

## Supplementary data:

### Gas permeation experiment

Before each measurement, all lines were initially vacuumed via a pump, and then the target gas was introduced to the membrane cell. The permeate gas was accumulated in a downstream constant volume chamber, and the rate pressure increment in the chamber versus the time was recorded via a pressure sensor with an accuracy of 1 mbar. The pressure gradient was found and the corresponding gas permeability was calculated by the following equation:

$$P = \frac{273.15 \times 10^{10} \nu l}{AT(p_0 \times 76)} \left( \frac{dp}{dt} \right) \quad (1)$$

where  $P$  is the gas permeability presented in Barrer (1 Barrer =  $10^{-10} \text{ cm}^3 \text{ (STP). cm. cm}^{-2} \text{. s}^{-1} \text{. cmHg}^{-1}$ ),  $\nu$  is the constant volume of the downstream vessel of the membrane holder ( $\text{cm}^3$ ),  $l$  is the membrane thickness (cm),  $A$  refers to the effective surface area of the membrane ( $\text{cm}^2$ ),  $T$  is the operating temperature (K),  $p_0$  is the feed gas pressure (atm), and  $(dp/dt)$  is the slope of pressure changes versus time ( $\text{atm. s}^{-1}$ ).

The ideal permselectivity of  $i$  gas than  $j$  gas ( $\alpha_{ij}$ ) was computed by dividing the gas permeability as follows:

$$\alpha_{ij} = \frac{P_i}{P_j} \quad (2)$$

The permeate gas for mixed gas mode was analyzed using a gas chromatography apparatus (GC-2552 TG, Iran) equipped with a thermal conductivity detector (TCD). The permeability was also measured by the following equations:

$$P_i = \frac{273.15 \times 10^{10} \times y_i \nu l}{AT(x_i \times p_0 \times 76)} \left( \frac{dp}{dt} \right) \quad (3)$$

where  $i$  is the desired gas composition in the gas mixture stream.

The gas mixture selectivity was appraised by the following equation:

$$\alpha_{ij} = \frac{y_i/y_j}{x_i/x_j} \quad (4)$$

where  $y$  and  $x$  are the volumetric fractions of the permeate and feed gas, respectively.

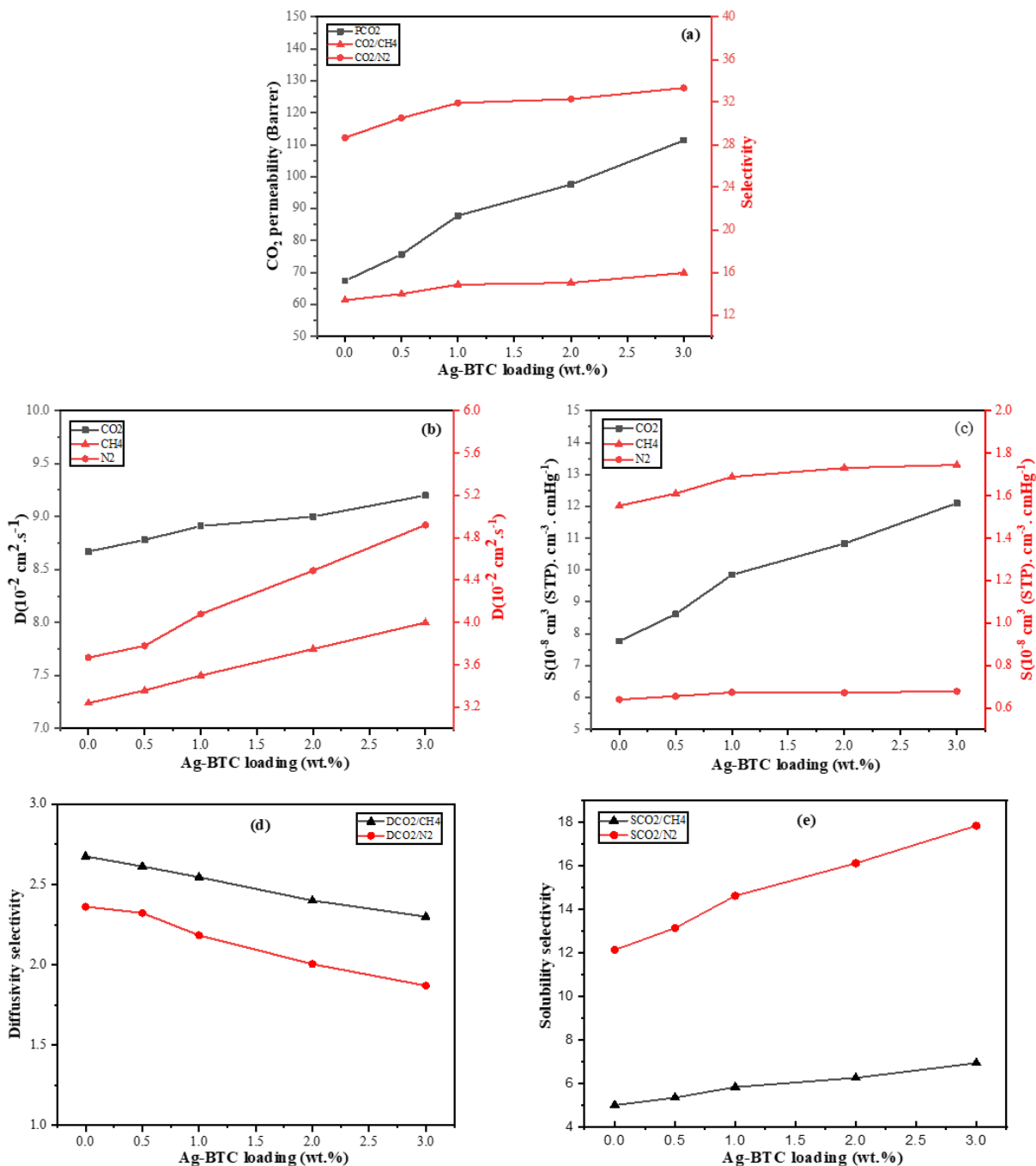
### Solubility and diffusivity analysis

Mixed matrix membranes with identical polymer concentrations with TFC/TFN membranes were prepared to determine solubility coefficients. After the calculation of permeability coefficients ( $P$ ) by using eq. S1, the time lag method was used in order to determine the diffusion coefficients ( $D$ ) as follows:

$$D = \frac{l^2}{6\theta} \quad (5)$$

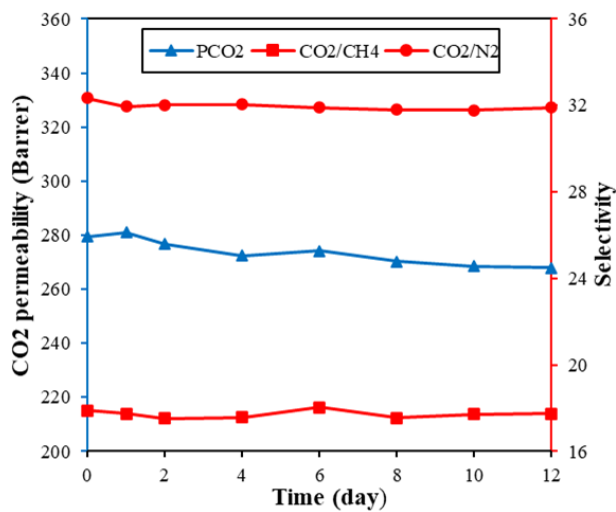
where  $l$  is the thickness of the membranes and  $\theta$  is the time lag (delay time) (s) evaluated from the pressure-time curve. The solution-diffusion mechanism is considered to be valid in gas transport through self-standing membranes. According to this model, the permeability of each gas through the polymeric membrane is the multiplication of the gas diffusivity ( $10^{-2} \text{ cm}^2 \cdot \text{s}^{-1}$ ) and solubility ( $10^{-8} \text{ cm}^3 \text{ (STP)} \cdot \text{cm}^{-3} \cdot \text{cmHg}^{-1}$ ). Thus, the solubility coefficient ( $S$ ) can be determined as follows:

$$S = P/D \quad (6)$$



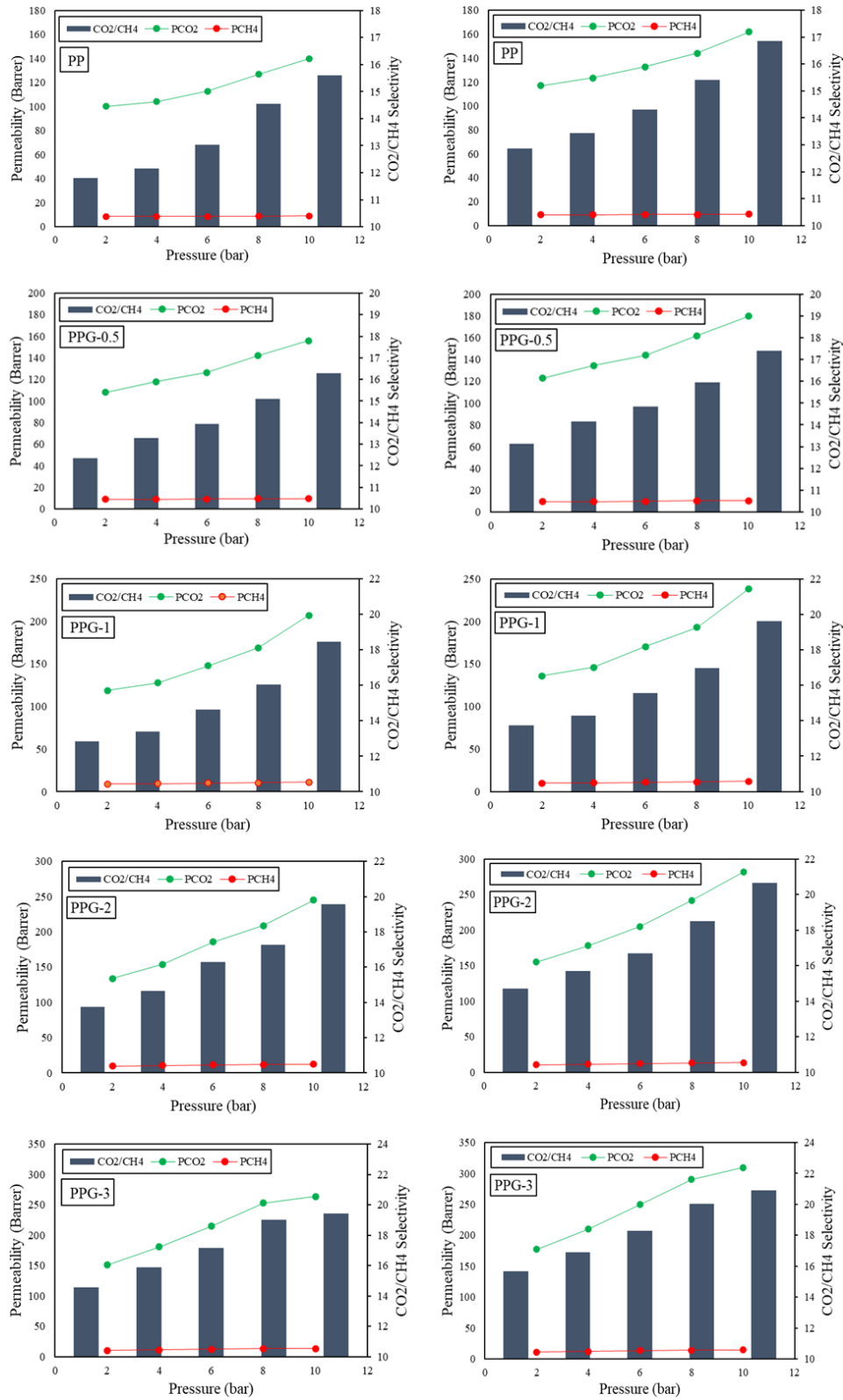
**Fig. S1.** (a) CO<sub>2</sub> permeability, (b) diffusivity coefficients, (c) solubility coefficients, (d) diffusivity selectivity, and (e) solubility selectivity of CO<sub>2</sub>/CH<sub>4</sub> and CO<sub>2</sub>/N<sub>2</sub> of Pebax1657 and Pebax1657/Ag-BTC MMMs. The experiment was conducted at 4 bar and 30 °C.

## Long-term stability

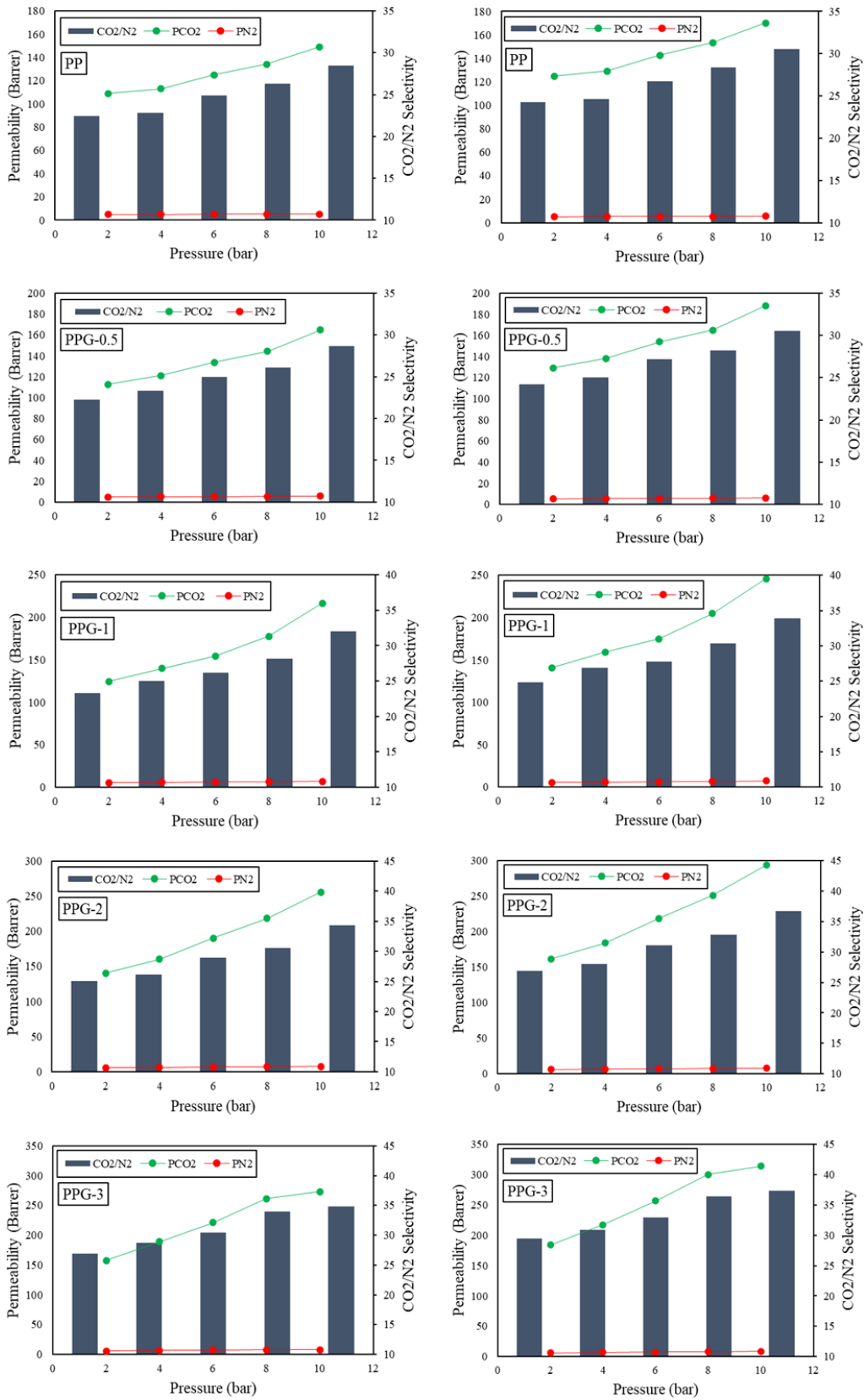


**Fig. S2.** The long-term stability of the PPG-2 film at 6 bar and 30 °C. The experiment was conducted at intervals of 2 days, for 12 days.

## Dry/humid-mixed gas permeation



**Fig. S3.** The effect of pressure on the CO<sub>2</sub>/CH<sub>4</sub> separation performance of the neat TFC and TFN membranes at 30 °C and 6 bar. Dry-mixed gas (left) and humid-mixed gas (right).



**Fig. S4.** The effect of pressure on the CO<sub>2</sub>/N<sub>2</sub> separation performance of the neat TFC and TFN membranes at 30 °C and 6 bar. Dry-mixed gas (left) and humid-mixed gas (right).