Surface-gel-conversion synthesis of submicron-thick MFI zeolite membranes to expedite shape-selective separation of hexane isomers

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ABSTRACT Ultrathin zeolite membranes are of paramount importance in accelerating gas transport during membrane separation, and lowering down their membrane thicknesses to submicron scale is deemed to be very challenging. Herein, we develop an advanced approach of surface gel conversion for synthesis of submicron-thick pure silica MFI (silicalite-1) zeolite membranes. Viscous gel is prepared by finely adjusting the precursor composition, enabling its reduced wettability. The unfavorable wetting of the support surface can effectively prevent gel penetration into alumina support voids. Aided by the seeds, the surface gel is directly and fully crystallized into an MFI zeolite membrane with minimal water steam. A membrane with a thickness of 500 nm is successfully acquired and it is free of visible cracks. Additionally, the as-synthesized membranes exhibit rapid and selective separation of hexane isomers by virtue of unprecedentedly high *n*-hexane permeance of $24.5 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ and impressive separation factors of 13.3–22.6 for *n*-hexane over its isomers. This developed approach is of practical interest for sustainable synthesis of high-quality zeolite membranes.

Keywords: zeolite synthesis, silica gel chemistry, thin membrane, hexane separation, isomer selectivity

INTRODUCTION

Zeolite membranes are of immense potential in separation technology owing to continuous working mode and energy efficiency. They are made of crystalline zeolites with uniform and molecule-level pores which are interconnected for mass transport [1]. Among different zeolites, MFI-type (ZSM-5 and silicalite-1) zeolite is a representative in membrane application because it has 5.5 Å multidimensional pores for sieving molecules of industrial interest (i.e., H_2/N_2) and tunable adsorption

properties favorable in separating important petrochemical compounds (i.e., alkanes) [2,3]. Although some progress has been made to enhance membrane performances by chemical synthesis (i.e., microwave irradiation) [4] and membrane engineering (i.e., crystal orientation) [5], MFI zeolite membranes still can hardly substitute existing technologies (i.e., distillation) due to their high fabrication cost [6–9]. One feasible way to compensate the high capital investment is to increase the flux in the separation process while maintaining the selectivity [10]. This means that the membrane thickness should be substantially reduced because the flux is inversely proportional to the thickness.

To achieve ultrathin (i.e., submicron in thickness) zeolite membrane is very challenging, evidenced by long-time endeavors from numerous scientists and engineers [11–17]. Currently developed synthesis methods for MFI thin films are categorized in the following: (1) controlled crystallization by additives such as fluoride ion and organic modifier. These additives have to be elaborately selected to slow down crystallization or to direct in-plane crystal growth [11,12]. (2) Support modifications by void plugging. Support pretreatments are sophisticated, requiring careful impregnation of extra substances into support voids [13,14]. (3) Seeding with zeolite nanosheets. Secondary growth of nanosheets-based seed layer can yield zeolite thin films [15–17]. In this method, generally speaking, very special and complex organic templates are needed for synthesis of MFI nanosheets.

The above syntheses are usually carried out in hydrothermal systems which require a lot of synthesis solution and produce a large amount of wastes. Alternative to hydrothermal synthesis, dry-gel conversion exhibits several advantages; for example, a small quantity of gel is

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needed for the membrane synthesis, zeolites are crystallized in high yields, and no liquid/solid wastes are disposed [18–22]. However, defects or cracks are prone to form in the membrane due to volume shrinkage in the gel-to-zeolite crystallization [18–23]. To acquire acceptable selectivity, the membranes have to be grown thick, which inevitably decreases the flux [21,24]. Thus, advanced methods are desirable for the green synthesis of highly permeable and selective MFI membranes. Herein, we develop a surface-gel-conversion approach, which can ideally meet the criteria of high permeability, good selectivity and sustainability for modern membrane technology. This approach involves first deposition of a thin layer of amorphous silica gel on the seeded support and its subsequent transformation into a membrane (Scheme 1). Hence, the top gel layer is determinative to zeolite membrane because the membrane thickness is a function of the gel thickness. In this regard, the microstructure of gel precursor is tuned to regulate its wettability and accordingly to precisely control the gel thickness. Assisted with minimal steam, the whole gel is fully crystallized to silicalite-1 zeolites (pure-silica MFI zeolite) on an alumina support, leading to a commensurately gel-thick membrane. The membrane thickness can be facily reduced to sub-1 μm by decreasing the gel layer. The membranes after additional growth are free of cracks and capable of separating hexane isomers with unprecedented permeance and impressive separation factor.

EXPERIMENTAL SECTION

Chemicals

Tetrapropylammonium hydroxide (TPAOH, 25% in wa-

Scheme 1 Schematic illustration of the surface-gel-conversion method for synthesis of the MFI zeolite membrane.

ter, Tianjin GuangFu Fine Chemical, China), tetraethyl orthosilicate (TEOS, 98%, Sinopharm Chemical Reagent Co., Ltd, China), and hexane isomers (*n*-hexane (nHEX), 2-methylpentane (2MP), 3-methylpentane (3MP), 2,3-dimethylbutane (23DMB), 2,2-dimethylbutane (22DMB), 98.0%, TCI) were purchased from the corresponding companies. Deionized water was used as the solvent. Porous Al_2O_3 discs (28 mm in diameter, 3 mm in thickness, 40% in porosity, 1–3 μm in pore size) were purchased from Guangdong Yuli Electronic Co., Ltd, China. These discs were polished with sand papers of 500, 1000 and 1500 mesh, washed with deionized water and then dried at 85°C for 5 h.

Syntheses of MFI seeds and membranes

MFI seeds in nanoscale were prepared with a synthesis solution in mole ratio of 1.0TPAOH:3.64TEOS:33.8 H_2O according to our published recipe [25]. Briefly, 8.0 g TPAOH solution (25%) was mixed with 7.46 g TEOS in a 25-mL polypropylene (PP) bottle to form a clear solution under stirring for 12 h at room temperature. This solution was transferred into a 25-mL Teflon-lined autoclave and heated at 120°C for 12 h. After cooling, the suspension was washed by repetitive centrifugations (18,000 r min^{-1} and 20 min for each run) until the supernatant was almost neutral. The seed solution of 1.0 wt% was prepared by dispersing seeds in deionized water. Seeding Al_2O_3 disc was carried out by immersing half height of the disc into 1.0 wt% seed solution with the polished surface downwards for 30 s. The wet disc was dried at 85°C for 30 min and the seeding procedure was repeated twice. The seeded surface was smoothed by finger with glove. The seeded Al_2O_3 disc was calcined at 550°C for 6 h (heating ramp of 1.0°C min^{-1}). MFI membranes were synthesized by the surface-gel-conversion method. The gel was prepared with a chemical composition of 1.0TPAOH:16.1TEOS:580 H_2O . Typically, 1.225 g TPAOH solution (25%) was mixed with 14.85 g water in a 25-mL PP bottle, and then 5.05 g TEOS was added in the bottle, with additional stirring for 48 h at room temperature. For comparison, reference solutions were prepared with compositions of 1.0TPAOH:16.1-TEOS:2765 H_2O (labeled as Ref.1) and 1.0TPAOH:3.2-TEOS:117 H_2O (labeled as Ref.2). The gel was deposited on the seeded support by the spin-coating technique. About 2 mL of the gel was added on the seeded Al_2O_3 disc and this disc was spun at 1500 r min^{-1} (<1 s to reach this speed) for 10 s (temperature of 25 $\pm$ 3°C, relative humidity of 40% $\pm$ 5%). The gel-coated disc was fixed horizontally on a sample holder and then placed in a 50-mL autoclave

with 0.2 mL water in the bottom. The autoclave was heated at 150°C in a static oven for 3 d for zeolite crystallization. The templates in the prepared membrane were removed by calcination at 550°C for 6 h with a heating rate of 1.0°C min⁻¹ in air.

Characterizations

The sizes and zeta potentials of silica colloids were measured on a Nano ZS90 laser particle analyzer (Malvern Instruments, UK) at room temperature (~25°C). The silica concentration was adjusted around 100 µg mL⁻¹ for the measurements. Fourier transform infrared (FT-IR) spectrum of freeze-dried gel was recorded on a Nicolet iS50 Fourier transform infrared spectrometer (Thermo Fisher Scientific, USA). Raman spectrum was recorded on the same sample using ultraviolet (UV) Raman spectroscopy (HORIBA Scientific, Japan), excited by 325 nm laser. X-ray diffraction (XRD) was performed on a Rigaku SmartLab X-ray diffractometer (Japan) with Cu-Kα radiation of λ=1.5418 Å (40 kV and 30 mA). The membrane morphology was inspected with field-emission scanning electron microscopy (FE-SEM, SU8010, Hitachi, Japan), which was equipped with energy-dispersive X-ray spectroscopy (EDX). The contact angle between gel and support was measured after gel stabilization for 10 s on the instrument of CA-100 (Shanghai Innuo Precision Instruments Co., Ltd, China). The viscosity values were collected on a rotational viscometer of DV-1 (Shanghai Yueping Scientific Instrument Co., Ltd, China). The depth of gel penetration (l_g) was estimated according to the following equation:

$$l_g = \frac{M_g}{\rho_g \times A \times \varepsilon},$$

where M_g is the weight of the penetrated gel (penetration time is about 10 s), ρ_g is the gel density (~1.0 g cm⁻³), A is the top surface area of the support (6.15 cm²), and ε is the support porosity (40%).

Membrane separation

The separation property was examined by the permeation of hexane isomer vapor (see Fig. S1). Hexane (nHEX) and its isomers (2MP, 3MP, 23DMB, and 22DMB) were stored in a saturator which was controlled from -20 to 0°C by a cooling apparatus. The zeolite membrane was fixed in a membrane module and sealed with xylene-resistant O-rings. This module was connected to a gas flow system. The carrier gas of argon passed through the saturator and supplied to the feed side of the membrane module with a flow rate of 50 mL min⁻¹. The sweep gas of

argon passed through the permeate side with a flow rate of 25 mL min⁻¹. The feed-to-permeate pressure drop was kept at 1.2 bar by a pressure gauge and the module temperature was maintained at 100°C in a tubular furnace. The concentration of hexane or its isomer in feed or permeate was monitored by online gas chromatography (Techcomp GC7900) equipped with TM-PLOT Al₂O₃/S column (30 m×0.53 mm×25 µm) and a flame ionization detector. H₂ or N₂ gas was detected on a thermal conductivity detector.

RESULTS AND DISCUSSION

A precursor solution with an initial mole composition of 1.0TPAOH:16.1TEOS:580H₂O (labeled as Gel) was optimized for the membrane synthesis. The silica chemical structure was studied by IR spectroscopy (Fig. 1a). IR spectrum contains strong bands at 1100, 800 and 460 cm⁻¹, corresponding to asymmetric, symmetric and bending vibrations of siloxane bonds [26,27]. The presence of siloxane rings is confirmed by the Raman shift at 490 cm⁻¹ (Fig. S2) [28]. Weak bands at 3630 and 965 cm⁻¹ (Fig. 1a) can be assigned to stretching and bending vibrations of silanol groups. Broadening of IR band at 3430 cm⁻¹ originates from hydrogen-bonded Si-OH with H₂O. XRD pattern (Fig. S3) reveals the amorphous nature of the silica, consistent with superimposed Raman shifts of 450–490 cm⁻¹ (Fig. S2). Reference precursors show the same band positions (Fig. 1a), indicating that they have the same chemical silica species, regardless of the addition of more water or concentrated TPAOH in solutions. Fig. 1b presents particle-size distributions of silica in the three precursors measured by dynamic light scattering (DLS). It can be seen that all precursors contain dispersed and stable colloids and the majority of particles are around 1.5 nm, showing that hydrolyzed silica after 48 h undergo similar sequential reactions of polymerization and particle growth. IR, Raman and DLS results reveal that silica particles in all precursors possess identical chemical structures and colloidal nature. Fig. 1c shows that the gel has the highest relative viscosity among the three precursors, indicating that the strongest interaction occurs between colloids in the gel. To unveil the underlying reason, the relative viscosities were investigated for different solutions by varying silica volume fraction (V_{fSiO_2}) and changing pH. The viscosity (Fig. S4a) increases rapidly as more silica is added in the solution, in particular for the high volume-fraction precursors (e.g., V_{fSiO_2} =0.03). High volume fraction means short particle-particle distance, resulting in strong inter-particle at-

Figure 1 (a) IR spectra of freeze-dried gel and reference precursors (1.0TPAOH:16.1TEOS:2765H₂O and 1.0TPAOH:3.2TEOS:117H₂O for Ref.1 and Ref.2, respectively). (b) Particle sizes of silica colloids measured by DLS. (c) Relative viscosities of the gel and reference precursors (viscosity relative to water). The insets are optical pictures of the precursors. (d) Penetration depths of the gel and reference precursors on the seeded Al₂O₃ support (penetration time is 10 s). The insets are contact angles of the precursors on the support.

traction *via* van der Waals force or hydrogen bonding [29]. The viscosity in Fig. S4b decreases when the solution has a high pH value. This can be rationalized by an increased repulsion between colloidal silica particles with high surface charges (zeta potentials of -25 , -26.5 and -38.6 mV for Gel, Ref.1 and Ref.2 solutions) [29]. The high content and low pH result in extensive and strong interactions between small silica colloids in the gel. As a consequence, the wettability of the gel precursor could be reduced. To prove this hypothesis, contact angles of silica-containing water drops on the seeded supports were measured in Fig. 1d. A contact angle of 45° is measured for the gel precursor. As comparison, contact angles of the other two precursors are approximately 0° . Higher contact angle describes less tendency of wetting the support [30], translating that the gel is difficult to penetrate into the support voids. A penetration depth of $75\ \mu\text{m}$ is mathematically determined for the gel (in 10 s, suitable for spin coating), which is significantly lower than those for reference precursors (2300 and $2030\ \mu\text{m}$ for Ref.1 and Ref.2).

In this study, MFI zeolite seeds were used to assist the conversion of the surface gel for the membrane synthesis. Without seeds, isolated crystals are formed in the gel and

loosely deposited on the Al₂O₃ support (Fig. S5). In contrast, by seeding the support with MFI nanocrystals, these crystals are inter-grown together after the secondary crystallization process and a compact membrane is yielded on the support (Figs 2 and 3). This promotion effect of seeds in zeolite growths of membranes is also observed in other studies [3,21,23]. Ref.1 and Ref.2 were also employed for the seeded syntheses of MFI membranes. It can be seen that the membrane prepared from Ref.1 is not continuous (Fig. S6) due to the very slow crystallization in dilute solution. In spite of membrane continuity achieved in Ref.2, the support pores are fully blocked by zeolite crystals, inevitably leading to an increase in membrane thickness (Fig. S6), consistent with the observation of precursor penetration depth (Fig. 1d). These findings show that the gel property and zeolite seeding are indispensable for obtaining high-quality membranes by surface gel conversion. In addition, the method of seed-assisted surface gel conversion allows the feasibility to accurately control the membrane thickness. Fig. 2 displays the relationship between the gel weight and membrane thickness. It can be seen that the membrane thickness is proportionally dependent on the gel weight. This result demonstrates that the MFI zeolite membrane

Figure 2 The thicknesses of MFI zeolite membranes as a function of the coated gel weights. The insets are SEM images of the prepared MFI membranes on porous Al_2O_3 supports.

Figure 3 (a, b) Top and side views of SEM images of the synthesized MFI zeolite membrane, and (c) the corresponding EDX mapping (Si and Al elements are presented by red and blue colors, respectively). (d) XRD patterns of the simulated MFI zeolite, porous Al_2O_3 support and synthesized membrane.

can be facilely thinned down to 500 nm through conversion of comparably-thick surface gel. This approach can be extended to other supports, exemplified by transferring porous alumina to silicon substrate. Linear correlation is observed for membrane thickness in function of gel thickness which can be easily varied by the weight of surface gel deposited on the substrate (Fig. S7).

Thin MFI zeolite membranes were synthesized after optimization of the synthesis conditions. Fig. 3a, b show top and side views of SEM images for the MFI membranes. The crystals are well inter-grown to form a con-

tinuous membrane and no cracks are inspected (Fig. 3a), showing that the obtained membrane is free of macroscopic defects. The membrane thickness is around 500 nm, determined from the cross-sectional SEM image (Fig. 3b). The membrane-support interface can be clearly distinguished from the EDX mapping (Fig. 3c), resulting from negligible gel penetration into the alumina support. As seen from the XRD pattern in Fig. 3d, the prepared membrane exhibits characteristic MFI zeolite diffractions and is free of other zeolite impurities. The XRD pattern also reveals that the membrane has a random orientation of crystals, in agreement with the SEM inspections (Fig. 3a, b).

The crystallization habit of the MFI zeolite in the membrane was investigated by SEM. Nano-zeolites were applied for seeding a representative flat substrate of silicon (Fig. 4a), similarly to Al_2O_3 support. MFI nanocrystals were first deposited on silicon and then a thick gel layer ($\sim 3.5 \mu\text{m}$) was intentionally coated on the seeds for a clear investigation. After 1 h crystallization, the individual seeds (100 nm) in spherical shape grow into larger crystals of $130 \text{ nm} \times 240 \text{ nm}$ by adsorbing silica nutrients from the top gel (Fig. 4b). In addition, the crystal growth proceeds faster horizontally than vertically,

Figure 4 Side views of SEM images of the silicon-supported seed layer (a) and zeolite membranes after crystallization of 1 h (b), 2 h (c), 4 h (d), 5 h (e) at 150°C . (f) The possible growth process of the MFI zeolite membrane. The dash lines present a defect in zeolite film in (c) and the gel-zeolite interface in (d).

evidenced by a larger crystal aspect ratio of ~ 2.0 (Fig. 4b) than that of ~ 1.0 for the seed (Fig. 4a).

This is beneficial for healing crystal boundaries. As the crystallization progresses for 2 h, zeolite crystals experience further growth and consequently merge into an inter-grown layer (Fig. 4c). At this stage, most crystals undergo preferentially vertical growth due to space limitation in the horizontal direction [25]. The crystallization process is briefly illustrated in Fig. 4f. As suggested in Fig. 4f and shown in Fig. 4d, some remaining defects (dash-lined area in Fig. 4c) can be eliminated by prolonging the crystallization time to 4 h. The nutrients in the upper gel (indicated by dash line in Fig. 4d) were completely consumed after 5 h and an entirely continuous zeolite membrane with well inter-grown crystals was yielded on the substrate (Fig. 4e).

The above results encourage us to evaluate the separation performance of this thin silicalite-1 zeolite membrane. Prior to separation, the quality of the silicalite-1 membrane was examined by standard hydrogen and nitrogen gas permeation tests. The selectivity for hydrogen over nitrogen measured at 25°C and 1.2 bar is 4.47, larger than the Knudsen constant of 3.73, supporting that the membrane is devoid of microscopic defects. Subsequently, the separation of hexane isomers was conducted under a standard Wicke-Kallenbach configuration using single or mixture vapor as the gas feed [31]. The gas permeation data at 100°C is compiled in Table 1. The permeance of $24.5 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ is measured for nHEX single gas. This is significantly larger than those for branched hexanes, translating to separation factors of 13.3–22.6. The separation factors follow the sequence of $13.7 \approx 13.3 < 21.5 < 22.6$ for nHEX/2MP, nHEX/3MP, nHEX/23DMB and nHEX/22DMB, respectively, in coincidence with the trend of their molecular sizes (Table 1), indicative of size-selective separation of hexane in the MFI membrane. For comparison, the performances of all relevant MFI zeolite membranes are summarized in Fig. 5 [32–43]. As shown, this submicron membrane gives

the highest permeance among all pertinent membranes. If the permeance is plotted *versus* the thickness (inset in Fig. 5), we have found that the nHEX permeance in general follows the decreasing trend with thickening the membrane. This finding soundly proves that reducing the membrane thickness is very effective in enhancing gas permeability. The separation factor exemplified by nHEX over 2,2-dimethylbutane is also quite good according to reported membranes, and it can be further improved by our coming effort in the future research.

When a binary gas is fed into the membrane, the gas permeance decreases and concurrently the separation factor increases in comparison with single gas, which can be interpreted by the competitive permeation mechanism of hexane isomers (nHEX, 2MP, 3MP, 23DMB, and 22DMB) [33]. The improved separation factor validates that the membrane is in good quality such as high continuity. To assess the practical application of this membrane, the separation test was carried out on a multicomponent mixture in volume composition of 13:20:18:19:30 for nHEX:2MP:3MP:23DMB:22DMB [44]. The permeance and separation factor were recorded as a function of time (Fig. 6). The permeance is very high initially at $6.0 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ and then slightly declines to $5.6 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ after reaching a steady state shortly within 10 h. The decreased permeance for nHEX is caused by an expansion of crystal lattice in the MFI zeolite [3], resulting in the permeation of some large molecules such as 23DMB and 22DMB through the membrane (Fig. 6). In spite of this, high separation factors of 16.6, 14.7, 18.3 and 23.6 for nHEX over 2MP, 3MP, 23DMB, and 22DMB at the plateau confirm that the membrane is free of pinholes and cracks. Both permeance and separation factor remain constant after an execution of 72 h, highlighting the high stability of this membrane.

CONCLUSIONS

In summary, we reported an advanced synthesis method

Table 1 Permeances of nHEX, 2MP, 3MP, 23DMB and 22DMB vapors and separation factors of nHEX over its isomers for the 500-nm-thick MFI zeolite membrane measured at 100°C with 1.2 bar feed gas

	Molecular diameter (Å)	Permeance ($10^{-7} \text{ mol s}^{-1} \text{ m}^{-2} \text{ Pa}^{-1}$)		Separation factor	
nHEX	4.3	24.5 (± 1.1) ^a	10.1 (± 0.9) ^b		
2MP	5.4	1.79 (± 0.2) ^a	0.64 (± 0.08) ^b	13.7 (± 0.8) ^a	15.7 (± 3.3) ^b
3MP	5.4	1.84 (± 0.3) ^a	0.69 (± 0.08) ^b	13.3 (± 2.7) ^a	14.7 (± 2.9) ^b
23DMB	5.8	1.14 (± 0.2) ^a	0.45 (± 0.06) ^b	21.5 (± 4.4) ^a	22.6 (± 4.9) ^b
22DMB	6.3	1.09 (± 0.1) ^a	0.33 (± 0.04) ^b	22.6 (± 3.0) ^a	30.5 (± 6.2) ^b

a), b) For single and binary (50:50 in volume) components, respectively.

Figure 5 A summary of separation performances in terms of nHEX/22DMB separation factor and nHEX permeance for relevant polycrystalline zeolite membranes (the black symbols are the results from previous reports and the red stars refer to the results for single and binary gases in this study). The inset is the plot of the nHEX permeance versus the membrane thickness.

Figure 6 Permeances of nHEX, 2MP, 3MP, 23DMB and 22DMB, and separation factors of nHEX over its isomers for a mixture (13:20:18:19:30 in volume for nHEX:2MP:3MP:23DMB:22DMB) as a function of test time for the 500-nm-thick MFI zeolite membrane measured at 100°C with 1.2 bar feed gas.

called surface gel conversion for sustainable fabrication of highly inter-grown pure-silica MFI zeolite membranes as thin as 500 nm on porous alumina supports. The chemical composition of the synthesized gel is determinative in decreasing its wettability on the support, which is crucial in the success of reducing the membrane thickness down to submicron level. The seeds and water vapor facilitate gel crystallization and crystal intergrowth. The thinnest MFI zeolite membrane exhibits a record high nHEX permeance exceeding $2.0 \times 10^{-6} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$ and an impressive selectivity greater than 13.3 for nHEX

over other isomers in the hexane separation. In addition to the efficiency in making high-quality membranes, this method has several advantages of facile operation, full materials utilization and no waste disposal, shedding light on possible transition of zeolite membrane chemistry to industrial separation through scalable, economic and reproducible production of high-performance zeolite membranes.

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Supplementary information Supporting data are available in the online version of the paper.

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表面胶转化法合成亚微米厚度MFI型分子筛薄膜应用于快速择形分离己烷异构体

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摘要 在膜分离中, 薄膜超薄化对促进气体传输至关重要, 而将膜厚降至亚微米级则极具挑战性. 本文中, 我们提出表面胶转化法来合成亚微米厚度的纯硅MFI分子筛膜. 通过精细调控前驱体化学组成制备了低浸润性的黏性胶, 有效防止凝胶从氧化铝载体表面渗透至空隙中. 在晶种的诱导下, 辅以少量水蒸气, 表面凝胶经历了直接和完全晶化过程, 形成了厚度为~500 nm并且连续致密的MFI分子筛膜. 合成的分子筛膜展示出对正己烷同分异构体的快速选择性分离性能: 超高的正己烷渗透量($24.5 \times 10^{-7} \text{ mol m}^{-2} \text{ s}^{-1} \text{ Pa}^{-1}$); 以及可观的分离系数(13.3–22.6, 正己烷对其同分异构体). 此外, 该方法还是一种绿色合成方法, 为可持续性制备高质量分子筛膜奠定了科学基础.