



Fabrication of zeolite/polymer composite membranes in a roller assembly



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ABSTRACT

Membranes are cost-effective solutions for many industrial separations. Polymer membranes are widely used in gas separations. Because of the solubility-diffusion mechanism of transport, the permeance and selectivity of polymer membranes are inversely related, and is considered a limitation. Zeolite membranes do not have this limitation, but zeolite membrane synthesis is a batch process with long synthesis times and presence of defects. The resulting high costs of manufacture make zeolite membranes non-competitive for most applications. In this study, we present a roll-to-roll method for zeolite synthesis on a polymer support exploiting a gel that leads to rapid zeolite crystallization and a bendable zeolite membrane structure. Membranes were grown under both compressive and tensile stress, and with zeolite structure both on top and within the pores of the polyethersulfone (PES) support. The structure of the membranes was evaluated by electron microscopy. Membranes were coated with a thin layer of polydimethylsiloxane, and evaluated for CO₂/N₂ separation, relevant for CO₂ capture from flue gas of power plants. As long as the zeolite membrane is grown within the PES support, highly reproducible CO₂/N₂ separation performance with CO₂ permeance of 1881 ± 204 GPU and CO₂/N₂ selectivity of 34 ± 4 was observed immaterial of the stress conditions under which it was grown. For zeolite membrane grown on top of the PES support, the compressive stress resulted in crack formation, with poor transport properties. Demonstration of zeolite membrane fabrication with roll-to-roll method has the potential for industrial level scale up.

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1. Introduction

Polymer membranes are cost effective platforms for a wide variety of gas separations, and in particular in carbon sequestration applications [1]. With increase in gas permeance, there is typically a decrease in selectivity for polymer membranes (Robeson limit) [2]. Inorganic zeolite membranes do not have this limitation. Computational studies have noted that faujasitic zeolite with pore size of 7.4 Å can have CO₂/N₂ selectivity higher than 500 and CO₂ permeability of 10,000 Barrer (1 Barrer = 3.35 × 10⁻¹⁶ mol m/(m² s Pa)) considerably better than polymeric membranes [3]. Because of their unique gas separation performance, zeolite membranes have been an active research area [4–7]. However, application of zeolite membranes on a commercial scale is limited to only one application, that of ethanol pervaporation, because of

their high cost [5]. Most zeolite membrane research is focused on the laboratory scale.

Polymer membranes have low manufacturing costs because of the roll-to-roll fabrication process [8]. Roll-to-roll process has also been applied for nanoparticle alignment on membrane support and pattern printing on films [9–12]. However, roll-to-roll fabrication method has never been applied in zeolite synthesis, because conventional zeolite synthesis takes long time and with the typical rigid alumina supports, roll-to-roll technology cannot be used [5].

Our group has reported methods to decrease zeolite crystallization time, synthesize zeolite membranes on polymer supports and fabricate bendable zeolite membranes [13–16]. A dehydration/rehydration strategy was developed to decrease the synthesis time of faujasitic zeolites with crystallization times of <2 h, and this time scale is potentially applicable for roll-to-roll synthesis. With this method, flat-sheet zeolite membranes were synthesized and examined for CO₂/N₂ gas separation [14]. A bendable zeolite membrane concept was developed by growing zeolite layer only within a polymer membrane [15]. The gas separation property was maintained after bending to a certain curvature.

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In this study, a roll-to-roll synthesis setup for zeolite membrane growth within a polymer has been designed and applied for membrane growth. Membranes were characterized with X-ray diffraction (XRD), scanning electron microscopy (SEM) and CO₂/N₂ gas separation. This study demonstrates the potential of producing zeolite membranes in similar fashion to polymeric membranes.

2. Experimental section

2.1. Chemicals

Ludox HS-30 colloidal silica (SiO₂, 30%), aluminum isopropoxide (Al(O-CH(CH₂)₂)₃, 98%), tetramethylammonium bromide ((CH₃)₄NBr, 98%) and Ludox SM-30 colloidal silica (SiO₂, 30%) were purchased from Aldrich (Milwaukee, WI, USA). Aluminum hydroxide (Al(OH)₃, 76.5%) was purchased from Alfa Aesar. Tetramethylammonium hydroxide ((TMAOH), 25% aqueous solution) was purchased from SACHEM Inc. Sodium hydroxide (NaOH, 99.0%) was purchased from Fisher Scientific. Dehesive 944 Polydimethylsiloxane (PDMS) was provided by Wacker Silicones, Inc. Helium (4.5 grade), carbon dioxide (4.0 grade) and nitrogen (4.5 grade) were purchased from Praxair. Polyethersulfone (PES) 300 kDa membrane was purchased from MILLIPORE Biomax. H₂O used in this study was purified by a Millipore ultrapure water system. All chemicals were used as received without further purification.

2.2. Zeolite membrane synthesis with roll-to-roll setup

Zeolite Y Nanoparticle Synthesis Nano sized zeolite Y seeds were synthesized according to literature with composition of 0.048 Na₂O:2.40 (TMA)₂O(2OH):1.2 (TMA)₂O(2Br):4.35 SiO₂:1.0 Al₂O₃:249 H₂O, where TMA⁺ is tetramethylammonium cations [17]. Briefly, 26.2 g Ludox HS-30 and 10.46 g TMAOH were mixed in a sealed bottle and stirred at room temperature for 30 min. 12.5 g aluminum isopropoxide was dissolved in mixture of 76.5 g H₂O and 52.3 g TMAOH solution, and heated in a water bath at 70 °C until complete dissolution. After cooling to room temperature, 13.1 g TMABr was added to alumina source solution followed by mixing with the silicon source. The clear sol was aged at room temperature with stirring for 3 days and then in an oil bath at 100 °C for 4 days. After synthesis, nano-zeolite particles were separated by ultracentrifugation (using Sorvall MX and Beckman Coulter Allegra 64RD centrifuges), and washed until pH of supernatant was 7. Purified nanozeolite seed dispersion was stored as a 1 wt% aqueous stock solution.

Deposition of Zeolite Y Nanoparticle on PES support Nanozeolite seed particles were deposited on PES supports by vacuum assisted dip-coating. PES supports were soaked in distilled water overnight and then in isopropanol for 1 h before washing with water again. Nanozeolite dispersion was ultra-sonicated for 1 h and diluted with distilled water to the required concentration. About 20 mL of nanozeolite suspension was placed in petri dish. The PES support was dipped in the petri dish for 3 s, A -25 psi vacuum was applied on the back of the PES support to pull the seeds onto the support. After coating, the support was dried at room temperature overnight and stored in plastic sample bags.

Roll-to-Roll Synthesis Setup The roll-to-roll synthesis setup designed in this study is shown in Fig. 1. This setup consists of 4 parts: rollers, reactor, temperature controller and a rehydration assembly. Two identical motor rollers and one fixed column all of diameter of 5.1 cm was used. Nonwoven fabric band was affixed to the rollers and fixed column, and could be moved with adjustable rolling speed. Temperature controller consists of a thermocouple, a digital temperature controlling box and four 100 W heaters placed in the holes at the bottom of the reactor. The reactor temperature is

set at 100 °C. Rehydration of gel was realized by continuous addition of water from a burette.

Roll-to-Roll Synthesis of Zeolite Membranes Gel composition used in the reactor is: 8.3 Na₂O:1 Al₂O₃:6.4 SiO₂:483.9 H₂O. After dissolving 4.416 g of Al(OH)₃ and 14.58 g NaOH in 170.48 g H₂O, 27.7 g Ludox SM-30 was added to the gel. Mixed gel was sealed in polypropylene bottle and aged at room temperature for 4 h. Aged gel was transferred to a dehydration/rehydration hydrothermal setup for removal of half the water in 1 h. This partially dehydrated hot gel was then immediately transferred to the reactor shown in Fig. 1 and heated to 100 °C. Seeded PES support was stapled onto the outer side of rolling non-woven fabric band. Entire PES support was immersed into the gel in the beginning of zeolite growth. During 1 h of zeolite growth process, water was added to the gel, diluting the gel from 120 mL to 200 mL. At the same time, PES support moved through the gel due to movement of the rolling non-woven fabric via the rollers. After zeolite growth, membrane sample was washed with flowing water and rubbed with a fur brush, soaked in water to remove residual surface species and dried for further study.

2.3. Polydimethylsiloxane (PDMS) coating

Zeolite membranes were spin coated with PDMS (Dehesive 944 kit) before gas separation test. Commercial PDMS comes in 3 separate bottles, containing PDMS monomer, catalyst and cross-linker (the exact nature of these chemicals is proprietary, and the procedure followed was supplied by the vendor). First, PDMS monomer solution was diluted with heptane. After complete dispersion, cross linker and catalyst were added with the ratio of 100:1:0.5 (PDMS: Cross linker: Catalyst) to prepare PDMS precursor solution. Before spin coating, zeolite membrane sample was taped on a spin coating support, which provides mechanical stability in spinning process. PDMS precursor solution was dropped to cover entire membrane surface area, and left for 3 s before spinning. Samples were spun at 2000 rpm for 5 s followed by 4000 rpm for 1 min. After coating, PDMS was polymerized at room temperature overnight.

2.4. Characterization

Bruker D8 X-ray Diffractometer with CuK α ($\lambda = 1.5405 \text{ \AA}$) was used to characterize the phase composition of zeolite membranes. FEI Helios Nanolab 600 Dual Beam Focused Ion Beam/Scanning Electron Microscope (FIB/SEM) was employed to characterize surface morphology of zeolite membrane samples.

2.5. Gas separation study

Fabricated zeolite membrane was applied for CO₂/N₂ separation. Gas flow compositions were controlled with a flow box and mass flow controllers from SIERRA Instruments Inc. Feed gas and sweep gas have flow rates of 60 mL/min and 30 mL/min, respectively. Compositions of permeate and retentate gas were investigated with a SRI 310C gas chromatograph equipped with a Hysep D column and TCD detector.

3. Results

3.1. Synthesis of zeolite membrane

The roll-to-roll synthesis setup shown schematically in Fig. 1a was employed to synthesize zeolite membranes while moving through a heated aluminosilicate gel. Fig. 1b is a picture of this setup. This setup consists of 2 moving rollers, a fixed column

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