).

References

[1] J. Liu. S. Fan and H. Dai, MRS Bulletin, (2004) 244 - 250.
[2] M. Pérez-Mendoza, C. Vallés, W. K. Maser, M. T. Martínez, A. M. Benito, Nanotechnology, 16, (2005) 224 - 229.
[3] T. Saito, W. -C. Xu, S. Ohshima, H. Ago, M. Yumura and S. Iijima, J. Phys. Chem. B, 110 (2006) 5849.