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A Novel Method for the Synthesis of Mesoporous Molecular Sieve MCM-41

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Abstract: Using glycerol, glycol and water as solvent, cetyltrimethylammonium bromide (CTAB) as template, tetraethyl orthosilicate (TEOS) as silica source, ethylenediamine (EDA) as base source, mesoporous molecular sieve MCM-41 has been synthesized at room temperature, characterized by X-ray power diffraction and N_2 adsorption. Compared with the samples synthesized by glycol and water, the samples synthesized by glycerol have larger pore diameter and high surface areas. Thus glycerol is an efficient solvent for preparing larger pore mesoporous MCM-41.

Keyword: MCM-41, synthesis, glycerol, glycol, weak base, room temperature.

Mesoporous materials are of great interest to many researchers since it possesses highly ordered periodic arrays of uniformly sized channel and large surface area (~1000m^2/g). MCM-41 is a well-known mesoporous molecular sieve first synthesized by Mobil Company researchers[1], it has been investigated in many applications as heterogeneous catalysis, catalyst support, adsorbent[2-4]. Generally, mesoporous molecular sieve MCM-41 was synthesized by hydrothermal method in strong base medium. But this method has some defects such as longer induce time, crystal-transforming phenomenon. Recently, MCM-41 was synthesized in ethylenediamine media at room temperature by Ge Shuxun[5]. Mesoporous molecular sieves synthesized in nonaqueous system are used to prepared thin films, fibers, spheres or monoliths through solvent (*e.g.* ethanol) evaporation and growth of fiber and thin films at interface[6,7]. These films and fibers with highly ordered mesochannels have potential and distinctive advantages in electronic devices[8]. Here we report the successful preparation of mesoporous molecular sieve MCM-41 in glycerol and ethylenediamine at room temperature.

Experimental

A typical synthesis was carried out as follows: the appropriate amount of cetyltrimethylammonium bromide (CTAB), template was dissolved in solvent glycerol, glycol, water, respectively. The pH of this solution was adjusted to 12 with ethylenediamine (EDA), followed by the addition of tetraethyl orthosilicate (TEOS) with vigorous stirring for about 30 min at room temperature. The molar composition of the reaction mixture was

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1 TEOS: 0.7 CTAB: 130 solvent(glycerol or glycol or water): 13 EDA. The mixture was stirred for 8 h at room temperature. Then the mixture was aged at room temperature for 24 h. The as-synthesized product was filtered and washed with deioned water to pH=7, dried at 50℃ for 24 h, then calcined at 550℃ in air for 5 h to remove the organic templates. The X-ray power diffraction (XRD) patterns were obtained with a Rigaku D/max-2500 diffractometer equipped with a rotating anode and Cu-K_α radiation: the diffraction data were collected by using a continuous scan mode with a scan speed of 2°(2θ)/min at room temperature; The Nitrogen adsorption/desorption was performed at 77 K using a Micromeritics ASAP2010 sorptometer. The surface area was determined from nitrogen adsorption isotherm by the Brunauer-Emmett-Teller (BET) method and determination of the pore size distribution from adsorption branch by the Barrett-Joyner-Halenda (BJH) method.

Results and Discussions

Figure 1 and **Table 1** show the XRD pattern of the calcined MCM-41 sample synthesized in different solvents. In every material, showed a sharp XRD peak around 2θ=2° and few weak peaks in 2θ=3~5°, which indicated well-hexagonal structure of MCM-41. The sample A clearly indicated stronger intensity in XRD spectrum, suggesting that the sample A has higher order. Furthermore XRD patterns showed that the 100 peak positions of the three samples (A, B, C **Figure 1**) are different, the sample A has larger d_{100} and a_0.

Figure 2 depicts the nitrogen adsorption-desorption isotherms and pore size distribution of samples (A, B, C), it is observed that the three samples show a type Ⅳ isotherm with a large hysteresis, and capillary condensation of nitrogen occurs at relative pressure p/p_0=0.2~0.4, which is the characteristic of mesoporous materials . The isotherm can be decomposed in three parts: the monolayer-multiple adsorption of N_2 on the wall of the mesopores, the capillary condensation of the nitrogen within the mesopores and then the saturation. From pore diameter distribution we can see that the sample A has larger pore diameter.

Figure 1 XRD pattern for mesoporous molecular sieve MCM-41 obtained of samples

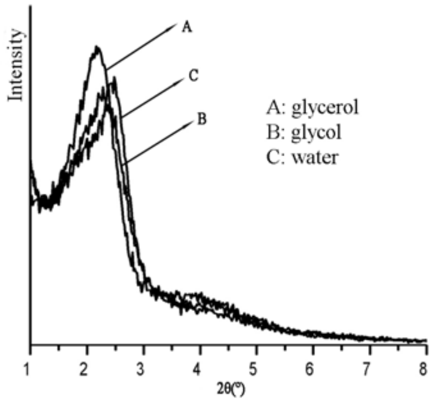
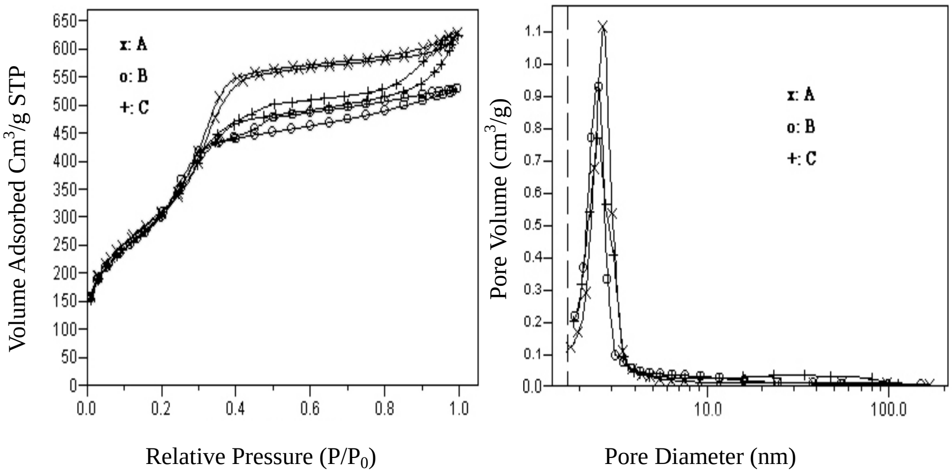


Table 1 The XRD data of mesoporous MCM-41 obtained with different solvents

No.	Solvent	2θ(°)	d_{100}/nm	a_0/nm	Dp(nm)	
					BET	BJH(adsorption)
A	glycerol	2.20	4.13	4.63	3.58	2.87
B	glycol	2.28	3.87	4.47	3.15	2.70
C	water	2.48	3.56	4.11	2.82	2.65

Figure 2 N_2 adsorption-desorption isotherm and pore size distribution MCM-41 of samples



Conclusion

Mesoporous molecular sieve MCM-41 has been synthesized at room temperature using glycerol as non-aqueous solvent. The product has larger pore diameter and narrow pore size distribution.

Acknowledgments

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