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A simple embedded-seeding method to prepare silicalite-1 membrane on porous α-Al_2O_3 hollow fibers

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ABSTRACT

High quality silicalite-1 membrane was successfully synthesized on the external surface of ceramic hollow fibers by a novel embedded-seeding method. The samples were characterized by XRD, SEM and gas permeation test. It is shown that the obtained silicalite-1 membrane was dense and continuous, with a thickness of about 4–5 μm. Specifically, silicalite-1 membrane exhibited high H_2 and N_2 permeances of 8.3×10^{-6} and 2.4×10^{-6} mol·m^{-2}·s^{-1}·Pa^{-1} with good H_2-N_2 perm-selectivity of 3.45. More finger-like pores in the hollow fibers minimized the gas transport resistance through the support, which is one of the reasons that the prepared supported silicalite-1 membrane is highly permeable.

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1. Introduction

Zeolite membranes have captured particular attention in recent years because of high thermal, chemical and structure stability under harsh conditions. One of the most studied zeolite membranes is MFI, which includes ZSM-5 and silicalite-1 (Sil-1), due to their high sieving properties [1]. Sil-1 membranes were prepared by different methods including in situ crystallization, secondary seeding growth and microwave heating approach [2–6]. In particular, secondary growth method is very attractive and widely used. It has been demonstrated that the support chemistry plays an important role in zeolite membrane synthesis [7,8]. Although pre-seeding can reduce their effects on zeolite deposition and control the membrane orientation and morphology [9], the quality of seeds on the support is still an important issue on the membrane formation. A layer of zeolite particle seeds should be introduced into the surface of support. Therefore, many pre-seeding approaches have been adopted including dip-coating, rub-coating, vacuum-coating and the combination of all these coating methods. Another critical factor is the seed particle size, which is determined by the pore size of support. Furthermore, the seeded support needs to be heated in order to bond the seed particles on the surface because the seeds are often physically and weakly adsorbed on the support. However, these unfavorable factors could be reduced if seed particles are integrated into support. A strong adhesion between the support and the zeolite crystals can be achieved. Compared to flat plates and large tubes which were widely used support, hollow fibers can provide the largest surface area per unit volume. Furthermore, the presence of more finger-like pores could minimize the transport resistance through the support layer.

This paper developed a novel embedded-seeding method to prepare Sil-1 membrane on the asymmetric α-Al_2O_3 hollow fibers. The Sil-1 particles were purposely added into the synthesis solution during the preparation of ceramic hollow fibers. The hollow fibers containing silica species were obtained by phase inversion technique. Then a continuous Sil-1 membrane was directly obtained on the surface of support through the hydrothermal treatment. The samples were characterized by XRD, SEM and gas permeation test.

2. Experimental

Sil-1 particles were prepared from 1TEOS (tetraethyl orthosilicate): 50H_2O: 0.35TPAOH (tetrapropylammonium hydroxide) at 373 K for 16 h according to prior works [7,10]. The Sil-1 particles were separated, dried and added into a mixture containing polyethersulfone, Al_2O_3 powder and N-methyl-2-pyrrolidone. The weight ratio of Sil-1 particles was controlled at 0.02 wt%. The hollow fiber precursor was prepared by phase inversion technique

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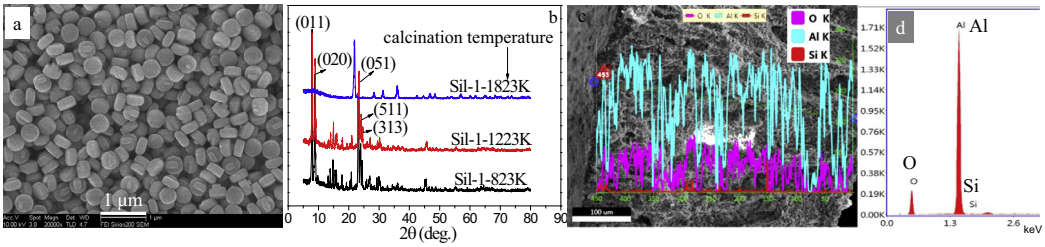

Fig. 1. SEM (a) and XRD patterns (b) of Sil-1 zeolite and EDX of Si-Al hollow fibers (c, d).

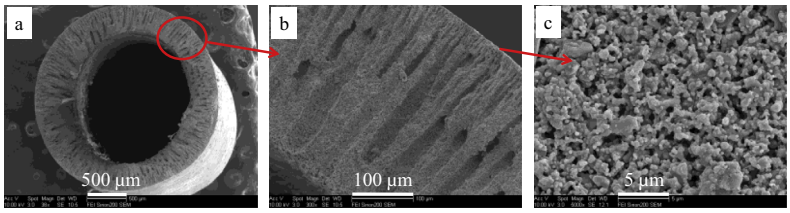

Fig. 2. SEM images of Si-Al hollow fibers (a, b: cross-section, c: external surface).

followed by high temperature sintering (1823 K), as described in previous works [11,12]. The spinning process was carried out at room temperature through a spinneret using water and a mixture of NMP and ethanol as the external and internal coagulants. The Al_2O_3 hollow fibers containing silica species were denoted as Si-Al support.

The solution used for preparation of Sil-1 membrane was prepared by mixing TEOS, TPAOH and water with a molar ratio of 1.0TEOS: 0.35TPAOH: 250H_2O. The solution was aged for 24 h at room temperature under stirring. The Si-Al support was immersed vertically in the synthesis solution in a Teflon vessel. The vessel was sealed in a stainless steel autoclave and placed in a 448 K oven for zeolite membrane deposition. The samples were washed, dried overnight at 373 K, followed by calcination at 823 K for 6 h. For comparison, Sil-1 membrane supported on bare Al_2O_3 hollow fibers was also prepared and denoted as A-1 and A-2 after one hydrothermal treatment and three hydrothermal cycles. The Sil-1 membranes supported on Si-Al support after one hydrothermal synthesis and three synthesis cycles were designated as B-1 and B-2. The samples were characterized by an X-ray diffractometer (XRD, Brucker D8 Advance, Germany) with a Cu-Kα radiation (λ = 0.1542 nm) and operated at a voltage of 35 kV and 30 mA. The morphology of support and membranes was ascertained by field emission scanning electron microscopy (FESEM, FEI Sirion 200, Netherlands). The permeability of membranes was tested by a soap-membrane flow meter at room temperature.

3. Results and discussion

Fig. 1a shows the SEM image of Sil-1 seed powders calcined at 823 K. A very regular size and shape with a mean particle size of about 200–300 nm was obtained. The XRD pattern of Sil-1 seed particles in Fig. 1b displays the characteristic peaks of MFI structure at (011), (020), (051), (511) and (313). The strong diffracted

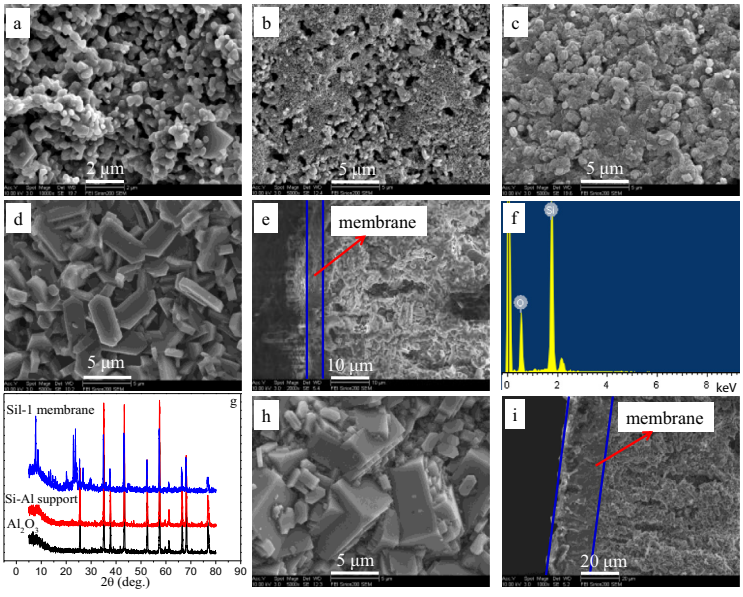

Fig. 3. SEM images of bare Al_2O_3 support (a) and Sil-1 membranes (b: A-1; c: A-2; d, e: B-1; h, i: B-2; a-d and h: surface; e, i: cross-section), EDX spectra (f) and XRD pattern (g) of B-1.

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