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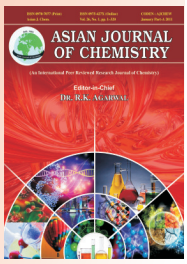

Facile Synthesis of Mesoporous Silica for Separation of Coalbed Methane

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Mesoporous silica as potential adsorbent for separation of coalbed methane was prepared by using one anionic surfactant containing poly(ethylene oxide) group as template in a facile method. Small angle XRD, TEM and N_2 adsorption-desorption isotherms were carried out to confirm the final product. From the results of characterizations, the diameters of the spherical particles have a narrow range of 100-130 nm and the mesoporous sizes are in 2-4 nm. In addition, the average pore size is 2.98 nm, the BET surface area is 572 m^2/g and the pore volume is 0.4150 cm^3/g.

Keywords: Mesoporous silica, Separation, Coalbed methane.

INTRODUCTION

Coalbed methane (CBM) is a form of natural gas extracted from coal beds. In recent years, it has become an important source of energy in China and in other countries. However, unlike much natural gas from conventional reservoirs, coalbed methane contains very little heavier hydrocarbons such as propane or butane and no natural gas condensate. It often contains up to a few per cent carbon dioxide. So separation of coalbed methane is very important. A lot of technology and adsorbent were developed to separate of coalbed methane. Carbon molecular sieve as adsorbent was studied widely, but other materials potential for adsorbent were reported rarely.

Mesoporous materials have drawn great interest from many researchers since mesoporous molecular sieve had been first synthesized by Mobil Oil Research and Development in 1992[1]. Due to its high surface area, uniform pore structure, shape and volume and photocatalytic properties, meoporous silica is widely used as filters, adsorbents[2], catalysts[3] and drug delivery carriers[4,5]., Recently, meosporous materials have been always prepared by using structure directing agent which is also called as template. Surfactants are usually used as template because of their self-assembly in the aqueous solution. However, the pore morphologies, including pore size and shape, are determined by the types of surfactants[6,7] which can be classified into cationic, anionic and nonionic surfactants. Nonionic surfactants become more and more powerful in the syntheses of mesoporous materials due to their wide variety of different chemical structures and attractive characteristics like low price, nontoxicity and biodegradability[8]. The main numbers of such nonionic surfactants are oligomeric alkyl poly(ethylene oxide) (PEO) surfactents, in which the poly-(ethylene oxide) part is hydrophilic and can form hydrogen bonds with water molecules. For example, Zhao's group[9] have synthesized highly ordered mesoporous materials by employing poly(ethylene oxide)-b-poly(propylene oxide)-b-poly(ethylene oxide) (PEO-PPO-PEO) triblock copolymers as templates under acidic aqueous media. Cationic surfactants, especially quaternary cationic surfactants, are also generally efficient for the synthesis of ordered mesoporous silicate materials[10,11]. However, cationic surfactants containing poly(ethylene oxide) parts are not common. Anionic surfactants include sulfates, carboxylates, phosphates, sulfonates are also used to template the synthesis of mesoporous silica[12]. Also poly(ethylene oxide)-contained anionic surfactants used in mesoporous materials have been rarely reported.

In this work, one anionic surfactant containing poly-(ethylene oxide) part, $C_{12}H_{25}(OCH_2CH_2)_2SO_4^-Na^+$, was used as template to prepared spherical mesoporous silica as potential adsorbent to separation of coalbed methane. The final products were confirmed by special characterizations. This investigation may expand the synthesis methods for mesoporous materials and adsorbent to separation of coalbed methane.

EXPERIMENTAL

Sodium alcohol ether sulphate ($C_{12}H_{25}(OCH_2CH_2)_2SO_4^-Na^+$, AESNa, 70 %) was produced by Hunan licheng company. Sodium hydroxide were purchased from Tanjin Chemical Reagents Co. Tetraethyl orthosilicate (TEOS, 98 %) was obtained from Aladdin Chemistry Co. Ltd. All the chemicals used in this work without any further purification. Double distilled water was used to prepare solution and clean the glass instruments.

Synthesis of mesoporous silica: The appropriate amount of sodium alcohol ether sulphate was dissolved in double distilled water, then the mixture was adjusted to pH = 9 by adding sodium hydroxide solution (0.01 mol/L). After stirring 0.5 h at 25 ºC, the TEOS was added dropwise in the mixture. The molar ratio of the mixture was n(TEOS):n(AESNa): n(ascorbic acid):n(H_2O) = 5:1:0.04:600. Then the mixture was stirred for another 8 h. After being filtered, washed with double distilled water and dried at 80 ºC for 8 h, white powder as-synthesis sample was obtained. Finally, the sample was calcined at 550 ºC for 4 h and the final product was the mesoporous silica.

Characterization: The X-ray power diffraction (XRD) were conducted with a Rigaku D/max-2500 diffractometer equipped with a rotating anode and CuK_α radiation, the diffraction data were collected by using a continuous scan mode with a scan speed of 2º (2θ)/min from 1º to 8º.

The morphology and topology images of final product were obtained from transmission electron microscopy, which was carried out with a JEM-1011 electron microscope at 100 kV.

The nitrogen adsorption/desorption was performed at 77.35 K using a Micromeritics ASAP2010 sorptometer. The samples were activated at 110 ºC in vacuum atmosphere about 10 μmHg for 10 h before measurement. The surface area was determined from nitrogen adsorption-desorption isotherm by the Brunauer-Emmett-Teller (BET). The pore size distribution was calculated from desorption branch by the Barrett-Joyner-Halenda (BJH).

RESULTS AND DISCUSSION

In order to obtain the information of pore structure of the final product, small angle XRD was carried out and the result was shown in Fig. 1. Form Fig. 1, only one single apparent peak appears at 2θ = 1.4, which indicates the existence of well-ordered mesoporous in the product[13-15]. In addition, according to Bragg equation (2d sinθ = nλ), the corresponded space d, the thickness of pore-pore wall, is 63.05 Å.

The pore structure and size can be observed intuitively from TEM images. Fig. 2 shows the TEM images of final product in different magnification. It is observed that the morphology of sample presents uniform spherical particles and the diameters of them are in the range of 100-130 nm.

The N_2 adsorption-desorption isotherms and the corresponding pore size distribution plots of final product are shown in Fig. 3. The isotherms can be classified as type IV according to the IUPAC[16]. The typical type IV isotherm with a hysteresis loop can also prove the product possesses meoporous. Besides, the type of the hysteresis loop is H1, which can indicate the pores are approximately uniform in fairly regular array and

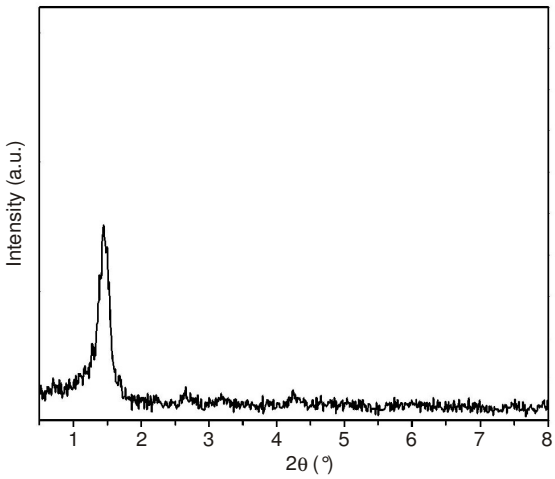

Fig.1. XRD pattern of prepared mesoporous silica

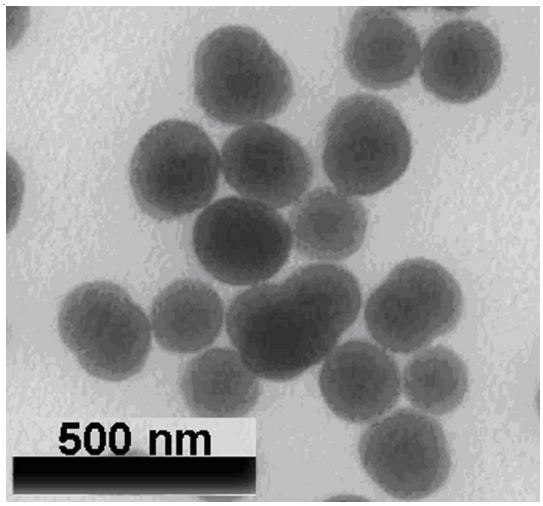

Fig. 2. TEM images of prepared mesoporous silica with different magnification

hence to have narrow distributions of pore size. Meanwhile, the narrow pore size distribution shown in Fig. 3 proves the above state. According to the BJH method, the average pore size is 2.98 nm. In addition, the BET surface area is 572 m^2/g and the pore volume is 0.4150 cm^3/g.

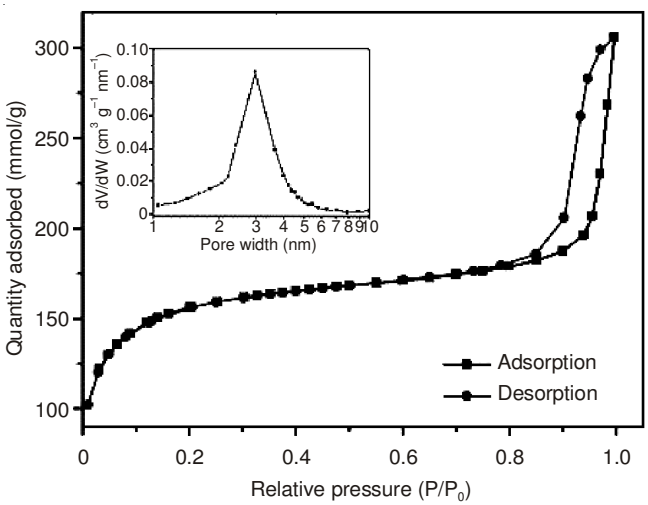

Fig. 3. N_2 absorption-desorption isotherms and pore size distribution (inset) of prepared mesoporous silica

Conclusion

In this work, silica with ordered uniform mesoporous was prepared by using sodium alcohol ether sulphate as template. The final structures of product were confirmed by the combination of small angle XRD, TEM and N_2 adsorption-desorption isotherms. The diameters of the mesoporous particles are in a narrow range of 100-130 nm with the mesoporous sizes in 2-4 nm. In addition, the average pore size is 2.98 nm, the BET surface area is 572 m^2/g and the pore volume is 0.4150 cm^3/g. Further investigations may be conducted to adsorption kinetics of coalbed methane in mesoporous silica and explore their applications.

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