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Nongyue Hea [b], Yueping Kuang [c], Qing Dai [a], Ping Li [a], Yuqing Miao [a], Zuhong Lu [a] & Chunwei Yuan [a]

[a] National Laboratory of Molecular & Biomolecular Electronics, Southeast University, Nanjing, 210096, P. R. China

[b] Department of Chemistry, Xiangtan University, Xiangtan, 411105, P. R. China

[c] Department of Chemistry, Hunan Normal University, Changsha, 410006, P. R. China

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NONGYUE HE[ab], YUEPING KUANG[c], QING DAI[a], PING LI[a], YUQING MIAO[a], ZUHONG LU[a] and CHUNWEI YUAN[a]

[a]*National Laboratory of Molecular & Biomolecular Electronics, Southeast University, Nanjing 210096, P. R. China,* [b]*Department of Chemistry, Xiangtan University, Xiangtan 411105, P. R. China and* [c]*Department of Chemistry, Hunan Normal University, Changsha 410006, P. R. China*

Hexagonal mesoporous molecular sieve materials (HMS) with different pore sizes were applied as supports of Fe-loading catalysts to catalytically synthesize carbon nanotubules. The larger the pore size of HMS is, not only the larger the pore size of the formed carbon nanotubes is, but also the longer the reaction time is. It was observed that the growth of carbon nanotubes can be orientated by the one-dimensional mesoporous structure of HMS.

Keywords: carbon nanotubules; mesoporous molecular sieve materials; pore size; growth direction

INTRODUCTION

Carbon nanotubule materials have wide-ranging implications to a variety of areas[1-8]. However, characterizations and applications of carbon nanotubules have been hampered because of their uncontrolled diameters and random growth directions. Although the growth directions can be controlled by catalytic synthesis of carbon nanotubules in the pores of Fe/silica or Fe/alumna substrate[4,5], the control over the pore size distribution of gel silica is dificult and only rough substrate size selectivity can be anticipated. Thus the support material

with a fixed pore size that precisely controls the pore size and growth direcion of carbon nanotubules would be an advancement. Here we reported another method to tailor the diameters of carbon nanotubules by using mesoporous molecular sieve materials with different pore sizes as catalyst supports.

METHODS AND EXPERIMENTS

Pure siliceous hexagonal mesoporous molecular sieve materials, called HMS, and microporous zeolite KL(Si/Al=2.9) were synthesized as described previously[6,7]. NaY zeolite with an atomic Si/Al ratio of 2.49 and unit cell parameter of 2.468 nm was commercially available.

Fe/HMS was prepared by depositing $Fe(Ac)_2$ on HMS as reported[7]. Fe/NaY, Fe/KL were also prepared in the similar procedure. All samples were loaded 5.0% of Fe in weight. The carbon nanotubules were formed by catalytically depositing acetylene following the reported procedure[8]. The quality of the carbon deposit on the catalysts was investigated by transmission electron microscopy (TEM) (JEOL 200 cx).

RESULTS AND DISCUSSION

The TEM pictures illustrated (not shown here) the hexagonally arranged patterns of pores for HMS(12) and HMS(18). The XRD patterns (not shown here) for both samples also matched that reported[6,9]. After the deposition of $Fe(Ac)_2$, the mesoporous structures of both samples remained.

From Table 1, we can find carbon nanotubules can formed on all samples except Fe/KL. The formation time of carbon nanotubules on Fe/NaY is much shorter than on Fe/HMS samples. Furthermore, the reaction times on Fe/HMS(12) is shorter than that on Fe/HMS(18), indicating the structure and pore size of supports greatly influences the formation of carbon nanotubules.

TABLE I Textural properties of used catalysts and data for carbon nanotubules formation

Catalysts	Pore size (nm)	S.A.[a] (m^2/g)	Favorable reaction time (min)	Pore size of carbon nanotube (nm)
Fe/NaY	~0.7[a], ~1.36[b]	697	10	~10
Fe/KL	~0.7[a]	287	--	Few carbon tubules
Fe/HMS(12)[c]	2.9	1031	30	~13
Fe/HMS(18)	3.8	1029	60	~17

[a]Diameter of window. [b]Diameter of supercage measured by ^{129}Xe MAS NMR. [c]Numbers in parentheses are carbon numbers in the chain of primary amine templates.

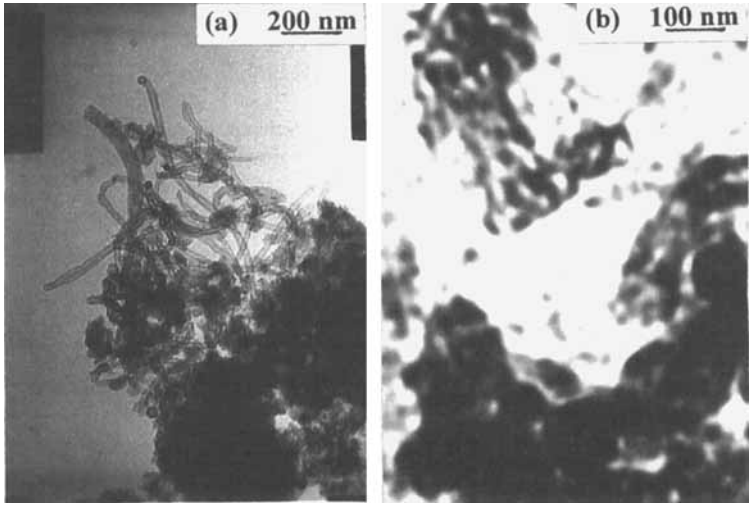

FIGURE 1 Carbon nanotubules grown (a) on the edges and (b) in the central part of an Fe/HMS(12) particles

One very interesting phenomenon is that the pore size formed on Fe/HMS

samples increased with the pore size of HMS supports (see Table 1), suggesting the diameter of carbon nanotubules can be tailored by adjusting the pore sizes of mesoporous materials. Another attractive result is that although the direction of carbon nanotubules formed on the edges of all samples are random (see Figure 1(a), take Fe/HMS(12) for example), nanotubules grown almost in same direction were found in the central part of the catalyst grains of Fe/HMS(12) and Fe/HMS(18). Shown in Figure 1(b) are the carbon nanotubules grown in the central part of an Fe/SiHMS(12) particle. Many black rings or white spots appear in this picture. We believe that they are the pores of carbon nanotubules grown vertically in a same direction from the catalyst. No such phenomenon was observed for Fe/NaY and Fe/KL. It seems that the one-dimensional mesoporous structure of HMS can orientate the growth of carbon nanotubes to a certain degree.

Acknowledgements

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