Supporting Information

Synthesis of NaA Zeolite Membranes with High Water Flux on Macroporous Si_3N_4

Chaochen Zou[a, b], *Yongfeng Xia*[a], *Dongxu Yao*[a], *Ming Zhu*[a], *Jun Zhao*[a], *Rui Yao*[c, **], *Yu-Ping Zeng*[a, *]

[a] State Key Laboratory of High-Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 200050, China

[b] Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China

[c] School of Materials Science and Chemical Engineering, Ningbo University, Ningbo, 315211, China

* Corresponding author.

** Corresponding author.

E-mail address: yaorui@nbu.edu.cn (R. Yao), yuping-zeng@mail.sic.ac.cn (Y.P. Zeng).

Full postal address: Shanghai Institute of Ceramics, 585 Heshuo Road, Jiading District, Shanghai, China

Table of Contents

Text S1: Raw materials

The chemicals employed in this work comprised tetraethyl orthosilicate (TEOS, SiO_2≥28wt.%, Shanghai Lingfeng Chemical Reagent Co., Ltd,), sodium aluminate with 98% purity (Shanghai Aladdin Biochemical Technology Co., Ltd.), sodium hydroxide (NaOH) with 96% purity (Sinopharm Chemical Reagent Co., Ltd). And the reagents used for porous Si_3N_4 support included ethanol (AR, purity ≥99.7 wt.%, Shanghai Titan Scientific Co., Ltd.), Commercial Si_3N_4 powder (purity ≥ 99.99 wt.%; α-phase ≥ 95.5 wt.%; d50 = 0.7 μm, Qingdao Cixing New Materials Co., Ltd, China), Y_2O_3 (d50 = 5 μm, purity ≥99.9 wt.%, Yuelong Co., Ltd.), and polyvinyl butyral (purity ≥99 wt.%, Sinopharm Chemical Reagent Co., Ltd., Shanghai, China).

Text S2: Preparation of the LTA membrane on porous alumina

The porous alumina was prepared by dry press pressing at 20 MPa. The ceramic slurry was first obtained by mechanically mixing commercial Al_2O_3 powder (Indonesia Chemical Alumina, A-12-40M), 2 wt.% polyvinyl butyral, and ethanol. The mixture was then dried at 80°C and sieved through an 80-mesh screen. After pressing, the green bodies were dewaxed at 600°C in air for 2 h and sintered in air at 1600°C for 2 h. The resulting porous Al_2O_3 exhibited a porosity of 38% and a mean pore diameter of 1.57 μm. All supports were polished with 1200-grit sandpaper to reduce surface roughness. The seeding procedure was identical to that employed for the silicon nitride supports, as detailed in Section 2.3, involving a bottom seed layer of 1.91 μm and a top seed layer of 0.47 μm. Hydrothermal synthesis was carried out following the same protocol used for membrane Q3 (Section 2.3). Under these consistent conditions, a total of seven membranes—designated M8 through M14—were fabricated.

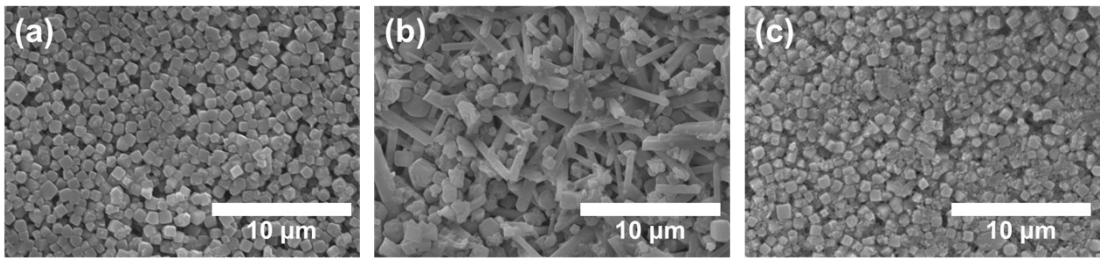

Fig. S1. Surface SEM images illustrating the sequential steps of the double-layer seeding process on Si_3N_4 supports: (a) after deposition of the primary seed layer using 1.91 μm seeds (2 wt.% suspension); (b) the primary-seeded support after

removal of loosely seeds by swabbing; (c) the final double-layer structure, consisting of the swabbed primary layer (1.91 μm seeds) overlaid with a secondary seed layer of 0.71 μm seeds (0.25 wt.% suspension).

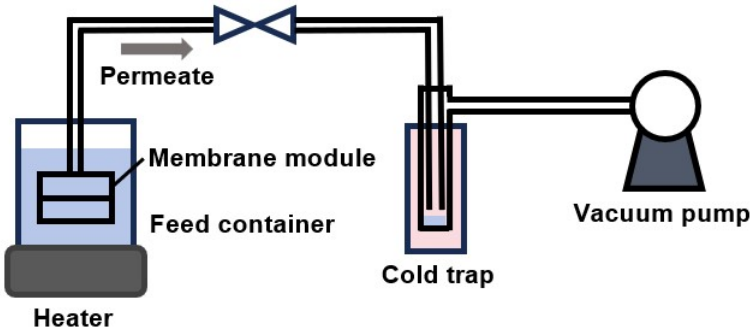

Fig. S2. Schematic diagram of the PV test apparatus for NaA zeolite membranes.

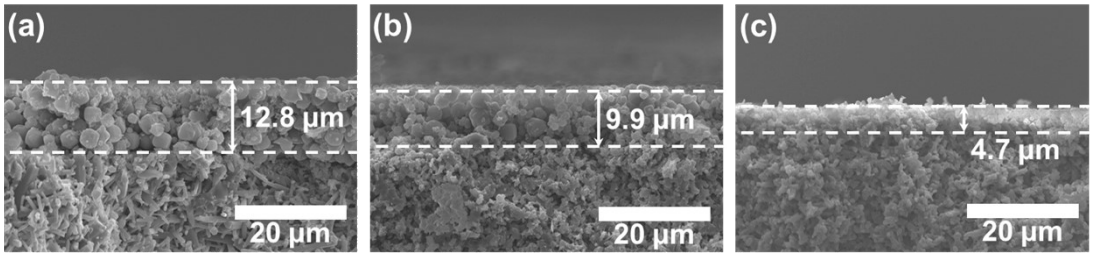

Fig. S3. The cross-sectional SEM images of seeded layer on macroporous Si_3N_4 by using (a) 1.91 μm, (b) 0.71 μm and (c) 0.47 μm NaA crystals as top seeds.

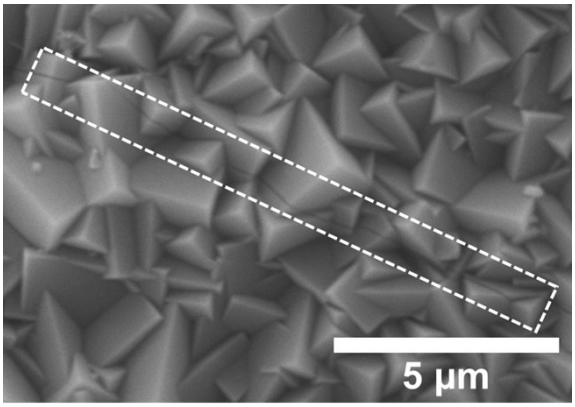

Fig. S4. Surface SEM image of NaA zeolite membrane (W4) prepared with 0.5 wt.% NaA seed suspension.

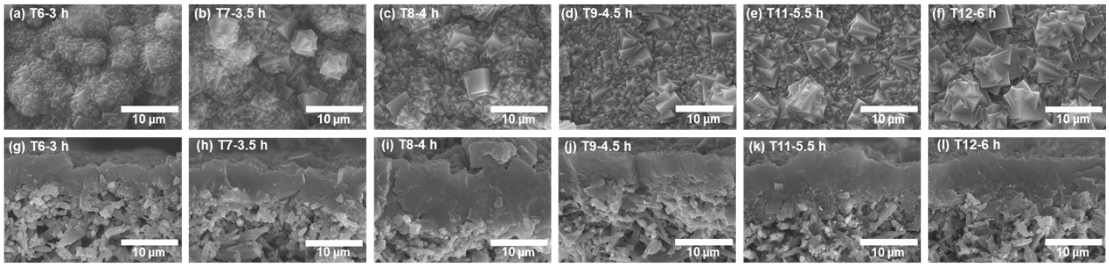

Fig. S5. Surface and cross-sectional SEM images of NaA zeolite membranes prepared with different crystallization times: (a)-(f) Surface SEM images; (g)-(l) Cross-sectional SEM images. Herein, T6, T7, T8, T9, T11, and T12 correspond to crystallization times of 3h, 3.5 h, 4 h, 4.5 h, 5.5 h, and 6 h, respectively.

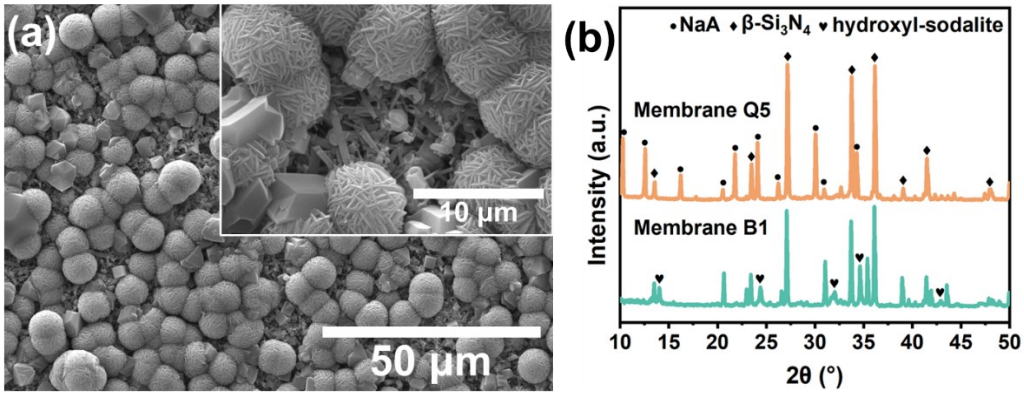

Fig. S6. Surface SEM image of zeolite membrane on unseeded bare Si_3N_4 support (Sample B1) (a) and XRD patterns of samples B1 and Q5 (b).

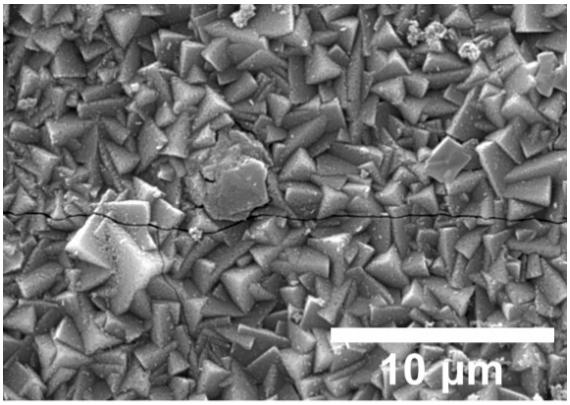

Fig. S7. SEM image of NaA zeolite membrane Q1 after pervaporation test.

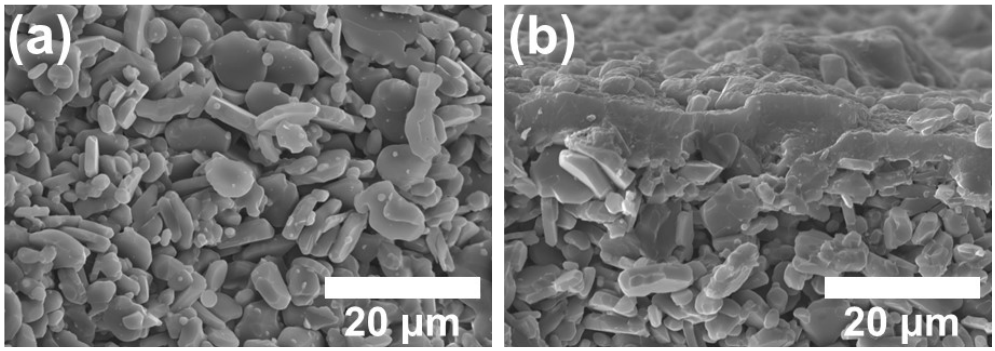

Fig. S8. Surface SEM image of porous alumina (a) and cross-sectional SEM image of NaA zeolite membrane M8 on porous alumina (b).

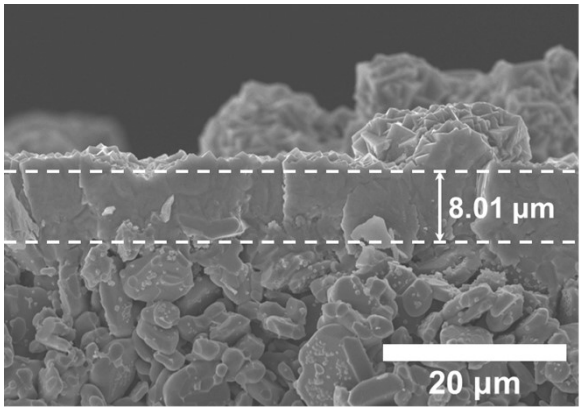

Fig. S9. Cross-sectional SEM image of NaA zeolite membrane M10 on porous alumina.

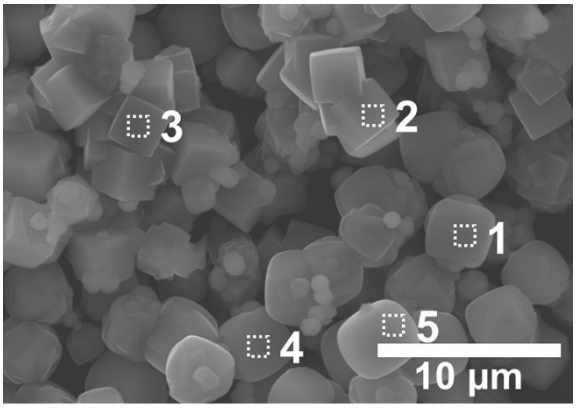

Fig. S10. SEM image of NaA crystals isolated from the synthesis gel used in preparing membrane Q5.

Table S1. Atomic concentration of NaA crystals isolated from the synthesis gel used in preparing membrane Q5.

Spectrum	Oxygen (O)	Sodium (Na)	Alumina (Al)	Silica (Si)	Si/Al
1	64.04	12.42	11.74	11.80	1.01
2	66.84	11.01	10.96	11.19	1.02
3	59.58	13.45	14.17	12.80	0.90
4	62.84	14.15	11.57	11.43	0.99
5	63.97	12.27	11.85	11.91	1.01
Mean	63.45	12.66	12.06	11.83	0.98
Sigma	2.62	1.20	1.23	0.62	0.05