S.1. Gas permeation experiments

A variable-pressure constant-volume method was employed to determine the gas permeation properties of Matrimid/NaY MMMs. The schematic of experimental set-up depicts in Figure S1. All the membrane samples were tested at operating temperature range of 35-75 °C and the test pressure in the range of 2-12 bar. At the steady state and under isothermal conditions, the pressure increase of accumulated permeating gases in the constant volume chamber was measured by an absolute pressure transmitter (type 691, Huba Control, Würenlos, Switzerland) and recorded respect to the time ($\frac{dp}{dt}$) and then the gases permeability were calculated using the following equation:

$$P = \frac{273.15 \times 10^{10} Vl}{760AT((P_0 \times 76)/14.7)}\left(\frac{dp}{dt}\right) \quad (1)$$

where P is the gas permeability across the membrane in Barrer (1 Barrer = 1×10^{-10} cm^3 (STP) cm/cm^2 s cmHg), V is the dead-volume of the downstream chamber (cm^3), l is the membrane thickness (cm) and was about 45-55 µm, T is the experimental temperature (K), A is the effective membrane area (cm^2) with a value of 11.34 cm^2, P_0 is the upstream feed gas pressure (psia) and finally as mentioned before ($\frac{dp}{dt}$) is the steady rate of pressure change gradient in the downstream side (mmHg/s). It should be noted that to ensure accuracy of the data acquisitions, each measurement was replicated on three different membrane samples with the same composition and the reported value was the arithmetic mean for the three samples.

Besides, the ideal selectivity of two pure components was calculated by dividing the respected permeabilities in the same conditions:

$$\alpha = \frac{P_A}{P_B} \tag{2}$$

where P_A and P_B are the permeability of pure A and B gases, respectively.

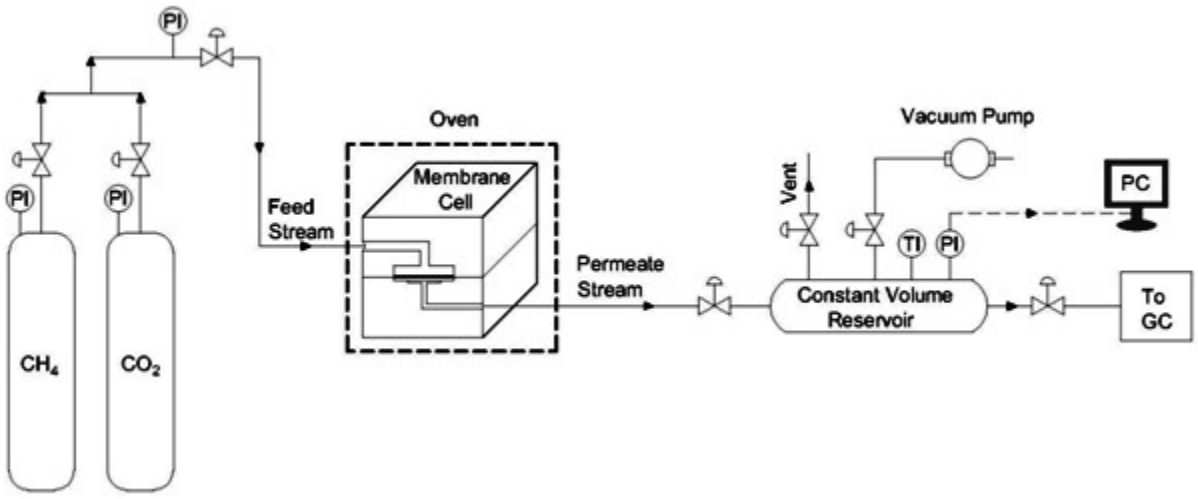

Figure S1. Schematic diagram of the experimental set-up.

S.2. Mixed gas permeation

A binary CO_2/CH_4 mixture containing 10% of CO_2 and 90% CH_4 (volumetric basis) was considered as a feed gas and permeability measurement of each gas conducted at 35 °C and 2 bar using the following equations:

$$P_{CO_2} = \frac{273.15 \times 10^{10} y_{CO_2} Vl}{760AT\left[x_{CO_2}(P_0 \times 76)/14.7\right]}\left(\frac{dp}{dt}\right) \tag{3}$$

$$P_{CH_4} = \frac{273.15 \times 10^{10}\left(1 - y_{CO_2}\right)Vl}{760AT\left[\left(1 - x_{CO_2}\right)(P_0 \times 76)/14.7\right]}\left(\frac{dp}{dt}\right) \tag{4}$$

where P_{CO_2} and P_{CH_4} are the permeability of gases across the membrane in Barrer, V is the dead-volume of the downstream chamber (cm^3), l is the membrane thickness (cm), T is the experimental temperature (K), A is the effective membrane area (cm^2), P_0 is the upstream feed gas pressure (psia) and $\frac{dp}{dt}$ is the steady rate of pressure change gradient in the downstream side (mmHg/s). The gas mixture selectivity was calculated by the following equation:

$$\alpha_{A/B} = \frac{y_A / y_B}{x_A / x_B} \quad (5)$$

where x and y are the mole fractions in the feed/permeate side, respectively. The composition of permeate streams was determined using a gas chromatograph instrument (GC model Agilent 7890A, Agilent Technologies Co., USA) equipped with a thermal conductivity detector (TCD) and a Q capillary column.

S.3. Membrane characterization

S.3.1. XRD

The XRD patterns of the zeolite and membranes were analyzed on a X'Pert MPD wide-angle X-ray diffractometer from Philips, The Netherlands. The measurements were carried out at room temperature using mono-chromatic radiation of α-rays emitted by Cu at a wave-length of 1.54 Å, accelerating voltage of 40 kV, and tube current of 40 mA. To identify the crystal structure, the scan range - the angle (2θ) of diffraction - was varied from 3 to 70° with a step increment of 0.02° s^{-1}. XRD was performed, in order to analyze the structural properties and quantitatively measure the inter-chain spacing of MMMs (d-spacing) at room temperature. This measurement was accomplished by using Cu Ka radiation in a scan range from 3º to 70º with a step increment

of 0.02°/s. it must be noted that the wavelength of λ= 1.54 Å was used in the experiments. Considering the Bragg's law, average d-spacing of the membranes was calculated:

$$n\lambda = 2\mathrm{d}\sin\theta \quad (6)$$

where n is an integral number (1, 2, 3, . . .), λ denotes the X-ray wavelength, d represents the dimension spacing, and θ is the diffraction angle.

S.3.2. FTIR-ATR

FTIR-ATR investigations were done on the zeolite and membranes using a Perkin-Elmer Spectrum, Frontier model, Version 10.03.06 (Perkin-Elmer Instruments, Norwalk, USA) in the range of 400-4000 cm^{-1}. The spectrum of each specimen was taken at an incidence angle of 458 with 32 scans at a wave number resolution of 4 cm^{-1}.

S.3.3. SEM

The morphology and compositional study of the zeolite and membranes were carried out by SEM. After sputter-coating with gold by a BAL-TEC SCD 005 sputter coater (BAL-TEC AG, Balzers, Liechtenstein), the samples were tested by a SEM (KYKY-EM3200, KYKY Technology Development Ltd., Beijing, China). In the case of cross sectional observation of the membranes, the samples were fractured in liquid nitrogen.

S.3.4. TG/DTG

Thermal analyses of the membranes were carried out in order to evaluate the temperature of possible decompositions and phase changes. The TG measurements were carried out using thermal gravimetric and derivative thermal gravimetric analysis (TG-DTG, STA BAHR 503,

Germany). TG runs were recorded under dried, ultra high pure argon gas flow from 0 to 700 ºC at a heating rate of 10 ºC/min.