# Supporting Information for

# Sharp Separation of C2/C3 Hydrocarbon Mixtures by Zeolitic Imidazolate Framework-8 (ZIF-8) Membranes Synthesized in Aqueous Solutions

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## Experimental Section

**Preparation of supports:** Porous α-alumina discs (2 mm thickness and 22 mm diameter) were used as supports, which were home-made from high purity α-alumina powder (Baikowski, CR-6). The discs were made by pressing α-alumina powder in a custom-made mold with the aid of a hydraulic press (Carver) and the resulting compacts were sintered at 1080 $^{o}$C for 30 h and then at 1190 $^{o}$C for 3 h. The nominal pore size of the support is around 200 nm and the porosity is around 40%. One side of the support was polished with sandpaper (Buehler, 1200 grit) to form a smooth surface for applying the ZIF-8 seed layers as described in the next section.

**Preparation of ZIF-8 seeds:** All chemicals were purchased from Sigma-Aldrich and used as supplied. 1.17 g (3.7 mmol) of $Zn(NO_3)_2$•$6H_2O$ and 22.7 g (276.5 mmol) of 2-methylimidazole were dissolved in 88 mL deionized (DI) water. The resulting mixture was stirred at room temperature for 12 h. The obtained white powder was washed with DI water and methanol for three times, respectively, and then re-dispersed in methanol to form a stable seed suspension (0.1 wt%) that will be used in the next section.

**Preparation of seed layers on porous α-alumina supports:** Seed layers were prepared by a simple slip-coating process which is described as the following. Firstly, the seed suspension was poured in a petri-dish placed on a height-adjustable jack and then slowly raised towards the polished surface of an inverted support disc held above, until the support soaked the suspension. The support was left for about 20 s in contact with the suspension and was subsequently removed tangentially and dried for 24 h at room temperature.

**Preparation of ZIF-8 membranes by secondary growth:** The secondary growth solution was prepared by dissolving 0.11 g (0.37 mmol) $Zn(NO_3)_2$•$6H_2O$ and 2.27 g (27.65 mmol) 2-methylimidazole into 40 mL DI water. A seeded support was first placed vertically in a Teflon cup. Then about 40 mL growth solution was poured to the cup until the seeded support completely immersed into the solution. The cup was covered and then transferred to an oven that was preheated to 30 $^{o}$C and stayed for 6 h. After the synthesis, the disc was taken out, rinsed with DI water and

methanol for three times, and then immersed in 30 mL fresh methanol for another 12 h. Finally, the disc was dried in ambient condition for 24 h before performing the permeation test.

**Characterizations:** X-ray diffraction (XRD) patterns were measured from a Bruker D8 Advance X-ray diffractometer with Cu Kα radiation. Field-emission scanning electron microscope (SEM) pictures were taken by a FEI Quanta 600 FEG, and the acceleration voltage was 30 kV. The mean particle size of the seeds was determined by manual measurement of about 300 crystals in SEM pictures. Nitrogen physisorption isotherm was measured at 77 K on an automatic volumetric adsorption apparatus (Micromertics ASAP 2420). The sample was filled into glass ampoules and outgassed in high vacuum at 473 K for 24 h before the sorption measurements. Thermal gravimetric analysis (TGA) was carried out on a Netzsch 449 thermoanalyzer. For this purpose, ca. 10 mg of sample was filled into alumina crucible and heated under an air flow from room temperature to 800 $^{o}$C at a ramp rate of 10 $^{o}$C/min.

**Gas permeation test:** All permeation tests were performed by the Wicke-Kallenbach (W-K) technique. Argon was used as the sweep gas at the permeate side. The permeate stream was analyzed on-line by a gas chromatography (Agilent 7890A). For single-gas measurements, the flow rates of the feed flow and the sweep flow were set to 10 and 100 mL/min, respectively. The pressure in the feed side was maintained at 1 atm. For the binary-mixture measurements the feed flow rate of each gas was kept at 10 mL/min. The two gases were well mixed at a mixer before entering to the feed side of the permeation cell. The flow rate of argon sweep gas was kept constant at 100 mL/min. The total pressure at the feed side and at the permeate side both were kept constant at 1 atm. The reported permeance and selectivity are average values recorded from at least three samples which were made from different batches.

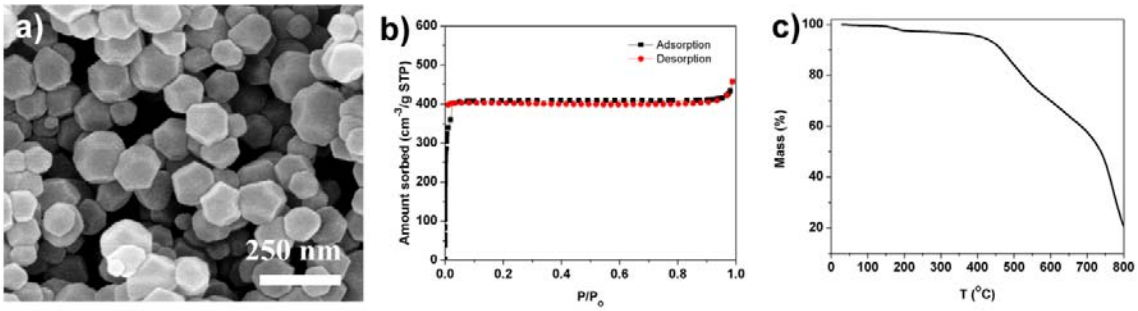


**Fig. S1** (a) SEM, (b) $N_2$ sorption isotherm and (c) Thermal gravimetric analysis (TGA) curve conducted at air of the as-synthesized ZIF-8 seeds.

Fig. S1 shows the SEM image, the $N_2$ physisorption isotherm at 77K, and the thermal gravimetric analysis of the seeds. The SEM image shows that the seed particles have a polyhedral shape with the mean particle size around 110 nm. The $N_2$ physisorption measurement showed a type I adsorption isotherm. The calculated BET surface area and the microporous volume are around 1250 $m^2$/g and 0.7 $cm^3$/g, which are close to the values reported in the

literature (Cravillon et al., *Chem Mater*, **2009**, *21*, 1410; Nune et al., *Chem Commun*, **2010**, *46*, 4878). The TGA curve performed under air flow shows the release of the guest molecules from the cavities or unreacted species from the surfaces of the seeds is occurred below 200 $^{o}$C (ca. 3%). The framework decomposes in the temperature range from 300 to 400 $^{o}$C. This result indicates that ZIF-8 seeds have high thermal stability.

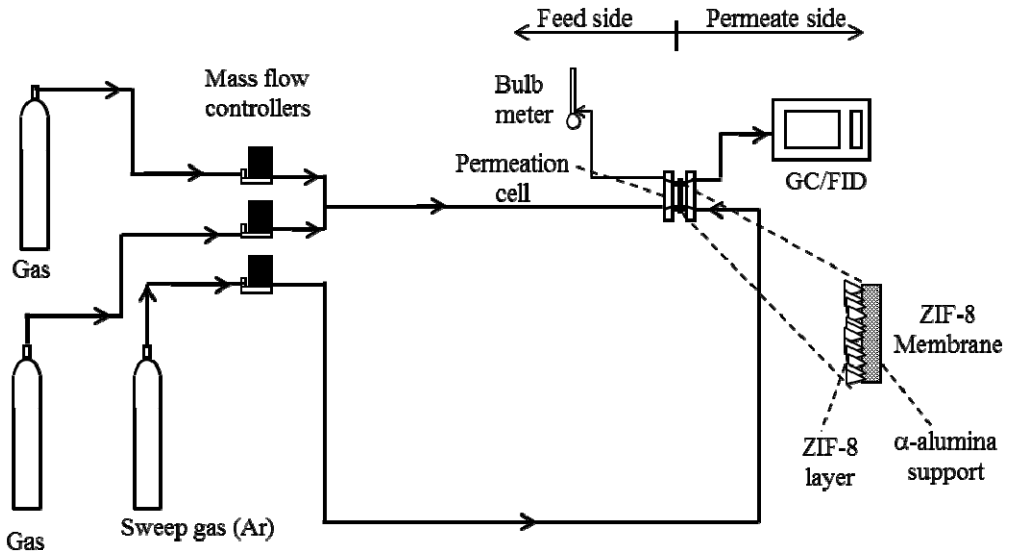


**Fig. S2** Permeation setup in Wicke-Kallenbach mode