



An ensemble synthesis strategy for fabrication of hollow fiber T-type zeolite membrane modules

Miaomiao Ji, Xuechao Gao, Xuerui Wang, Yuting Zhang, Ji Jiang, Xuehong Gu*

State Key Laboratory of Materials-Oriented Chemical Engineering, College of Chemical Engineering, Jiangsu National Synergetic Innovation Center for Advanced Materials, Nanjing Tech University, 5 Xinnofan Road, Nanjing 210009, PR China

ARTICLE INFO

Keywords:

Zeolite membrane
Hollow fiber
Membrane module
Pervaporation
Dehydration

ABSTRACT

T-type zeolite membranes were prepared on four-channel α -Al₂O₃ hollow fibers by secondary growth method with an ensemble synthesis strategy. Prior to the membrane synthesis, a bundle of α -Al₂O₃ hollow fibers were assembled on the ceramic bases by enamel sealing. The membrane modules with membrane areas of 0.03 m² and 0.54 m² were then seeded with ball-milled T-type zeolite crystals by dip-coating. A uniform and continuous T-type zeolite seed layer was obtained for a seed concentration of 1.5 wt% and a coating time of 15 s. The seeded modules were then immersed in synthesis solution for ensemble hydrothermal crystallization. After optimizing the synthesis parameters, high-quality hollow fiber zeolite membrane modules could be prepared with good reproducibility. The membrane module with 0.03 m² exhibited a water permeation flux of 2.25 kg m⁻² h⁻¹ and a separation factor of 1348 for PV dehydration of 90 wt% ethanol/water mixture at 348 K. The membrane modules had long-term stability in acidic solution (pH ~ 3) for 450 h as well. A pilot-scale apparatus composed of two 0.54 m² membrane modules connected in series was further constructed for PV dehydration of 90 wt% isopropanol/water. The ensemble synthesis strategy paves a way for the industrialization of hollow fiber T-type zeolite membranes.

1. Introduction

Zeolite membranes with uniform microporous structures and extraordinary thermochemical stability have indicated great potential applications in organic solvent dehydration [1–3], gas permeation [4], membrane reactors [5], seawater desalination [6] and so on. NaA zeolite membranes have been commercialized for organic solvent dehydration since 1990s. Due to its excellent separation performance, more than 200 industrial plants have been built up by Mitsui Engineering and Shipbuilding (Japan), Jiangsu Nine Heaven (China) and GFT Membrane Systems GmbH (Germany) [7–9]. However, all of these plants are limited to the neutral systems (pH = 7–9) due to the low acidic resistance of NaA zeolite crystals with a Si/Al ratio of 1 [10,11]. T-type zeolite membranes (OFF-ERI intergrowth), containing an effective pore size of 0.36 × 0.51 nm² with a Si/Al ratio of 3–4, have been considered as an attractive alternative for dehydration of solvents with high stability under moderate acidic conditions [12,13].

The high separation performance and strong acidic resistance were demonstrated in tubular T-type zeolite membranes [13–19]. However, the high fabrication cost and low permeation flux have limit their industrial applications [20]. Caro et al. reported that 70% of the

investment was contributed by the substrate rather than the zeolite membrane layer [21,22]. Thus, porous ceramic hollow fibers with thin walls are considered as a promising substrate for zeolite membranes, which involve T-type [23], LTA [24–26], CHA [27] and MFI [28]. As reported in our previous work, the packing density of a hollow fiber membrane module was 600 m² m⁻³, which was almost three times higher than that of a tubular membrane module [24,25]. Meanwhile, it is anticipated to enhance the permeation flux by the asymmetric structures [29,30]. So far, the reported zeolite membranes were usually developed based on single channel ceramic hollow fibers. The single channel hollow fibers should be carefully handled during the synthesis, assembling into the module, and practical applications, to avoid fracture due to the poor mechanical strength [31,32].

In our previous work, four-channel α -Al₂O₃ hollow fibers with high mechanical strength have been developed for supporting NaA and DDR zeolite membranes [33–35]. Herein, four-channel α -Al₂O₃ hollow fibers are further explored to prepare T-type zeolite membranes. We proposed an ensemble synthesis strategy for preparation of hollow membrane modules, which is more practically applied for scale-up production. In the synthesis strategy, hollow fiber supports were first assembled on ceramic bases, and the hollow fiber bundle was then coated with ball-

* Corresponding author.

E-mail address: xuehonggu@yahoo.com (X. Gu).

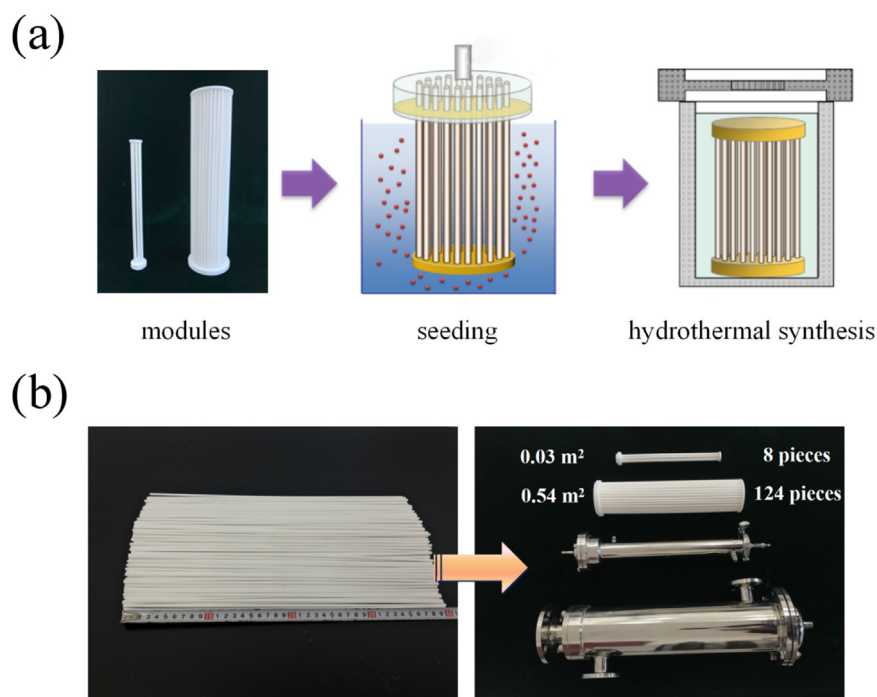


Fig. 1. Flow diagram of ensemble synthesis strategy for module fabrication (a) and photos of hollow fiber support and membrane module (b).

milled seeds and finally used for ensemble hydrothermal synthesis. The membrane modules were evaluated by pervaporation (PV) performance and acidic stability. A separation apparatus with an area of ca. 1 m² was also constructed for the dehydration of isopropanol.

2. Experimental

2.1. Ensemble synthesis of hollow fiber membrane modules

Hollow fiber T-type zeolite membrane modules were prepared by an ensemble synthesis strategy, as shown in Fig. 1. A bundle of α -Al₂O₃ hollow fibers with 8 or 124 pieces were first grafted on two bases by enamel sealing, and the modules with the membrane areas of 0.03 or 0.54 m² were derived, where one end of the module is open and the other is dead. The hollow fiber support have an average pore size of 0.27 μ m and a porosity of 45%, as well as a bending load of 35 N. For the preparation of T-type zeolite membranes, the α -Al₂O₃ hollow fiber modules were first coated with original seeds or ball-milled seeds. The ball-milled seeds were obtained by milling original seeds (average particles size of $\sim$ 1.2 μ m) with a planetary ball mill (Retsch. Ind. Ltd., PM-100) using zirconium oxide balls as grinding media. The ball-milled seeds had an average particle size of $\sim$ 200 nm. For seeding, the hollow fiber module was dipped into an aqueous 1.0–2.0 wt% seed suspension for 10–20 s. After being dried for overnight, the seeded modules were immersed in a synthesis solution for hydrothermal crystallization. The synthesis solution was prepared by dissolving sodium aluminate, colloidal silica, sodium hydroxide and potassium hydroxide in D.I. water at room temperature. The molar ratio of the synthesis precursor was 1 SiO₂: 0.05 Al₂O₃: 0.26 Na₂O: 0.09 K₂O: 14 H₂O, where all the chemicals were industrial grade and purchased from commercial companies in China. After being vigorously stirred for 24 h, homogeneous synthesis gel was transferred into a Teflon-lined stainless steel autoclave containing the seeded hollow fibers modules held vertically. The hydrothermal synthesis was carried out at 375 K for 35–45 h. After crystallization, the hollow fiber membranes were washed with D.I. water and dried at 333 K overnight.

2.2. Pervaporation

The PV performance of as-synthesized hollow fiber membrane modules was evaluated by dehydration of ethanol/water and isopropanol/water mixtures at 348 K. The schematic diagram of the experimental apparatus was depicted in our previous paper [36], where the T-type zeolite membrane module was mounted in a stainless steel module, with the liquid mixture feeding to the shell side at a constant flow. The permeate was extracted from the lumens of the hollow fibers by a vacuum pump, maintaining at a pressure below 200 Pa. Fig. 2 shows the pilot-scale dehydration apparatus operated in pervaporation. The liquid mixture is firstly fed to the heat exchanger with a constant flow pump and then flow through the outer side of membrane modules, finally circulates to the feed tank. The permeate is removed from the lumen side by a vacuum pump and trapped by a condensing tank. The separated product is finally collected into the product tank. Two membrane modules with each membrane area of 0.54 m² were connected in series. The separation temperature was controlled at 348 K. The feed pressure was operated at atmospheric pressure while the permeate pressure reached 100 Pa.

Both of the samples in the feed and permeate sides were analyzed by

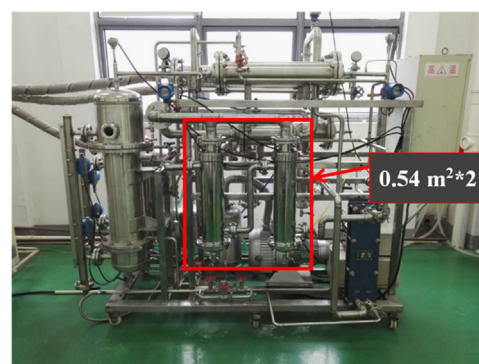


Fig. 2. Schematic diagram for the pilot-scale PV apparatus with hollow fiber T-type zeolite membrane modules.

Download English Version:

<https://daneshyari.com/en/article/7019711>

Download Persian Version:

<https://daneshyari.com/article/7019711>

[Daneshyari.com](https://daneshyari.com)