

Zeta potential measurements of zeolite Y: Application in homogeneous deposition of particle coatings

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Abstract

Zeolite membranes are typically grown from thin coatings of sub- μm size zeolite particles on porous supports. The particles have the function of seeds in forming a dense zeolite membrane by a secondary growth process. This study focuses on how seed layer coatings can be optimally produced, crack- and agglomerate-free, by dip coating from stable dispersions of nano-crystalline zeolite Y. Surface charge and colloidal charge stabilization were optimized by adjusting the pH to 11.5 at a low ionic strength. The coatings were formed in a combination of film and slip coating on quasi-homogeneous flat porous $\alpha\text{-Al}_2\text{O}_3$ supports with a typical surface pore diameter of 40 nm. The optimum pH was established with zeta potential measurements of the dispersion. Optimum conditions resulted in excellent homogeneity, <500 nm thickness of the seed layers with a deposition technique that makes it possible to coat large areas. This makes this process suitable for the production of thin, defect-free, and stable supported zeolite Y membranes by subsequent secondary growth. © 2007 Elsevier Inc. All rights reserved.

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1. Introduction

Zeolites are a class of inorganic crystalline materials with well-defined sub-nanometer pores and unique structures. This has resulted in important applications as catalysts, adsorbents, ion-exchangers, and molecular sieves [1–7]. For many applications, performance can be significantly increased by preparing thin films and/or membranes [8].

Existing wet-chemical methods of zeolite membrane synthesis use a two-step approach in which a particle seed layer is prepared first, followed by densification through secondary growth. While this method generally gives good results [1,3,4,9–13], the quality and thickness of the eventual membrane depend strongly on the quality of the seed

layer, in particular its uniformity and thickness. The seed layer needs to cover the (porous) support completely, and consist of a dense-packing of individual dense particles with similar morphology and dimensions. Therefore, improvements in seed layer deposition techniques are important for the development of zeolite chemical syntheses [1,11,12,14].

Methods that are currently available for the deposition of previously formed small zeolite crystals include: rubbing [1,3,4,15], spin coating [16,17], electrostatic adsorption [2,9,11,12,18,19], electrophoretic deposition [20–22], and dip coating [1,10,12]. The latter method is preferred for practical applications [1,12] since it does not require any time-consuming and costly surface pre-treatment of support and/or particles, and because it can be applied over large areas. The pre-treatment is needed, for instance, for the electrostatic and electrophoretic adsorption techniques. An important requirement for the dip coating method to be successful is that the zeolite particle dispersions must be stable in the dispersed state. Pre-mature agglomeration

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due to colloidal instability leads to inhomogeneous coverage and the formation of micro-cracks and delamination during subsequent processing. Colloidal stability can be improved in two ways:

- Application of surface chemical modification of the zeolite crystals by charged polymers to achieve electro-steric colloidal stabilization.
- Utilizing spontaneous charge-stabilization by adjusting the aqueous dispersion medium pH while maintaining a low ionic strength.

In this work, the latter approach is used in view of its simplicity and viability. It is shown how thus optimized, state-of-the-art nano-zeolite Y (FAU-type framework) dispersions can be used to obtain the desired seed layer characteristics on optically flat, homogenous macroporous α -Al₂O₃ supports with a typical pore \varnothing of 40 nm. In addition, it is shown how the selection of proper pH conditions can be carried out with straight-forward zeta potential measurements of the dispersions as a function of pH.

2. Experimental

2.1. Zeta potential measurements

The zeta potential of dispersed zeolite Y crystals was determined by an electro-acoustic technique with a ZetaProbe Analyzertm (Colloidal Dynamic Inc., Warwick, RI) at room temperature. The measurements were performed with commercially available 1 μ m \varnothing zeolite Y crystals (Union Carbide, Danbury, CT), dispersed in distilled water (Gradient A10 Milli-Q water distillator, Millipore, Billerica, MA) at a solid load of 1 wt.%. The choice of commercial material is determined by the fact that the ZetaProbe instrument cannot perform zeta potential measurements for nano-sized particles. This problem is related to interference by Brownian motions and insufficient overall charging of small particles. The Si/Al ratio for the 1 μ m zeolite Y was \sim 2.5, which is the closest available to our synthesized nano-Y (see Section 2.2).

After 15 min ultrasonication of the suspension, the zeta potential was measured as a function of pH by titration with 4 M HNO₃ (EMD Chemicals, Gibbstown, NJ) to pH = 2.0 or 10 wt.% tetramethylammonium hydroxide (TMAH) (prepared from 25 wt.% solution, SACHEM, Austin, TX) to pH = 12.5.

2.2. Zeolite crystal synthesis

Zeolite Y crystals with $\varnothing \approx$ 150 nm were synthesized from an initially clear reactant solution as described in [23,24]. The reactant solution had a molar composition of 0.037Na₂O:1.0Al₂O₃:3.13(TMA)₂O:4.29SiO₂:497H₂O (NaOH, Mallinckrodt, Hazelwood, MO; Al(OH)₃, Alfa Aesar, Ward Hill, MA; SiO₂, Ludox 30, Sigma-Aldrich, St. Louis, MO) and resulted in nano-Y crystals with Si/

Al ratio of \sim 1.7. The synthesis mixture was placed in a Teflon bottle and heated at 98 °C in an oven (Blue M B-2729-Q, New Columbia, PA) for 3 days.

After quenching the reaction, the crystals were collected using an ultra-high speed centrifuge (Allegratm 64R, Beckman Coulter, Fullerton, CA) at 23 000 rpm for 25'. The sediments were washed repeatedly with distilled water until the pH of the rinsing solution became neutral.

Zeolite Y dispersions for dip coating were prepared from 20 mL of aqueous 0.2 wt.% polyvinylpyrrolidone (PVP, $M_r \sim$ 360 000, Fluka, Neu-Ulm, Switzerland) solution mixed with 17 mg of nano-Y crystals at different pH 8.5, 9.5, 10.5, 11.5 (measured with Orion 710Aplus pH/ISE meter, Richmond, VA). Mixing was enhanced by ultrasonic treatment for 3.5 h at a power of 130 W (45 kHz, Model FS30, Fisher Scientific, Fair Lawn, NJ). During this treatment, the temperature of the mixture was kept below 23 °C by adding ice to the ultrasonic bath vessel.

2.3. Supports and dip coating procedure

The supports were macroporous α -Al₂O₃ disks with thickness of 2 mm and diameter of 42 mm, routinely produced by our group from AKP15 α -Al₂O₃ powder (Sumitomo Chemical Co. Ltd., Tokyo, Japan). The average pore diameter, surface roughness, and porosity of the supports were about 40 nm, 25 nm, and 30%, respectively [25].

Dip coating was performed with the supports mounted on a computer-controlled arm that makes a large radius rotational movement to drag the support through the surface of the dispersion (MEMDIP 1, Pervatech BV, 7468 MC Enter, Netherlands). The linear speed before and after dipping was 15 cm/s, and 1 cm/s during the actual dipping process. In all experiments, single coatings were performed. The dipping process took place in a clean-room environment with an air quality of better than class 1000 to reduce airborne contamination of the surface.

After the dip coating procedure was complete, the supports were covered and dried at 40 °C in air for 24 h in a drying stove (Lindberg/Blue M gravity oven, Asheville, NC). To remove the organics and enhance the bonding between the zeolite layer and the support [10], calcination was done in a programmed box furnace (Model 650-14, Fisher Scientific, Fair Lawn, NJ) to a maximum temperature of 400 °C for 3 h in air using a heating and cooling rate of 0.5 °C/min.

2.4. Sample characterization

The surface microstructure before and after dipping was characterized by scanning electron microscopy (SEM) with an XL-30 ESEM (Philips Electron Optics, Eindhoven, Netherlands). The layer thickness was estimated with the same technique by investigating perpendicular fracture surfaces. The average crystal size of the deposit was estimated by measuring the apparent dimensions of 30...50

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