11th International Conference
on Advanced Materials

ICAM2009

Rio de Janeiro Brazil
September 20 - 25

Carbon nanotubes synthesis on FeMoMgO catalysts monitored by a TPRe system

B. R. S. Lemos,[(1)*] A. P. C. Teixeira,[(2)] L. A. Magalhães,[(2)] R. M. Lago,[(1)] C. A. Furtado,[(2)] A. P. Santos[(2)]

(1) DC, Universidade Federal de Minas Gerais, MG, Brazil, brunoroch@ufmg.br
(2) Centro de Desenvolvimento da Tecnologia Nuclear, MG, Brazil

Abstract – In this work, TPRe (Temperature Programmed Reaction) has been used to monitorated carbon nanotube production during CVD (Chemical Vapor Deposition). The experiments were carried out using four different catalysts based on Fe and FeMo supported on MgO via impregnation (imp) or co-precipitation (ppt). These systems were characterizated by SEM (Scanning Electronic Microscopy) and Raman Spectroscopy. The (ppt) catalysts produced more reactive Fe species which produce carbon at lower temperatures favoring more defective forms of carbon. On the other hand, the (imp) catalysts are very active in a narrow temperature range (760-790°C) producing more organized carbon, e.g. SWNT.

Nowadays, a lot of catalysts and carbon source have been studied for production of carbon nanotubes by CVD (Chemical Vapor Deposition). However, the process kinetics isn't completely understood[1].In this work, preliminary results on the use of a TPRe (Temperature Programmed Reaction) system, a well known catalysis technique, showed TPRe as a powerful tool to monitor carbon nanotube production during CVD. The experiments were carried out using four different catalysts based on Fe and FeMo supported on MgO via impregnation (imp) or co-precipitation (ppt).

In a first step, the catalyst was reduced with H_2 and the hydrogen consumption was monitored by a normal TPR (Temperature Programmed Reduction) procedure. After reduction the gas stream was changed to CH_4/Ar and the sample heated up to 900°C where the temperature was kept for 30 min. SEM analyses of the samples after reaction showed the formation of large amounts of carbon nanotubes and filaments. Raman spectra (laser 514 nm) showed important differences for the catalysts prepared, i.e. only FeMgO(imp) and FeMoMgO (imp) showed RBM modes with high I_G/I_D ration. Figure 1 shows the production of H_2 during TPRe experiments. The H_2 production is directly related to the carbon formation:

CH_4 (catalyst)→ C + 2 H_2 (Equation 1)

From the TPRe profiles several important information can be obtained, such as the reactivity of the catalyst, the carbon yield, the kinetics of carbon formation and catalyst deactivation[2].

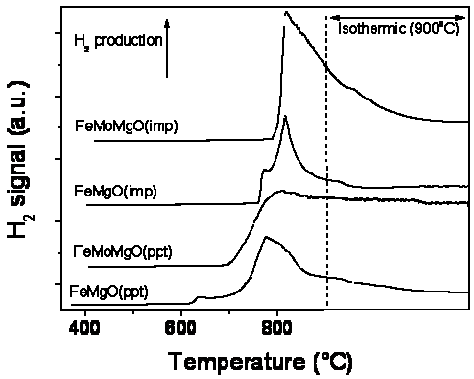

Figure 1. TPRe profiles for the reaction of CH_4 with Fe/MgO(imp), FeMo/MgO(imp), Fe/MgO(ppt) FeMo/MgO(ppt).

It can be observed that the catalysts (ppt) and (imp) showed significantly different TPRe profiles. FeMgO(ppt) and FeMoMgO(ppt) showed more reactive species producing carbon at lower temperatures, i.e. 600-680 compared to 760-790°C for the (imp). This reaction at lower temperature for (ppt) is likely related to the formation of more defective/amorphous carbon. The increase on the H_2 signal affords information on the C formation kinetics. The catalysts (ppt) showed a gradual increase in the H_2 signal suggesting a slower carbon formation. On the other hand, the catalysts (imp) produced a sharp increase in the H_2 signal suggesting a rapid carbon formation which could be related to the formation of SWNT as revealed by Raman. The larger peak areas (Figure 1) suggest that the presence of Mo in the catalyst leads to much higher carbon yields. In conclusion, the (ppt) catalysts produced more reactive Fe species which produce carbon at lower temperatures favoring more defective forms of carbon. On the other hand, the (imp) catalysts are very active in a narrow temperature range (760-790°C) producing more organized carbon, e.g. SWNT.

References

[1] Y.A. Kasumov, A. Shailos, I.I. Khodos, V.T. Volkov, V.I. Levashov and V.N. Matveev. Journal Appl Phys A, 4 (2007) 687-91.
[2] S. Besselmann, C. Freitag, O. Hinrichsen and M. Muhler. Physical Chemistry Chem. Phys, 3 (2001) 4633.