Rapid Synthesis of a CHA Membrane Using a Small Tubular Reactor

Rizqan Jamal [1], Manabu Miyamoto [2,*], Yasuhisa Hasegawa [3], Yasunori Oumi [4] and Shigeyuki Uemiya [2]

[1] Department of Engineering Science, Graduate School of Engineering, Gifu University, 1-1 Yanagido, Gifu 501-1193, Japan; rizqan.jamal.p7@s.gifu-u.ac.jp
[2] Department of Chemistry and Biomolecular Science, Faculty of Engineering, Gifu University, 1-1 Yanagido, Gifu 501-1193, Japan; uemiya.shigeyuki.j8@f.gifu-u.ac.jp
[3] National Institute of Advanced Industrial Science and Technology, 4-2-1 Nigatake, Sendai 983-8551, Japan; yasuhisa-hasegawa@aist.go.jp
[4] Institute for Advanced Study, Gifu University; 1-1 Yanagido, Gifu 501-1193, Japan; oumi.yasunori.x2@f.gifu-u.ac.jp
* Correspondence: miyamoto.manabu.v5@f.gifu-u.ac.jp

Table of Contents

1. Hydrothermal synthesis procedure **2**

2. CHA seed crystal..... **3**

3. Effect of dip-coating solution..... **4**

4. Effect of draying condition for seeded support..... **7**

5. Effect of hydrothermal synthesis time..... **8**

1. Hydrothermal synthesis procedure

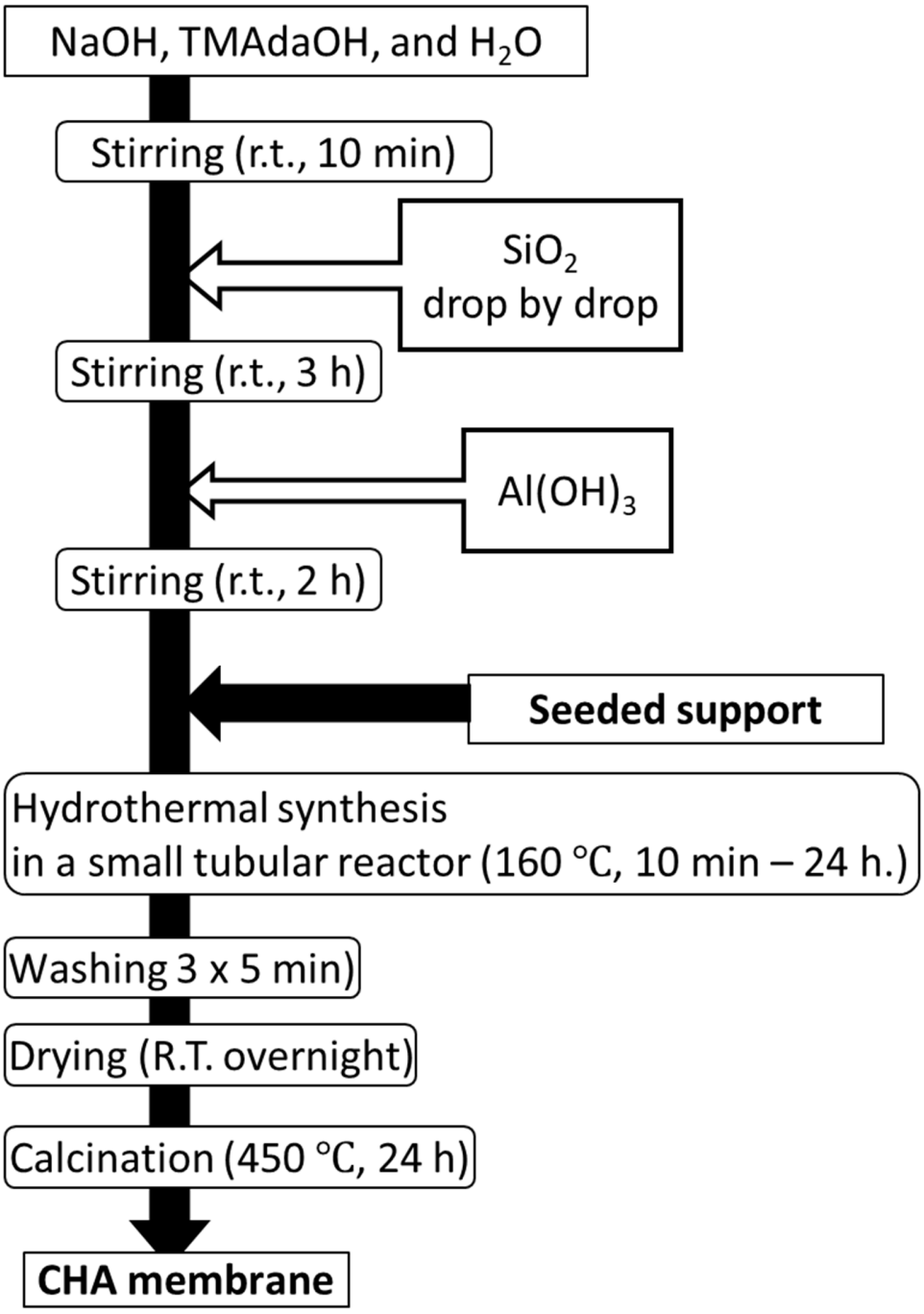


Figure S1. CHA membrane synthesis procedure.

2. CHA seed crystal

We successfully synthesized seed crystals of CHA zeolite, which were confirmed to exhibit a similar structural pattern to the IZA standard, the XRD patterns can be seen in Fig S.1, and the XRD patterns were compared to the reference standards for Al_2O_3 obtained from the Materials Project database (Material ID: mp-1143 for Al_2O_3). The crystals have an average size of approximately 1 µm, as shown in Fig. S2, as can be seen in the figure, and a Si/Al ratio of around 14.

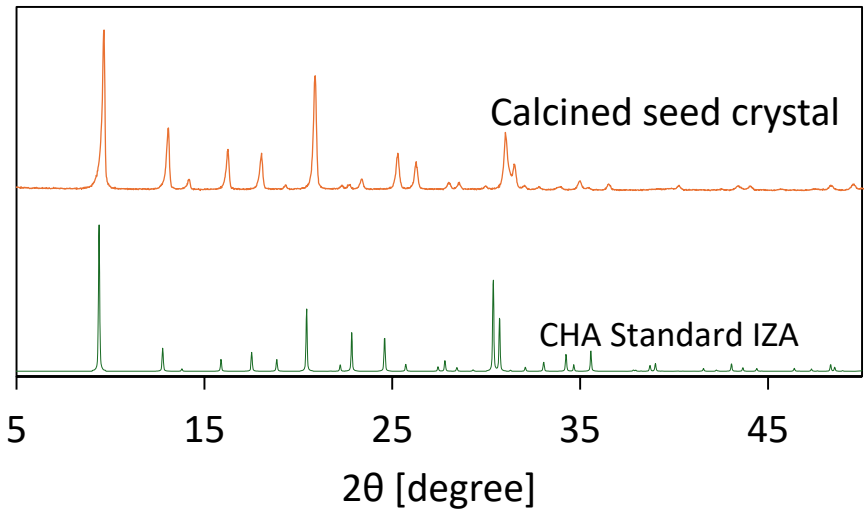


Figure S2. XRD pattern of CHA seed crystal compared to the standard from IZA.

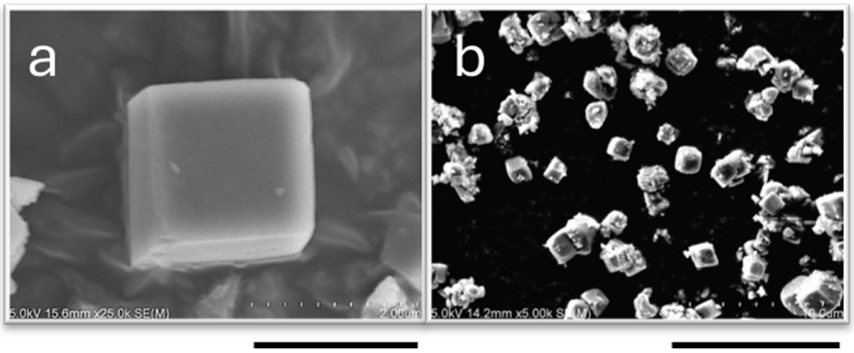


Figure S3. SEM image of CHA seed crystal, the scale bar represents (a) 2 µm and (b) 10 µm.

3. Effect of dip-coating solution

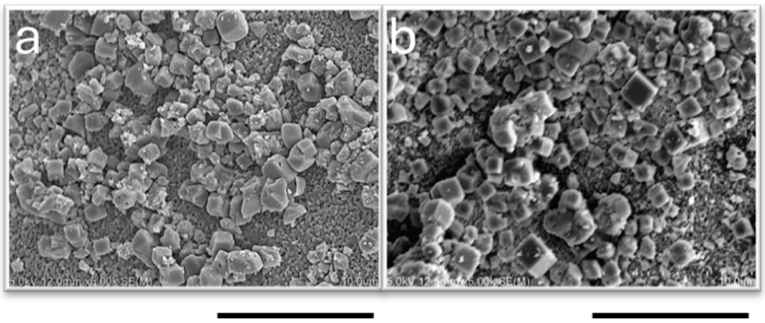


Figure S4. SEM images of seeded supports coated with solution 1 wt% of (a) pH 7 and (b) pH 8, (scale bar: 10 µm)

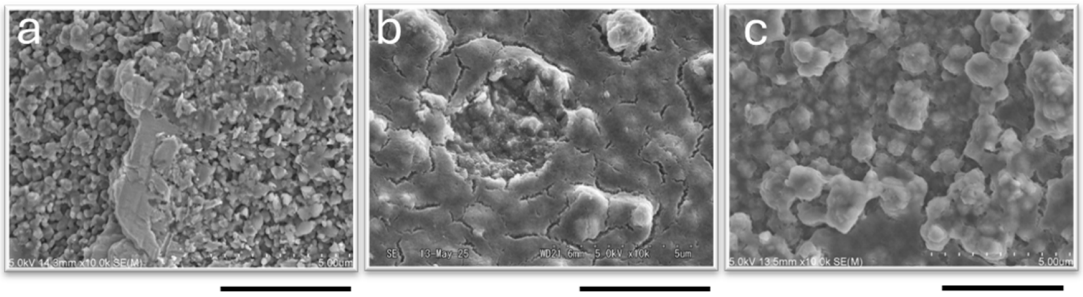


Figure S5. SEM image of the surface of membranes of 60 min (a) no seed, (b) 1 wt%-pH 7, and (c) 1 wt%-pH 8, the scale bar represents 5 µm.

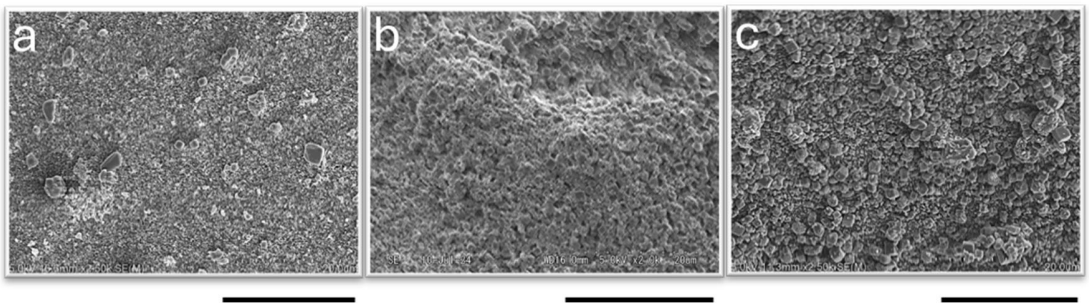


Figure S6. SEM image of the surface of seeded support coating using coating solutions at pH 8 with (a) 0.1 wt%, (b) 2 wt%, and (c) 4 wt% of seed crystal. Scale bar: 20 µm.

.

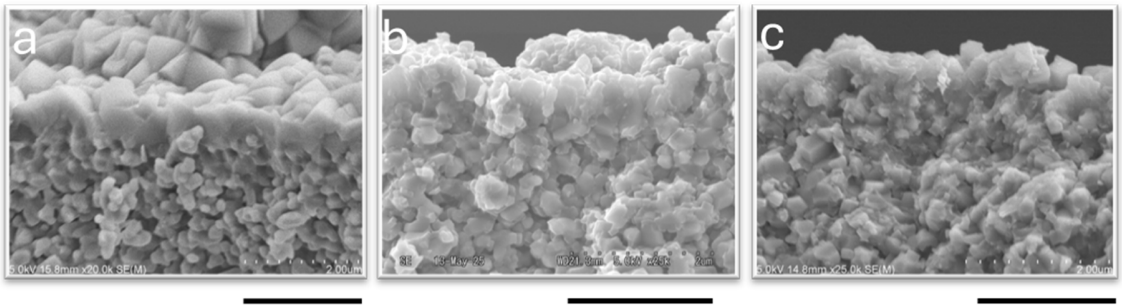


Figure S7. SEM images of the cross section of membranes synthesized for 70 minutes using coating solutions at pH 8 with (a) 0.1 wt%, (b) 2 wt%, and (c) 4 wt% of seed crystal. Scale bar: 2 µm.

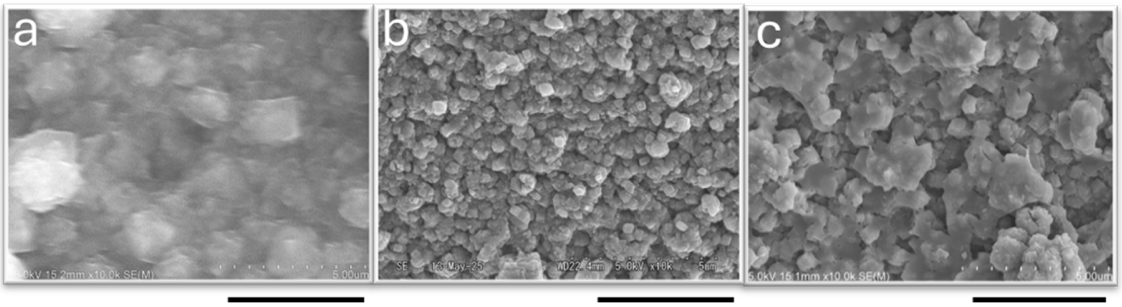


Figure S8. SEM images of the surface of membranes synthesized for 70 minutes using coating solutions at pH 8 with (a) 0.1 wt%, (b) 2 wt%, and (c) 4 wt% of seed crystal. Scale bar: 5 µm.

4. Effect of draying condition for seeded support

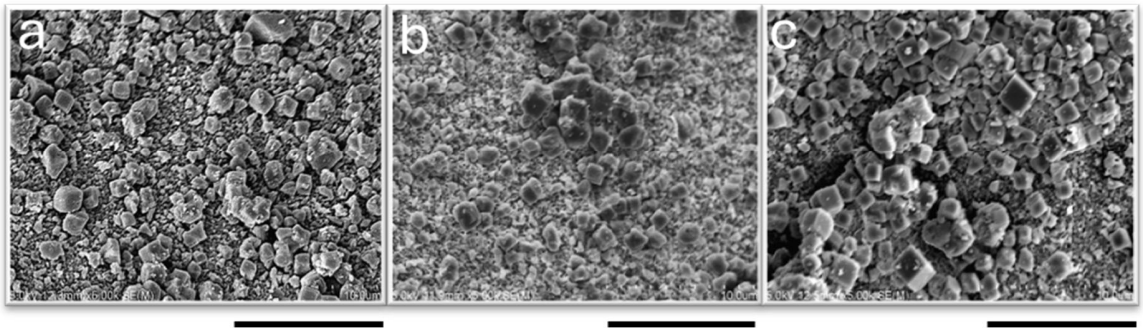


Figure S9. SEM image of the surfaces of seeded supports using a 1 wt% pH 8 seed solution dried at (a) R.T., (b) 200 °C, and (c) calcined at 450 °C, the scale bar represents 10 µm.

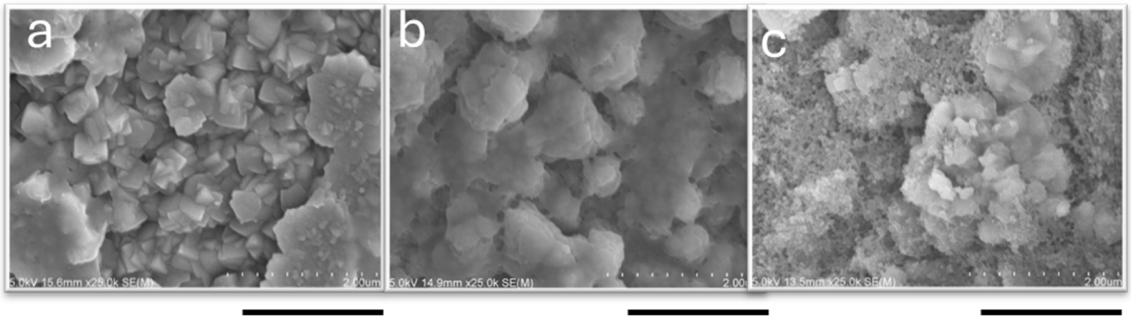


Figure S10. SEM image of the membrane surfaces synthesized for 60 min using 1 wt%-pH 8 seed solution dried at (a) R.T., (b) 200 °C, and (c) calcined at 450 °C, the scale bar represents 2 µm.

5. Effect of Effect of hydrothermal synthesis time

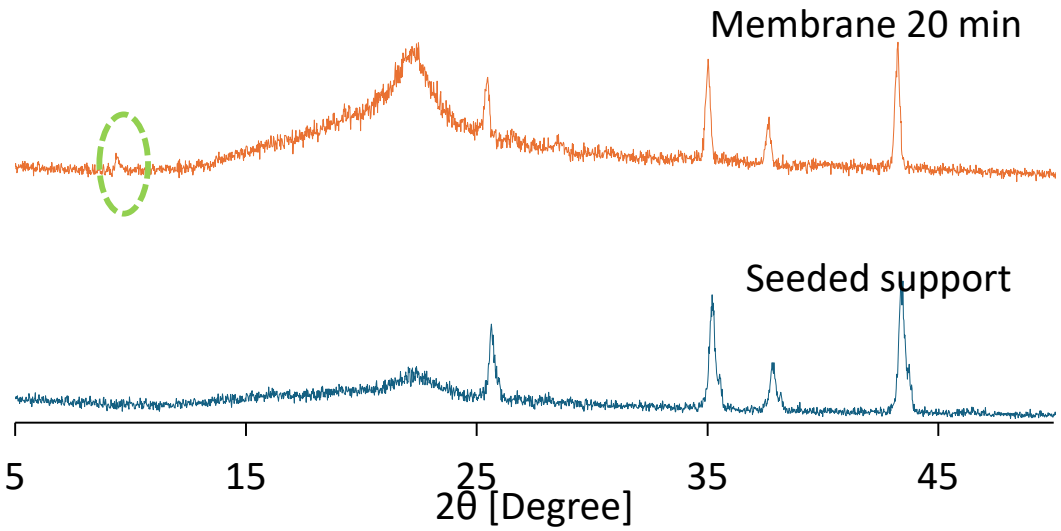


Figure S11. XRD patterns of seeded support and CHA membrane of 20 min.

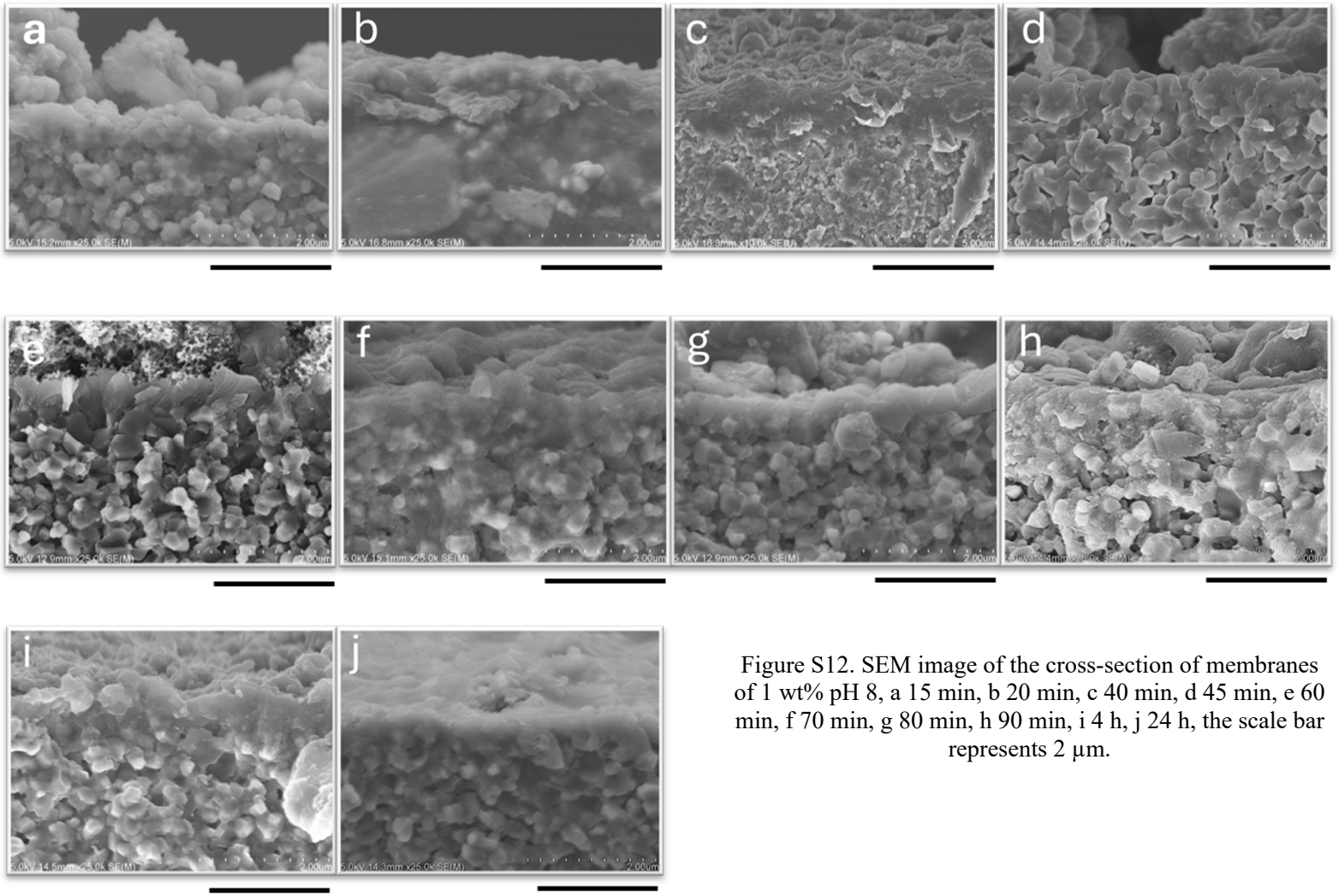


Figure S12. SEM image of the cross-section of membranes of 1 wt% pH 8, a 15 min, b 20 min, c 40 min, d 45 min, e 60 min, f 70 min, g 80 min, h 90 min, i 4 h, j 24 h, the scale bar represents 2 μm.

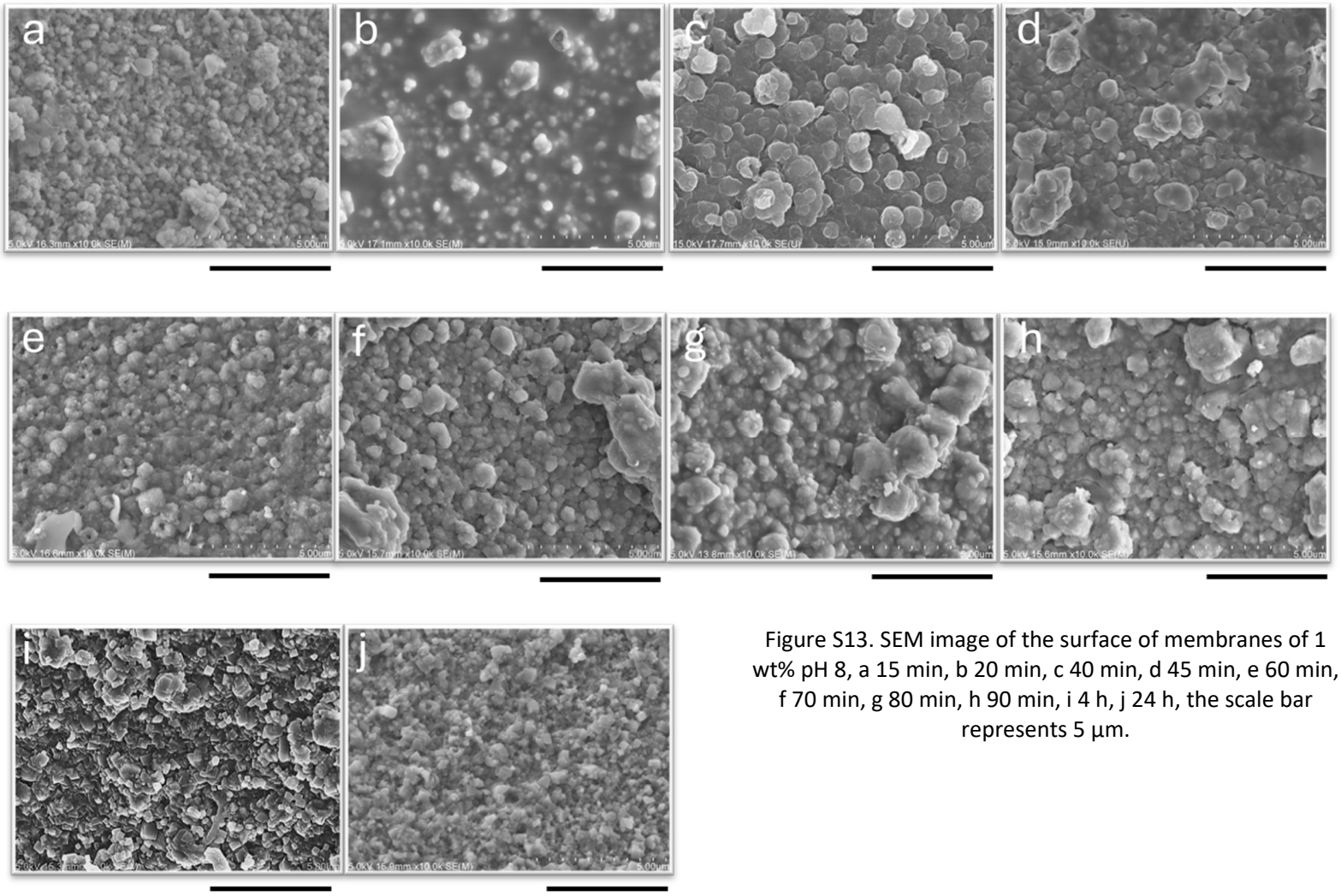
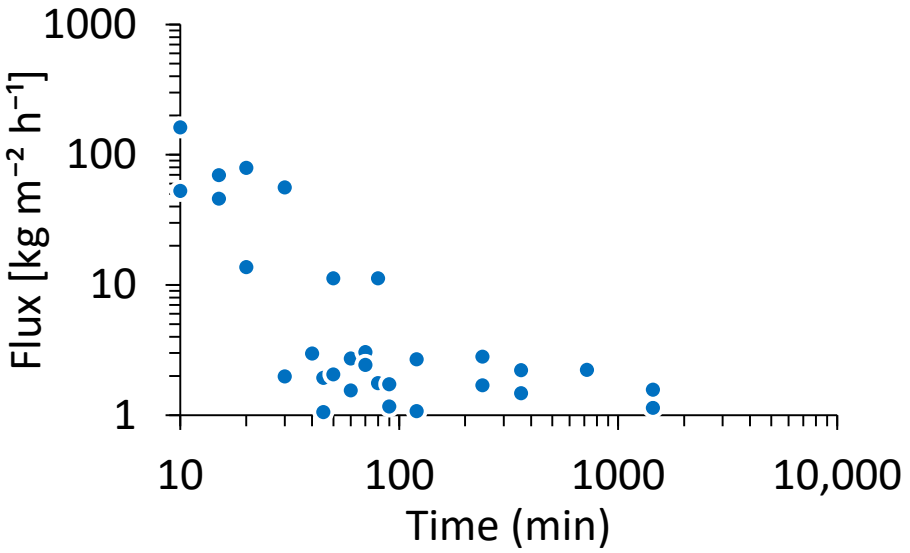
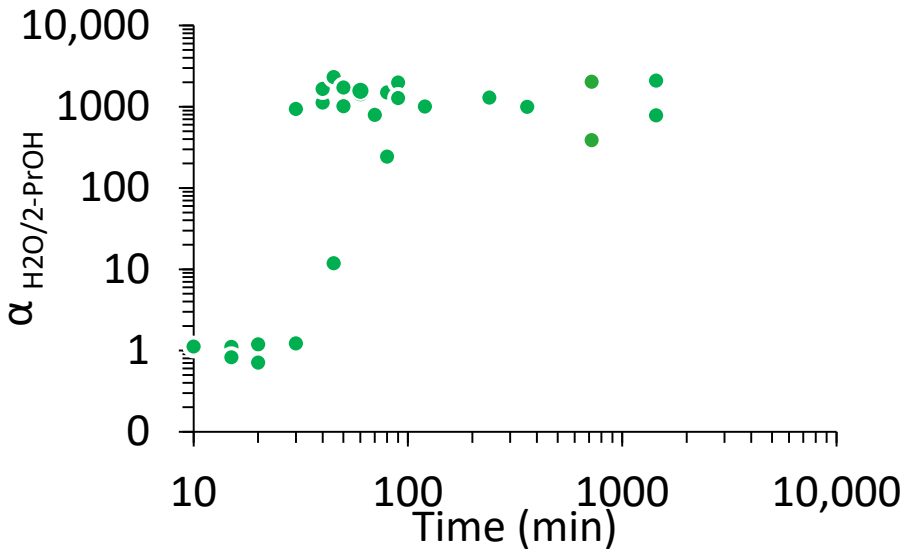


Figure S13. SEM image of the surface of membranes of 1 wt% pH 8, a 15 min, b 20 min, c 40 min, d 45 min, e 60 min, f 70 min, g 80 min, h 90 min, i 4 h, j 24 h, the scale bar represents 5 µm.

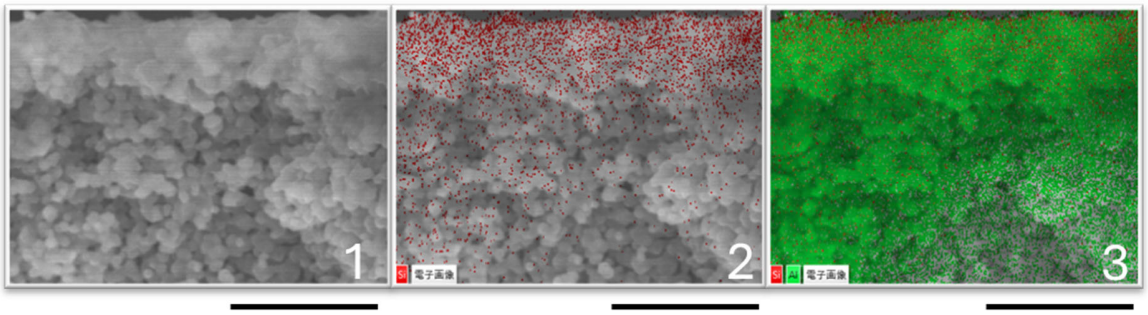


(a)

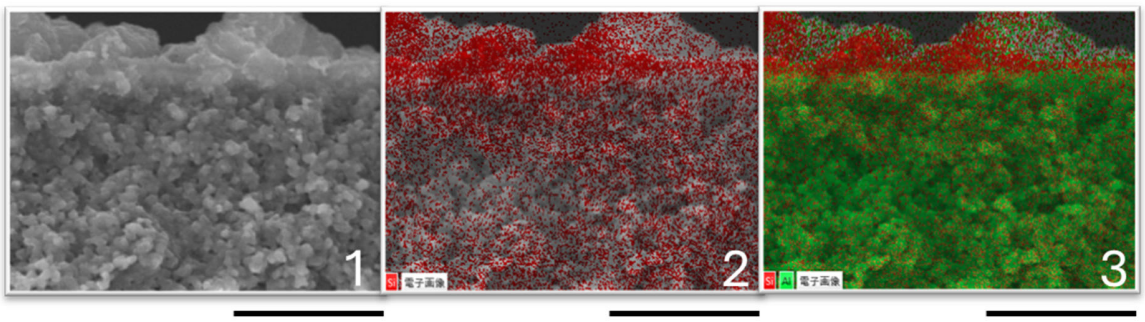


(b)

Figure S14. (a) Relation of total flux vs synthesis time, (b) Relation of separation factor vs synthesis time of the CHA membrane prepared with 1 wt% pH 8 dip-coating.



(a)



(b)

Figure S15. EDX-SEM cross-sectional analysis of membranes prepared of 1 wt% pH 8 for (a) 40 min and (b) 120 min. For each condition: (1) SEM micrograph, (2) elemental mapping of Si (red), and (3) combined elemental mapping of Si (red) and Al (green). The scale bar represents (a) 2.5 µm and (b) 5 µm.