

Supplementary information for:

Multi-lab study on the pure-gas permeation of commercial polysulfone (PSf)

membranes: measurement standards and best practices

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Table of Contents

1. Normality tests, Z-scores, and fitting deviations	3
3. Sample preparation	6
4. Equipment information	7
5. Testing conditions	9
6. Literature comparison	12
Appendix 1. Word and Excel templates for submissions	14
1.1. Word reporting template	14
1.2. Excel file submission template	17
Appendix 2. Analysis of co-dependencies for H₂ and CH₄	18
Appendix 3. Raw data for time lag calculations	20
Appendix 4. Detailed experimental procedure for each lab	24
Lab 1	24
Lab 2	25
Lab 3	28
Lab 4	29
Lab 5	30
Lab 6	32
Lab 7	35
Lab 8	36
Lab 9	38
Lab 10	39

1. Normality tests, Z-scores, and fitting deviations

Table S1. P-value for the Shapiro-Wilk normality test.

Gas	Pressure (bar)		
	10	15	20
H ₂	0.0432	0.0606	0.2476
O ₂	0.1246	0.1478	0.1242
CH ₄	0.1168	0.3624	0.1889
N ₂	0.3368	0.2945	0.5064

The null hypothesis of the Shapiro-Wilk test is that the average permeability from replicates reported by each lab follows a normal distribution. The null hypothesis is rejected when the p-value is less than 0.05. In this case, there is less than 5% probability that the null hypothesis is true. Therefore, the H₂ permeability data at 10 bar does not follow a normal distribution, but all other distributions can be considered normal.

Table S2. Z-scores for as-submitted permeabilities.

Z-score for permeability												
Gas	H ₂			N ₂			O ₂			CH ₄		
Pressure (bar)	10	15	20	10	15	20	10	15	20	10	15	20
Lab 1	-1.25	-0.76	-0.87	-0.61	-0.42	-0.26	-	-	-	-0.72	-0.72	-0.53
Lab 2	1.62	1.93	1.81	1.49	1.72	1.64	1.69	1.76	1.65	2.37	1.98	1.95
Lab 3	1.82	1.58	1.16	1.72	1.06	1.37	1.36	0.98	0.79	1.53	1.10	0.73
Lab 4	-0.11	0.03	0.04	-0.03	0.74	0.08	-0.13	-0.45	-0.40	0.49	0.01	0.38
Lab 5	-1.10	-1.26	-1.10	-1.14	-1.30	-1.14	-1.15	-	-	-1.24	-1.14	-1.14
Lab 6	-0.09	-0.03	-	0.84	0.79	-	-0.35	-0.10	-	1.49	0.83	-
Lab 7	-0.09	-0.40	-0.40	-0.51	-0.57	-0.37	-0.49	-0.79	-0.78	-0.17	-0.33	-0.11
Lab 8	-0.26	-0.25	-0.21	-0.48	-0.66	-0.50	-0.20	-0.47	-0.42	-0.29	-0.49	-0.42
Lab 9	-0.12	-0.32	-	-0.34	-0.38	-	-	-	-	-0.08	-0.29	-
Lab 10	-0.42	-0.53	-0.44	-0.92	-0.98	-0.83	-0.73	-0.93	-0.84	-1.01	-0.96	-0.86

Table S3. Fitting parameters for H₂, O₂, N₂, and CH₄ permeability versus upstream pressure plots shown in **Figure 5**.

Gas	x	y	z
H ₂	14.4	0.00000402	4660
O ₂	0.792	330	0.00201
CH ₄	0.178	2.55	0.0609
N ₂	0.240	0.228	5000

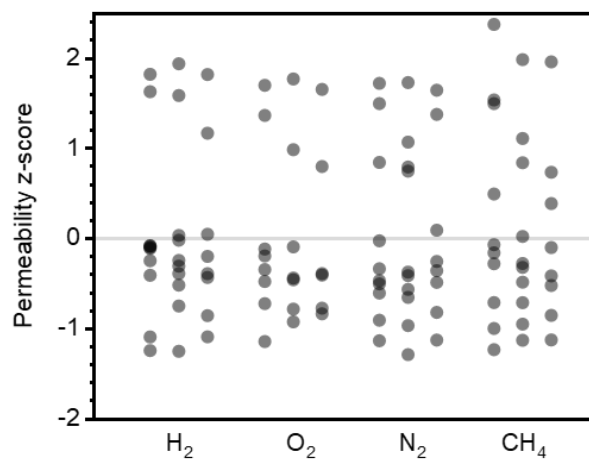


Figure S1. Z-scores for as-submitted permeabilities.

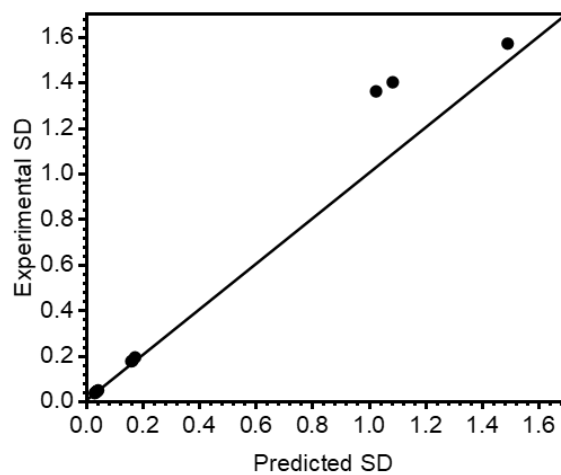


Figure S2. Experimental versus predicted standard deviations from the model fittings. Experimental standard deviations were collected from box and whisker plots and predicted standard deviations were collected from the model fitting shown in **Figure 5**.

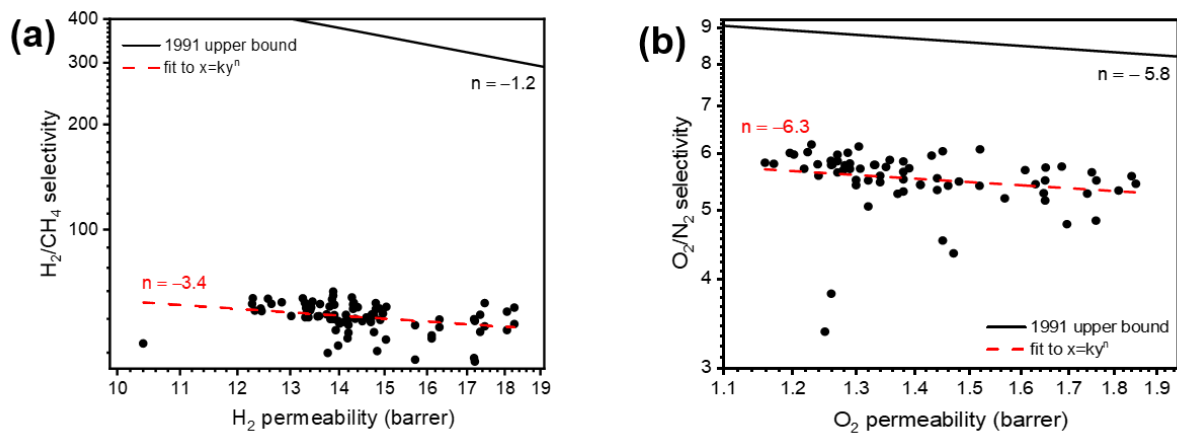


Figure S3. O₂/N₂ and H₂/CH₄ upper bounds for as-submitted permeability data.

3. Sample preparation

Table S4. Sample preparation conditions.

Lab	Sample preparation					
	Sample support	Adhesive material	Micrometer	Storage conditions	Area measurement	Area (cm ²)
1	copper tape	1 part DEGBA based epoxy and 2 parts reactive polyimide	Mitutoyo (Series 239)	lab bench	ImageJ	6.347 ± 0.001
						6.451 ± 0.030
						4.265 ± 0.008
2	stainless steel disk	aluminum tape	Guanglu (110-221FG)	vacuum dessicator	ruler	3.137 ± 0.0002
						2.926 ± 0.0002
						3.079 ± 0.0002
3	porous iron disk covered with polyethylene non-woven	Viton O-ring for sealing	Mitutoyo (Series 293-240)	dry, room temperature	ruler	1.91
4	aluminum tape	Devcon 5-min epoxy (20945)	Mitutoyo (Code No. 547-401)	35 °C in a convection oven	Use 25 mm round punch for making effective area in the aluminum cell	4.909
5	aluminum tape	Devcon 10-min epoxy (14255)	Mitutoyo (293-344-30)	epoxied samples stored in vacuum desiccator	ImageJ	1.877 ± 0.01
						1.999 ± 0.01
						1.95 ± 0.01
6	brass disk	Devcon 5-min epoxy (14240)	Mitutoyo (Series 293)	stored as received in original packing.	ImageJ	0.64 ± 0.03
						0.65 ± 0.04
						0.61 ± 0.01
7	aluminum tape (3M 438)	Acrylic adhesive from tape	Farnell (439-5578)	stored in a plastic bag in air	ImageJ	1.605 ± 0.008
						1.745 ± 0.003
						1.708 ± 0.005
8	brass disk	Devcon 5-min epoxy (14240)	Mitutoyo (C112EB)	cool, dry drawer	ImageJ	0.8323 ± 0.003
						1.0452 ± 0.0006
						1.0826 ± 0.003
9	copper shim	5 min-epoxy (300007-392)	Mitutoyo (293-340-30)	dry condition at room temperature	ImageJ	0.676 ± 0.002
						0.917 ± 0.003
						0.776 ± 0.004
						0.670 ± 0.004
10	brass disk	Devcon 5-min epoxy (14250)	Mitutoyo (293-340-30)	lab drawer in ambient conditions	ImageJ	0.6962 ± 0.0009
						0.730 ± 0.006
						0.606 ± 0.004

4. Equipment information

Table S5. Equipment information.

Lab	Equipment information			
	Gas purity and provider	Upstream pressure transducer	Downstream pressure transducer	Temperature control
1	Praxair: H ₂ (99.999%), O ₂ (99.993%), N ₂ (>99.9999%), CH ₄ (99.97%)	Wika (S-20, 0-50 psig, 0.125% of span)	MKS Baratron (627F12TBC1B, 0.12% rd.)	water bath with circulator (ThermoFisher Neslab RTE 7)
2	Dalian Datai Gas Co. LTD: H ₂ (99.999%), N ₂ (99.9993%), O ₂ (99.995%), CH ₄ (99.9%)	CEMPX-4 (0-3000 KPa, 0.2% FS)	Inficon AG, FL-9496 Balzers (CDG025, 0.2% FS)	air bath
3	Air Liquide: H ₂ (>99.999%), O ₂ (>99.995%), N ₂ (>99.999%), CH ₄ (>99.9995%)	Swagelok Stainless Steel Back-pressure regulator (KBP1J0G4D5A200B0)	MKS Baratron (627F11TBC1B, 0.12% rd.)	electrical resistance wire at the bottom of the cell
4	Air Korea: H ₂ (99.999%), O ₂ (UHP), N ₂ (>99.999%), CH ₄ (>99.95%)	Digital pressure gauge (SBSA0010MCG, SZUN60, 0-10 MPag, 0.5% FS)	MKS Baratron (626B11TBE, 1-1000 Torr (0.25%), < 1 Torr (0.5%))	air bath
5	Matheson: H ₂ (99.999%); Airgas: O ₂ (99.999%), N ₂ (99.999%), CH ₄ (99.999%)	Baratron MKS (728A52TCEFA, 0.5% Rd.)	MKS Baratron (750C33PCD3GA, 1% rd.)	electric fiberglass heating jacket (A510-UCAL1016-21) with ambient gas/tubing
6	Airgas: >99.9%	Honeywell STJE (AP112CV, 0.05% FS)	MKS Baratron (626C11TBE, 0.25% rd.)	water bath with circulator (Thermo Scientific SC150, 115VAC)
7	BOC UK: H ₂ (Zero Grade, 270065-F, 99.995%), O ₂ (N5.5, 284916-L, 99.9995%), N ₂ (Research Grade, 293679-L, 99.9995%), CH ₄ (CP grade, 291372-L, 99.5%)	Keller digital manometer (LEO2EI-1-30BAR, 0-31 bar, 0.1% FS)	Keller (P-AA-33X-10-10V-DIN, 0-10bar abs, 0.05% FS)	air bath
8	Airgas: UHP 99.999%	Honeywell STJE (060-1885-19, 0.05% FS)	MKS Baratron (626C12TBE, 0.25% rd.)	water bath with circulator (ThermoFisher ARTIC A10 153-5108)
9	Airgas: 99.98%	McMaster-Carr digital pressure gauge (3834K111, 0-300 psi, 1% FS)	MKS Baratron (627F13TBC1B, 0.12% rd.)	air bath
10	Airgas UHP300: H ₂ (99.999%), O ₂ (99.994%), N ₂ (99.999%), CH ₄ (99.99%)	Honeywell STJE (060-C517244451917, 0.5% rd., 0.05% FS)	MKS Baratron (622C11TBE, 0.25% rd.)	water bath with circulator (Thermo Scientific SC150L)

Table S6. Volume calibration procedures for each lab.

Lab	Downstream volume (cm ³)	Downstream volume calibration
1	60.12 ± 0.37	Fill known volume to a pressure and measure pressure decrease to new volume. Calculate new volume from pressure difference
2	42.77 ± 0.6	Gas permeability of a standard polycarbonate film (LabThink) was measured with our system, then based on the tested value and already-known value, the downstream volume could be determined. We also use tiny stainless steel balls to fill in the downstream volume then by counting the ball numbers we can get the volume. Both values are close with an uncertainty around 0.6 cm ³ .
3	75	None, volume is fixed.
4	20.35 ± 0.066	Measuring He and N ₂ permeability of Kapton® in different membrane areas and pressures three times for each gas using non-calibrated time-lag apparatus. We already know the He and N ₂ permeability of Kapton®. So, we calculated six downstream volumes, and used the average value of downstream volume.
5	29.6 ± 0.05	Burnett expansions in triplicate using nitrogen.
6	36.20 ± 0.16	One volume was calibrated using methanol at 25 °C, average of 10 measurements were taken. Once the volume was known, all other volumes were calibrated using low pressure He at 30 °C. The known volume was loaded with He and the change in pressure on opening a valve between the known and unknown volumes was measured using the MKS Baratron Transducer. The ideal gas law was used to estimate the unknown volume based on the change in pressure between the known and unknown volume. Uncertainties were calculated by running several measurements with different pressure starting conditions, all at p _{He} ~ 0.3 Torr.
7	2.86 ± 0.02	The downstream volume (V ₀) includes dead volume in membrane cell and volumes of tubing, tube connections and valves. The stainless steel gas sampling cylinder (Swagelok® 304L-HDF2-40, V1) connected to the downstream side of permeate cell was used to calibrate the downstream volume through the isothermal expansion of N ₂ .
8	33.275 ± 0.005	Burnett expansion procedure using helium.
9	72.6 ± 0.7 for O ₂ ; 166.5 ± 0.3 for H ₂ ; 18.2 ± 0.2 for N ₂ and CH ₄	Using polycarbonate thin films (20 μm) from Goodfellow and He and N ₂ permeability data from the literature and our lab.
10	40 ± 0.1, 39.9 ± 0.1, 35.6 ± 0.1	Burnett expansion in triplicate using helium.

5. Testing conditions

Table S7. De-gas testing conditions.

Lab	Initial de-gas time (h)	De-gas time between gases (h)	Switching between gases	Hold time for each gas (h)			
				H ₂	O ₂	N ₂	CH ₄
1	22	After H ₂ > 1 h, O ₂ > 3 h, N ₂ overnight, and CH ₄ overnight.	Evacuate system to <100 Torr. Close off membrane holder and pressure purge the upstream volume twice. Then, evacuate system to 0.1 Torr or less.	0.22–0.62	1.7–5.2	2.9–5.3	3.8–7.7
2	12	After H ₂ >20 h, O ₂ > 17 h, N ₂ > 18 h, CH ₄ was tested last.	Vent the residual gas in the reservoir. Connect the gas supply line to the desired gas cylinder. Open Feed valve to fill gas reservoir to 0.11MPa. Vent the reservoir 5 times. After waiting 24h for the sample to equilibrate, purge the system with the test gas. Open the feed valve to fill the upstream section with the test gas and adjust to the desired pressure.	0.04–0.09	0.2–0.9	1.4–2.5	1.5–3
3	16 h in vacuum oven	In-situ de-gassing involved 2 h of flushing with N ₂ .	Nitrogen was used to flush for 1 h.	4–5	5–6	16	16
4	> 6 h	6 to 24 h, until there is no downstream pressure fluctuations when the downstream-pump valve is closed.	Vacuuming upstream and downstream for at least 6 hours. Sweeping all the external gas lines with the target gas before gas injection.	Until the downstream pressure gauge reached 10 torr.			
5	1	1 hour between gasses, 20 minutes between pressures.	Degas under vacuum for 1 hour, then fill and purge upstream volume with new feed up to 80 psi 4 times.	0.1–0.3	0.2–0.6	0.35–1.34	0.9–2.7
6	18	At least 6h between each gas. After CH ₄ : 12h, O ₂ : 6h, N ₂ : 12h, and H ₂ was tested last.	The high pressure on the upstream side was vented and vacuum was pulled on the downstream and upstream sample for at least 20x the time lag (θ) of the previous gas. Pressure on the downstream was allowed to reach the minimum pressure achievable before the experiment began. While the leak rate is measured, the upstream is purged at least three times with the gas of interest. This involves alternating between pressurizing the upstream and pulling vacuum. Once this exercise was completed, the upstream is left pressurized at a pressure larger than the maximum testing pressure used in the experiments.	0.2	0.6-1	0.6–1	1
7	17	0.5 h, 1 h and 2 h under vacuum when switching from H ₂ , O ₂ and N ₂ , respectively.	Degassing under vacuum at different times depending on the sample and gas of interest.	Until the downstream pressure reached 30 mbar.			
8	24	At least 20 times the time-lag for each gas.	Turn off the feed gas supply. Vent feedline and upstream to atmosphere. Hold feedline, upstream, and downstream under vacuum until system reaches 2 Torr. Disconnect feed line from gas and connect to desired gas. Shut off valve connecting feedline to vacuum and degas sample. Hold feedline under vacuum for the duration of the leak test.	0.2–0.25	0.2–0.25	0.2–0.5	0.6–3
9	18	Overnight.	The samples were exposed to vacuum overnight to be degassed and get ready for the next test.	Each pressure was tested for around 2–3 h.			
10	8	1h after H ₂ , 2 h after O ₂ , and 3 h after N ₂ .	The entire system is flushed with approximately 1 bar of helium and then held under dynamic vacuum to remove residual gas (8 h, 1 h, 2 h, and 3 h for H ₂ , O ₂ , N ₂ and then CH ₄ , respectively)	0.8	1.5	1	1

Table S8. System testing conditions for each lab.

Lab	Minimum downstream pressure (psia)	Leak rate (10^{-6} torr s^{-1})	Seal (10^{-7} torr s^{-1} cm^{-3}) ^a	$\left(\frac{dp_1}{dt}\right)_{ss} / \left(\frac{dp_1}{dt}\right)_{leak}$ ^b
1	0.0021	1.67	0.28	478
2	0.0005–0.0006	0.44	0.10	1643
3	At 20 bar: 0.0015–0.02	1.48–6.24	0.83	29
4	0.0000193	2.31	1.14	724
5	N/A, the zero baseline of the transducer shifts so it is difficult to determine.	1.0–27.6	9.25	13
6	0.0006–0.001	0.4–5	19.34	270
7	N/A	17.89–88.79	128.64	100
8	0.00029	0.46–1.23	0.47	73
9	0.001	0.177–2.8	0.018	773
10	0.00019	3.39–11.99	0.039	32

^aThe seal is calculated as the leak rate divided by the downstream volume. We used one downstream volume provided by study participants, or if multiple volumes were reported, we used the downstream volume for the first sample tested for the first gas (hydrogen) at 10 bar.

^bThe steady state transport rate vs. the leak rate measured for CH₄ at 10 bar.

Table S9. Percent contributions to the overall error propagation from the thickness (l), downstream volume (V_d), area (A), and the upstream pressure (p). Error propagations were performed for each data point. The contributions from triplicate data points were then averaged to give an overall percent contribution to error for measurements from a particular sample for each lab.

Lab	Sample	Percent contribution to overall error propagation (%)				Total error propagation (%)
		A	V_d	l	p	
2	1	0.114	25.0	71.3	3.57	5.61
	2	0.122	25.0	71.3	3.57	5.61
	3	0.116	25.0	71.3	3.57	5.61
5	1	9.34	2.96	70.2	17.5	5.70
	2	23.2	2.51	59.4	14.9	6.73
	3	9.52	3.14	68.7	18.6	5.39
6	1	34.8	3.28	53.0	8.91	13.5
	2	54.7	3.93	30.7	10.7	11.2
	3	25.2	6.79	49.6	18.4	5.42
7	1	12.0	12.6	69.4	6.00	4.17
	2	3.76	11.5	79.3	5.46	4.58
	3	9.44	16.9	65.6	8.06	3.10
9	1	9.74	0.406	83.1	6.75	3.70
	2	41.2	1.07	39.8	17.9	1.39
	3	8.01	0.434	84.3	7.22	3.46
10	1	6.98	17.6	72.6	2.83	2.14
	2	7.66	17.5	72.1	2.81	4.32
	3	11.6	16.8	69.0	2.69	4.51
Average		14.9	10.7	65.6	8.9	5.3

6. Literature comparison

For PSf samples cast from CHCl_3 (chloroform) or CH_2Cl_2 (dichloromethane, DCM), the H_2 , O_2 , N_2 , and CH_4 permeabilities fell within the range of values calculated in this work. However, when the films were cast in $(\text{CH}_3)_2\text{NCH}$ (N,N-Dimethylformamide, DMF) or melt extruded, O_2 , N_2 , and CH_4 permeabilities were lower than those reported in this study. Such variation can be attributed to differences in free volume structure of the films cast in different conditions. In particular, McHattie et al. evaluated the effect of melt-extrusion on the gas transport properties of Udel P1700 PSf [1]. Compared to solution-cast films, melt-extruded films showed consistently lower gas permeabilities for CH_4 (-24%) and CO_2 (-12%) [1–3]. On the other hand, solvent-polymer interactions can also influence the packing structure of the film, resulting in variations in transport performance. For instance, solvents with very similar Hansen solubility parameters to a polymer can sometimes provide high mobility for chain rearrangement, resulting in tighter packing structures, and thus reduced permeability [4].

Self-reported diffusion data was also compared against literature values, as shown in **Table S10**. When sorption data was reported, the diffusion coefficients were calculated from permeation experiments using the sorption–diffusion model [5]. Similar to the permeation results, the diffusion coefficients for the melt-extruded films were much lower than those of other samples due to the tighter packing structure of the polymer matrix. For all other samples, the sorption-based diffusion coefficients reported in the literature were slightly lower but still within the reported ranges in this work. This small deviation can be attributed to expected differences between diffusion coefficients calculated from direct sorption and time-lag diffusion approximations, which was previously investigated by Robeson et al. [6]. At 10 bar, for instance, N_2 and CH_4 diffusion coefficients calculated from time lag were predicted to exceed those from direct sorption by 7% and 11%,

respectively. Here, as shown in **Table S10**, time-lag diffusion coefficients calculated by McHattie et al. at 5 bar were higher than the other measurements calculated from sorption, and even closer to the average range of diffusion coefficients reported in this study.

Table S10. Reference permeability and diffusion coefficients for PSf measured at 35 °C.

Transport metric	Permeability (barrer)								Diffusion coefficient (10^{-8} cm ² /s)			Ref.
	H ₂	O ₂	N ₂			CH ₄			O ₂	N ₂	CH ₄	
Pressure (bar)	10	10	10	15	20	10	15	20	10			
Cast in CH ₂ Cl ₂ ; dried at 10 °C above T _g ~ 186 °C in vacuum	13.5 ^a	1.4 ^b	0.24	0.24	0.24	0.26	0.24	0.24	(4.4 ^c)	0.84 (1.2 ^c)	0.34 (0.33 ^c)	[1]
Cast in CHCl ₃ ; dried at 100 °C, 24h in vacuum	-	1.29	0.22	0.224	0.22	0.24	0.22	0.211	3.29	0.83	0.27	[7]
Cast in CHCl ₃ ; dried at 100 °C, 24h in vacuum	-	-	0.22	0.227	0.221	0.24	0.225	0.215	-	0.84	0.27	[8]
Oven cast in DMF; dried at 75 °C for 4h in vacuum	-	0.92	0.21	-	-	0.21	-	-	-	-	-	[9]
Melt extruded; CO ₂ conditioned for 24h at 20 atm	-	-	0.18	0.17	0.17	0.18	0.16	0.15	-	0.65	0.25	[10]
Commercial film from Goodfellow & Co.	14 ± 1	1.4 ± 0.2	0.26 ± 0.04	0.26 ± 0.04	0.25 ± 0.05	0.27 ± 0.05	0.26 ± 0.04	0.25 ± 0.01	3.5 ± 0.7	1.2 ± 0.4	0.33 ± 0.06	This work

^a1.5 bar.

^b5 bar.

^capparent diffusion coefficient from time-lag data measured at 5 bar.

Appendix 1. Word and Excel templates for submissions

1.1. Word reporting template

Contact Information			
PI Name (First, Middle, Last):		E-mail:	
Associate Name (First, Middle, Last):		E-mail:	
Associate Phone Number:		Institution:	
Physical Address of Participants (for manuscript preparation)			
Materials Specifications			
Material: Polysulfone (PSf) films with a thickness of 0.025 mm ordered from Goodfellow (http://www.goodfellowusa.com/) (Part number: SU341025; Order code: 238-653-30)			
Ordered material	Yes, from Goodfellow <input checked="" type="checkbox"/>		
Sample support	Aluminum tape <input type="checkbox"/> Brass disk <input type="checkbox"/> none <input type="checkbox"/> Other:		
Adhesive & materials used	Epoxy <input type="checkbox"/> Filter paper: <input type="checkbox"/> Other(s):		
Permeation cell type	In-line stainless steel filter holder <input type="checkbox"/> Other:		
Gases	Supplier(s):	Purity (%):	
Preparation Procedure			
Micrometer type (if applicable):			
Storage conditions			
Date PSf was received (MM/DD/YYYY):			
Date(s) each PSf sample was tested (MM/DD/YYYY):			
Equipment Information (please define the type of error reported)			
Temperature control	Water bath with circulator <input type="checkbox"/> Air bath <input type="checkbox"/> Other:		
Constant-volume, variable-pressure working mechanism used: <input type="checkbox"/>			
Upstream pressure transducer model (if known, please provide the zero-point and span uncertainty):			
Downstream pressure transducer model (if known, please provide the zero-point and span uncertainty):			
Downstream volume size and uncertainty (if multiple volumes used, please specific which volume used for each gas/pressure):			
Calibration procedure used for downstream volume			

What is your procedure for switching between gases while testing?									
Raw data									
Raw data file <u>using the Excel template provided in the appropriate units:</u> <input type="checkbox"/>									
Actual upstream pressures (psia)									
Actual operating temperatures (°C):									
Testing times and sequences (hours)									
De-gassing time prior to testing (hours):									
Minimum pressure in downstream under vacuum (psia)									
Film thickness with standard deviation (µm)									
Area measurement with standard deviations (cm ²):									
Technique used to measure area (e.g., ruler, imageJ):									
Leak rate*:				Steady-state pressure rise for each sample (dp/dt)*:					
Calculated permeation data									
Order in which the gases were tested:									
Permeability (barrer)	10 bar			15 bar			20 bar		
Sample #	1	2	3	1	2	3	1	2	3
H ₂									
O ₂									
N ₂									
CH ₄									
Time-lag (in seconds) at 10 bar, if tested/accessible.									
Sample #	1			2			3		
H ₂									
O ₂									
N ₂									
CH ₄									

* See corresponding Excel sheet. If leak rates were recorded prior to each gas measured, please provide the information for each gas in the paragraph below.

Detailed Experimental Procedure (paragraph summarizing a step-by-step procedure used to test the samples, including any information that was not included in the previous questions.)

Note: these paragraphs will be included in the Supplementary Information of the published work.

1.2. Excel file submission template

The spreadsheet is organized as follows:

- Row 1:** Date of test (MM/DD/YYYY)
- Row 2:** Leak rate* (blank)
- Row 3:** Hydrogen Downstream pressure (Torr)
- Row 4:** Leak rate* (blank)
- Row 5:** Nitrogen Downstream pressure (Torr)
- Row 6:** Leak rate* (blank)
- Row 7:** Oxygen Downstream pressure (Torr)
- Row 8:** Leak rate* (blank)
- Row 9:** Methane Downstream pressure (Torr)
- Row 10:** Leak rate* (blank)
- Row 11:** Time (s)
- Row 12:** Hydrogen Downstream pressure (Torr)
- Row 13:** Time (s)
- Row 14:** Nitrogen Downstream pressure (Torr)
- Row 15:** Time (s)
- Row 16:** Oxygen Downstream pressure (Torr)
- Row 17:** Time (s)
- Row 18:** Methane Downstream pressure (Torr)
- Row 19:** Time (s)
- Row 20:** Hydrogen Downstream pressure (Torr)
- Row 21:** Time (s)
- Row 22:** Nitrogen Downstream pressure (Torr)
- Row 23:** Time (s)
- Row 24:** Oxygen Downstream pressure (Torr)
- Row 25:** Time (s)
- Row 26:** Methane Downstream pressure (Torr)
- Row 27:** Time (s)
- Row 28:** Hydrogen Downstream pressure (Torr)
- Row 29:** Time (s)
- Row 30:** Nitrogen Downstream pressure (Torr)
- Row 31:** Time (s)
- Row 32:** Oxygen Downstream pressure (Torr)
- Row 33:** Time (s)
- Row 34:** Methane Downstream pressure (Torr)
- Row 35:** Time (s)
- Row 36:** Hydrogen Downstream pressure (Torr)
- Row 37:** Time (s)
- Row 38:** Nitrogen Downstream pressure (Torr)
- Row 39:** Time (s)
- Row 40:** Oxygen Downstream pressure (Torr)
- Row 41:** Time (s)
- Row 42:** Methane Downstream pressure (Torr)
- Row 43:** Time (s)
- Row 44:** Hydrogen Downstream pressure (Torr)
- Row 45:** Time (s)
- Row 46:** Nitrogen Downstream pressure (Torr)
- Row 47:** Time (s)
- Row 48:** Oxygen Downstream pressure (Torr)
- Row 49:** Time (s)
- Row 50:** Methane Downstream pressure (Torr)
- Row 51:** Time (s)

Four empty line graphs are provided for each gas, with the following axes:

- Hydrogen:** Y-axis: Downstream pressure (Torr) [0 to 1.2]; X-axis: Time (s) [0 to 1.2]
- Nitrogen:** Y-axis: Downstream pressure (Torr) [0 to 1.2]; X-axis: Time (s) [0 to 1.2]
- Oxygen:** Y-axis: Downstream pressure (Torr) [0 to 1.2]; X-axis: Time (s) [0 to 1.2]
- Methane:** Y-axis: Downstream pressure (Torr) [0 to 1.2]; X-axis: Time (s) [0 to 1.2]

At the bottom of the spreadsheet, there are four tabs: Pressure 10 bar, Pressure 15 bar, Pressure 20 bar, and a plus sign for additional tabs.

Appendix 2. Analysis of co-dependencies for H₂ and CH₄

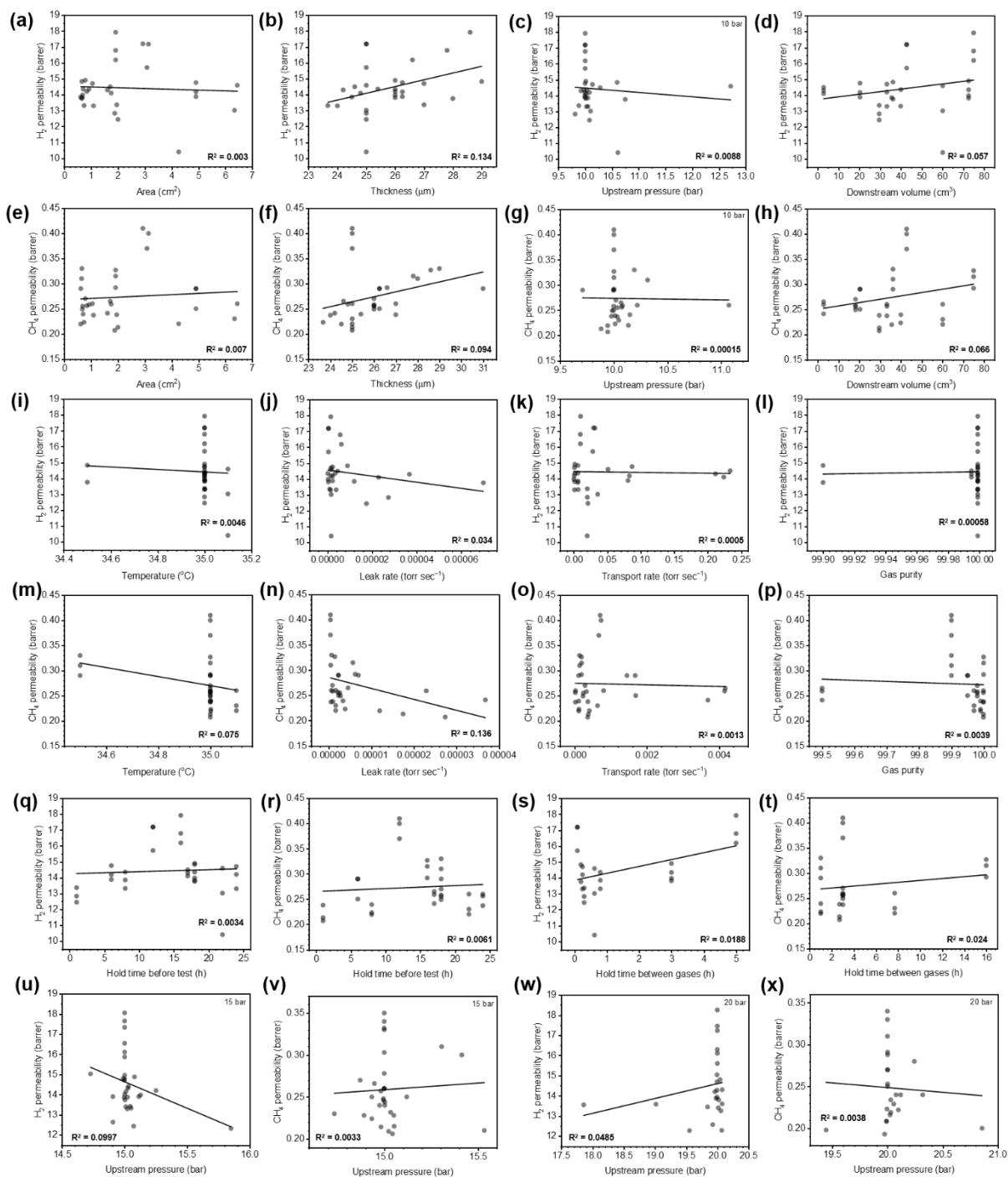


Figure S4. Co-dependency analysis for H₂ permeabilities versus (a) area, (b) thickness, (c) upstream pressure (10 bar), (d) downstream volume, (i) temperature, (j) leak rate, (k) transport rate, (l) gas purity, (q) hold time before the test, (s) hold time between gases, (u) upstream pressure

(15 bar), and (w) upstream pressure (20 bar). Co-dependency analysis for CH₄ permeabilities versus (e) area, (f) thickness, (g) upstream pressure (10 bar), and (h) downstream volume, (m) temperature, (n) leak rate, (o) transport rate, (p) gas purity, (r) hold time before the test, (t) hold time between gases, (v) upstream pressure (15 bar), and (x) upstream pressure (20 bar). R² values were calculated from linear fits using OriginLab 2018b.

Appendix 3. Raw data for time lag calculations

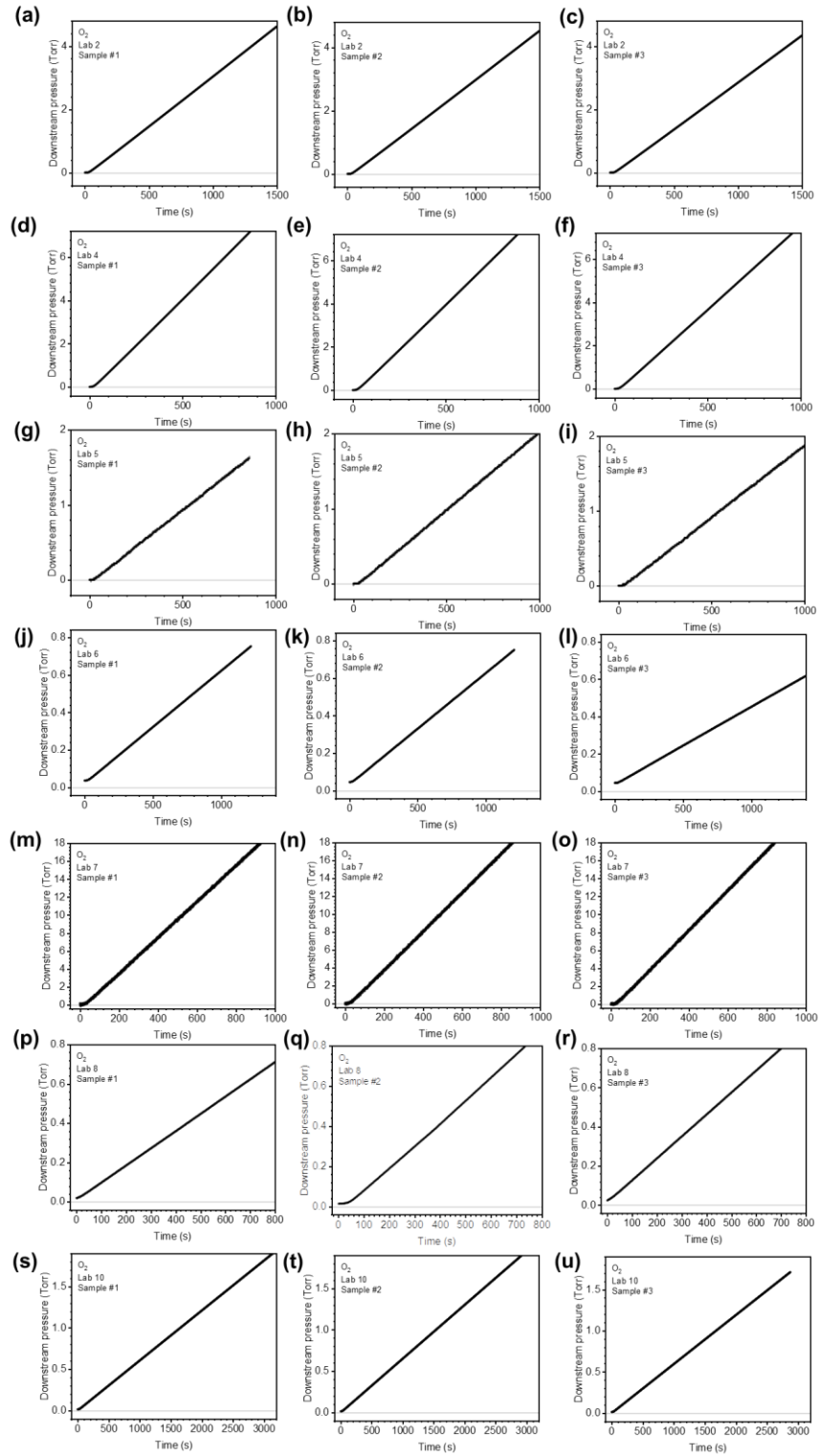


Figure S5. Raw data for O₂ time-lag calculations.

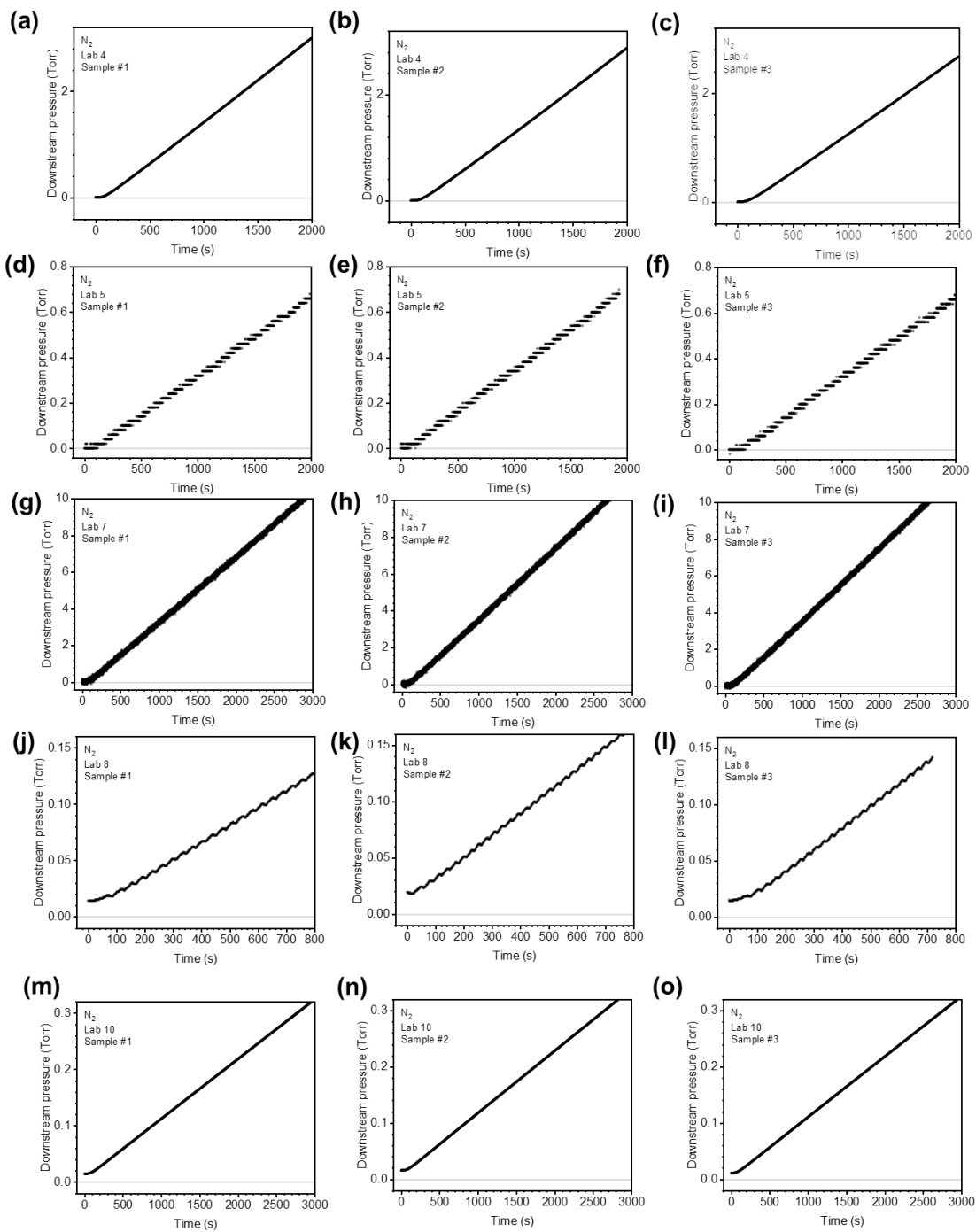


Figure S6. Raw data for N_2 time-lag calculations.

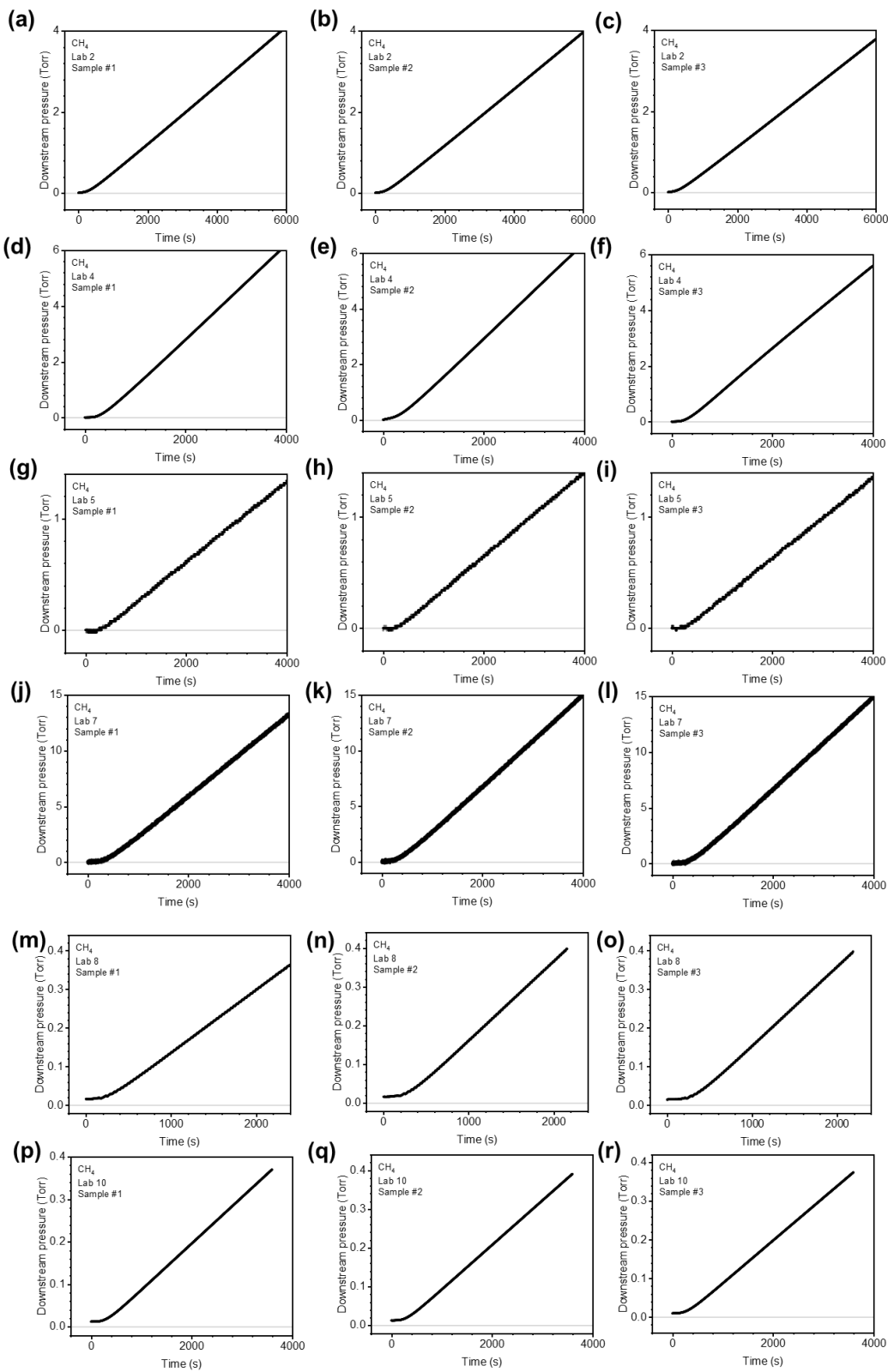


Figure S7. Raw data for CH₄ time-lag calculations.

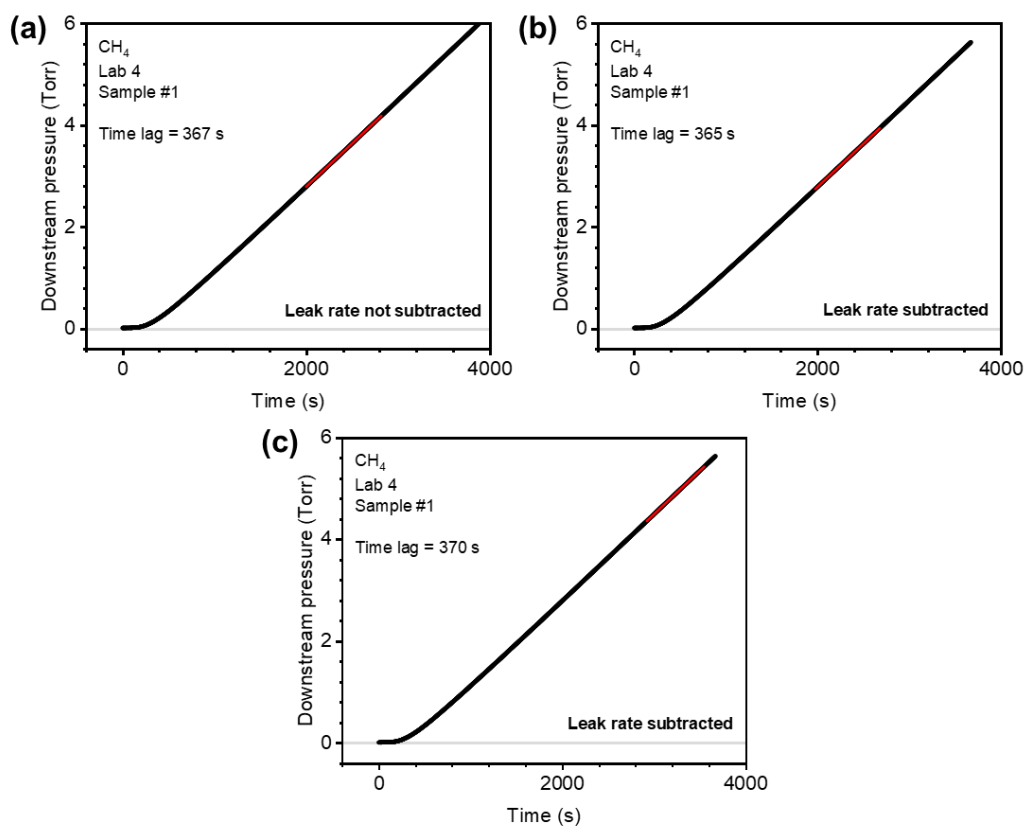


Figure S8. Time-lag estimation for an example data set from Lab 4 where (a) the leak rate was not subtracted from the data, and (b and c) the leak rate was subtracted from the data but two different time ranges were used for the fit.

Appendix 4. Detailed experimental procedure for each lab

Lab 1

1. System purge
2. Choose which gas will be used in the test.
3. Pressurize feed tubing with desired gas. Vent gas to snorkel. Repeat once more.
4. Fill feed system with test gas. Vent gas to snorkel. Repeat once more.
5. Vacuum entire system and pump down according to list.
 - a. Before H_2 – overnight
 - b. Before N_2 –minimum 1 hr
 - c. Before CH_4 –overnight
 - d. Before O_2 –overnight (different unit)
6. Pressurize feed tubing with desired gas. Vent gas to snorkel. Repeat once more.
7. Start data collection.
8. Slowly increase gas pressure over at least 10 seconds to avoid damaging membrane.
9. Monitor pressure as gas permeates membrane and passes to low pressure side of membrane. (At least 10 Torr)
10. To end run, stop data collection, vent upstream gas to snorkel, then evacuate system.
11. Repeat including system purge for each gas run.

Lab 2

A Sample mounting and evacuating the system

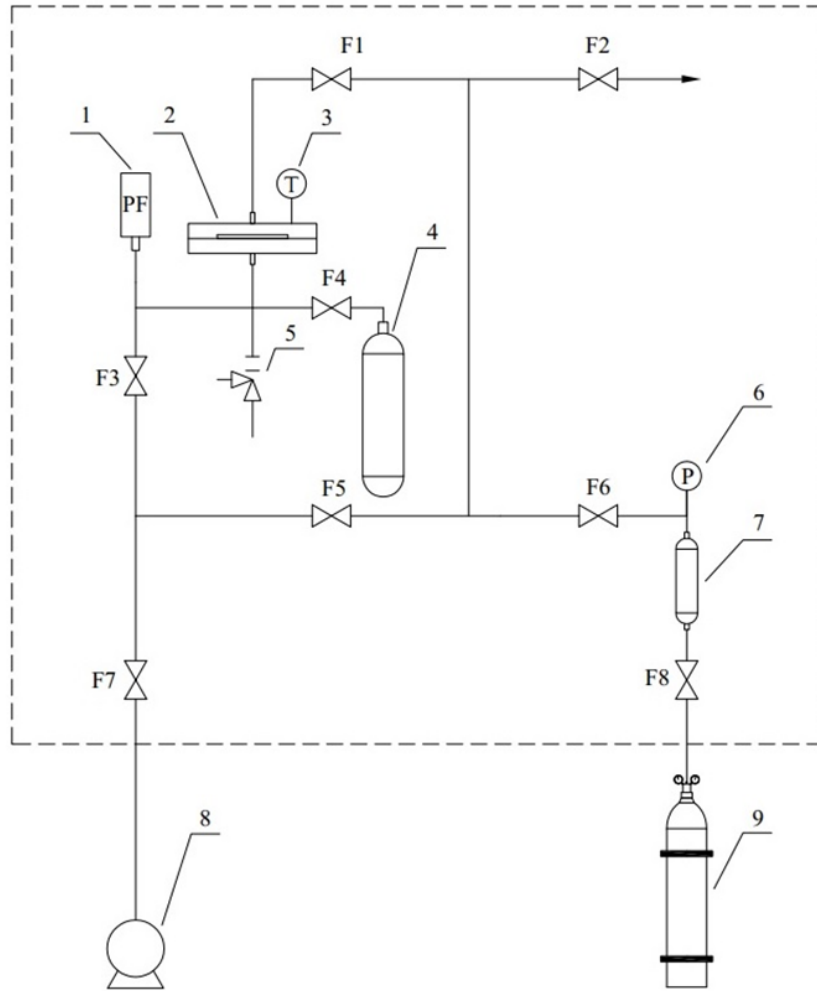
1. Prepare and mount the sample (make sure the membrane is free defect) onto the permeation cell. Flatten down the Al foil.
2. Measure the thickness and the diameter of the membrane.
3. Open vacuum pump and F7; open F3; Vacuum will suck sample towards the cell surface.
4. Replace top cover of permeation cell and lock in all 6 screws.
5. Set the test temperature.
6. Open F5 then open F1 slowly.

B Purging gas reservoir

1. Close F1 and F5.
2. Open F2 slowly. This will vent the residual gas in the reservoir.
3. Close F2.
4. Connect the gas supply line to the desired test gas cylinder.
5. Turn on the gas supply valves on the gas cylinder and adjust outlet pressure to about 50 psi. Open F6 and F8 to fill up gas reservoir. Fill to about 20 psi (check from pressure gauge).
6. Close F8.
7. Open F2 to vent reservoir.
8. Close F2.
9. Repeat steps 3 times.
10. Close F2 and F8.

C Conducting the test

1. Allow enough time for sample to equilibrate the test temperature and vacuum has stabilized (Check Pressure transducer PT). As a guide, this takes about 24 hours after the last step in “Sample mounting and evacuating the system”.
2. Close F1 and F5; Close vacuum pump.
3. Open F6 to fill the upstream section with the test gas. Adjust to till required pressure is obtained.
4. Ensure chart recorder settings have been set properly.
5. Open F1 and start chart recorder simultaneously.
6. Stop chart recorder in computer.
7. Open vacuum pump; open F7 and F3.
8. Open F2 to evacuate the upstream system.
9. Close F2.
10. Open F5.



The system leak rate is measured every month.

Lab 3

Experimental procedure: 8 polysulfone coupons were cut and placed in a vacuum oven at 50 °C overnight. After degassing, the coupons were placed in the high-throughput gas separation set-up (HTGS). The set-up was pressurized with N₂ at the desired pressure and a vacuum was applied downstream of the membranes. The membranes were tested for leaks/defects. 4 coupons were defect and thus not considered for testing. Out of the other 4 coupons, one coupon showed a significantly higher gas flux and was also considered defect. The three remaining coupons were tested with gases in order of increasing critical temperature and with increasing feed pressure. This means that first H₂, N₂, O₂ and CH₄ at 10 bar feed were tested, then at 15 bar and finally at 20 bar. Between each gas type, the set-up was flushed with N₂ for 2 h for safety reasons.

Membrane steady-state was assessed by frequently monitoring the gas flux (by monitoring the pressure rise). Once the flux had stabilized (3 consecutive measurements with less than 2% error), the last measurement was considered the steady state flux and used for permeability calculation. Steady-state time depended on membrane thickness, gas type and feed pressure, as indicated above, and varied between 4 - 7 h.

After measuring the membrane gas permeation, leak rates were recorded. All 16 position valves were closed and a vacuum was applied for 15 h. Next, the leak rate of each position was measured consecutively.

Lab 4

Sample preparation: Membrane film cut into 3 x 3 cm and sandwiched between two Al foils with 2.5 cm holes. The membrane was attached on the foil using epoxy resin. The membrane cell was stored in ambient conditions for at least 15 min for epoxy curing before used.

Measurement: Membrane cell was inserted into the sample holder. Upstream and downstream were vacuumed for at least 6 hr before gas injection. Upstream was filled with gas up to testing pressure. Waited until the oven temperature became constant at 35 °C. Open the sample valve, and detected downstream pressure with time up to 10 Torr.

Plot: Time-lag (second) was obtained from linear plot of downstream pressure curve. Here, the section for the linear plot was selected from the linear part of the curve.

Lab 5

Samples prep: 3 membrane samples were punched from film with a 3/4" bore punch. Thickness of each sample was measured using a micrometer. Samples were next sandwiched between a 42.5 mm diameter piece of filter paper and a 50 mm diameter aluminum foil tape disk with a 5/8" hole punched from the center. Samples were epoxied with Devcon 10 minute black epoxy and allowed to cure at ambient conditions for 16 hours before being stored under vacuum until use.

Permeation test: Membrane surface area was first analyzed using imageJ and a photo of the epoxied membrane with a fine ruler for reference. Then the membrane was loaded into a stainless steel permeation cell (Millipore), which was then loaded into the permeation system. The permeation cell was evacuated at 35°C for at least 2 hours to degas the membrane and equilibrate at temperature. Leak tests were performed by isolating the permeation system from vacuum for at least 20 minutes during which the downstream pressure change was measured and reported as the leak rate for that membrane sample for all subsequent tests. Next the upstream volume was manually charged with the desired gas at the desired pressure with the cell temporarily isolated. Data collection commenced concomitantly as the upstream volume opened to the cell. The permeation test proceeded for at least 14 times the time lag. The steady-state permeability was calculated from either 10-14 times the time lag for slow penetrants (O₂, N₂, CH₄) or from a stretch of at least 300 seconds past 14 times the time lag for H₂ to reduce the effect of noise. The permeation system was evacuated and the membrane degased for at least 20 minutes between different pressures, and at least 1 hour between different gases. The feed line and permeation system was purged 4 times after gas switches before charging to the desired pressure. Samples

took several days to complete all tests, and the cells were kept under vacuum, unheated overnight. Cells were allowed to re-equilibrate at 35°C for 2 hours before performing the first test each day.

Lab 6

Polysulfone films (membranes) with known thickness were mounted on to brass discs (outer diameter – 47 mm) with known inner diameter (0.375 in) using Devcon 5-min epoxy to properly adhere them to the brass. The membranes were supported at the bottom by 0.45 μm Whatman filter paper. The paper has negligible resistance to transport. Films are loaded into the sample holders of the permeation cell (High Pressure Stainless Steel Filter Holder, EMD Millipore) and the edges of the sample holders were coated with vacuum grease for optimal sealing of the sample cell. To maintain temperature of the bath, deionized water is added to completely submerge the membrane cell and the temperature is controlled with a circulating heater (Thermo Fischer Scientific) and the temperature is set to 35 $^{\circ}\text{C}$ (308 K). The time taken to reach thermal equilibrium is around 3 hours for our current setup. Once this step is complete, the downstream of the cell is evacuated until a pressure of ~ 1 torr is reached. Subsequently, the volume upstream of the membrane is also evacuated and held under continuous vacuum overnight (\sim at least 12 hours) until a steady state pressure of $\approx 30\text{-}50$ mTorr is achieved downstream. Pressure is monitored using a Honeywell STJE (0-1000psia) for the high pressure upstream and a MKS Baratron 626C transducer (0-e10 Torr) for the low pressure downstream.

The membrane holder and downstream volume was then isolated from the vacuum and the rate of pressure increase in the downstream chamber of known volume is measured to determine the “leak rate” of the sample. In all cases the leak rate was < 20 mTorr/h (lowest leak rate recorded was ~ 3 mTorr/h; contrasted with a typical pressure increase of > 300 mTorr/h for the least permeable sample. While the leak rate is being measured, the high pressure side of the cell is purged with the gas of interest (penetrant) at least three times to ensure that permeabilities measured are free of contamination from other gases. Purging involves – a) Evacuating the high

pressure tubes of the cell with the vacuum pump, b) Pressurizing the upstream with ~ 2 bar of the gas of interest; c) Evacuating the tubes once again when the system was pressurized; d) Repeating this process at least 3 times; e) Set the pressure of the upstream side with a manual pressure regulator.

After the leak rate is measured for about 1 h, the system is returned to the earlier ultimate vacuum before beginning the experiment. Once ultimate vacuum is re-obtained, the downstream is again isolated from the vacuum and the upstream is charged with ≈ 10 bar of the desired penetrant gas. Both the upstream (high) and downstream (low) pressure were recorded as a function of time as gas diffused through the film using a python based graphical user interface made available to us by the Freeman Lab at UT Austin. The test continued up to the time the rate of increase of pressure on the downstream side was steady with time and the time lapsed from the start of the experiment to the end was at least 10x the time lag of the penetrant. The permeability of the gas, P_1 , was calculated from the steady-state rate of downstream pressure increase:

$$P_1 = \frac{V_d \ell}{p_u ART} \left[\left(\frac{dp_d}{dt} \right)_{ss} - \left(\frac{dp_d}{dt} \right)_{leak} \right]$$

The rate of change of downstream pressure, $\left(\frac{dp_d}{dt} \right)_{ss}$ was calculated from the last 10% of the data collected, which was well into the steady state regime of the experiment. One dataset showed an unexpected upturn for the last 5% of the data even after steady state had been reached. For that particular dataset, the last 5% of the data was discarded and the last 10% which displayed the steady state behavior. The steady-state data of pressure vs time was extrapolated back to intersect the x-axis at the starting pressure of the experiment to estimate the time lag. Prior to calculating the time lag, the instrumental leak rate was NOT subtracted from the downstream pressure. Difficulties in estimating the true zero-time i.e. the start of diffusion through the membrane due to

manual operation of the valves makes the calculated time lags sensitive to operator error. Thus the time lags measured on the instrument are useful for estimating the diffusion coefficient, but small changes in diffusivity are difficult to distinguish due to the measurement errors.

Once enough data was collected, the pressure was increased to the higher pressure and allowed to equilibrate at the higher pressure before starting data collection. The volume downstream of the membrane was also evacuated concurrently to reduce the pressure to an acceptable starting point. Once the membrane had equilibrated, data collection was restarted in a new file and continued until steady state had been unequivocally achieved.

After each experiment was completed (both the 10 and 15 bar measurements) the upstream gas supply was shut off and the downstream was re-opened to the vacuum to remove the gas that had permeated through the film. The film, downstream and upstream were subsequently degassed for at least 6 hours (much larger than the experimental time lags of the gases). The leak rate was once again measured before the next gas to be measured. Uncertainties for each measurement were calculated using error propagation of the variables.

Lab 7

Pure gas permeation tests were carried out using a constant-volume pressure-increase apparatus developed following the guidance of reference [1]. A stainless-steel membrane holder (Millipore, model XX4404700, diameter of 47 mm) was used as the membrane cell where the membrane was supported on filter paper and a porous metal disk at the bottom. The gas permeation system is placed in an oven with temperature of 35.6 °C. Membrane samples were evacuated for at least 17 hours before gas permeation tests under each pressure to ensure complete removal of adsorbed gases. Leakage rate was measured after degassing and before permeation tests. The gases were tested in the following order: H₂, O₂, N₂, and CH₄. The sample was evacuated for 0.5 h, 1 h and 2 h before being switched to O₂, N₂, and CH₄ for complete removal of the previous gas. The gas feed pressure was set to 10, 15 or 20 bar, as measured by a Keller digital manometer (LEO2EI-1-30BAR, 0-31 bar, 0.1% FS).

The permeability is derived from the following equation:

$$P = \frac{Vl}{A} \frac{T_0}{P_f P_0 T} \left[\left(\frac{dP}{dt} \right)_{ss} - \left(\frac{dP}{dt} \right)_{leak} \right]$$

where P is the permeability of the gas through the membrane, generally expressed in Barrer (a non-SI unit), 1 Barrer= $10^{-10} \text{ cm}^3(\text{STP})\text{cm}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}\cdot\text{cmHg}^{-1}$. V is the permeate volume (cm^3), l is the thickness of membrane, A is the effective area of the membrane (cm^2), P_f is the feed pressure (cmHg), P_0 is the pressure at standard state (76 cmHg), T is the absolute operating temperature (K), T_0 is the temperature at standard state (273.15 K), $(dP/dt)_{ss}$ and $(dp/dt)_{leak}$ are the rate of pressure increase in the permeate volume at steady state and leakage (cmHg/s), respectively, recorded by a pressure transmitter (Keller PAA 33X, Accuracy of 0.01% F.S.).

Lab 8

Once the polysulfone film arrived, it was stored in a dry drawer. A brass disk with a hole of 0.5 inch diameter was cleaned using Kim Wipes and isopropyl alcohol. The film was inspected and a seemingly defect-free portion of the film was cut out. The film was cut to leave approximately a quarter inch overlap around the entire hole. Ten to thirteen thickness measurements were taken using a micrometer while taking special care not to dent the film. Fisherbrand P8 filter paper was cut in order to have a quarter inch overlap around the entire polysulfone film. The brass disk was placed concave down onto a nut. Devcon two component 5-minute epoxy was squeezed into a catch can and a small brush was used to mix the two components for thirty seconds. The brush was then used to apply moderate amounts of epoxy onto the brass disk. The polymer film was carefully laid onto the center of the brass disk followed by the filter paper. Using tweezers, the entire structure was taken off of the nut and placed onto a sheet of weigh paper. A nut with a hole slightly larger than the hole of the brass disk was placed onto the structure followed by a one kilogram weight. The epoxy was left to dry for at least two hours. Once dried, the films were placed into an image scanner and scanned for the purpose of measuring the area. Using ImageJ, the area was computed by tracing a polygon where the film meets the epoxy. This was done seven times and the average area was used in the permeability calculation.

The permeation system that was used for this study is outfitted with a water bath. The water was drained, the cell was wiped dry, and the bolts were removed. Once open, the interior of the cell was wiped clean of vacuum grease and the O-rings were removed. New O-rings coated with vacuum grease were inserted into their proper crevices and the edges of the cell are lightly dabbed with any excess vacuum grease to prevent water contamination. The sample was placed onto the downstream portion of the cell and the vacuum was connected to the downstream to cinch

the sample down and to test if any obvious leaks were present. The upstream side of the cell was carefully placed onto the downstream and bolted into place. If permeation properties for the specific material being tested are not known as was the case in this study, the sample is left to degas for 24 hours (the first degas of the first sample). If the properties are known, the sample is left to degas for at least $20 \cdot \theta$ (last gas the sample was exposed to). After the degas, the feedline was connected to the appropriate gas and held under vacuum for the duration of the hour long leak test. The entire leak test is used to calculate the leak rate. The system was then evacuated for five minutes. During the ramp for each pressure step, the gas was fed into the upstream at a rate of ~ 2 psi per second. Each pressure step was tested for $6 \cdot \theta$ or twelve minutes, whichever was larger. The last 10% of each pressure step was used to calculate the flux. The downstream was evacuated for five minutes between each step. Once all of the pressure steps were complete, the gas supply was turned off. The feedline and upstream were purged and evacuated to ~ 2 Torr. The feedline was then connected to the desired gas and the sample degassed.

Lab 9

Pure-gas permeability was measured using a constant volume/variable pressure method at 35 °C. After the film was partially masked by copper shim using epoxy adhesive and installed in a permeation cell, both upstream and downstream sides of the sample were exposed to vacuum for degassing overnight. The leak rate of the permeation system was measured before introducing the feed gas to the system. After the feed gas was introduced at desired pressures, the pressure increase of the downstream volume was recorded as a function of time. The permeability coefficient (P_A) was calculated according to the following equation [11,12]:

$$P_A = \frac{V_d l}{p_2 A R T} \left[\left(\frac{dp_1}{dt} \right)_{ss} - \left(\frac{dp_1}{dt} \right)_{leak} \right]$$

where V_d is the downstream volume, l is the thickness of the sample, p_2 is the upstream pressure, T is temperature, R is the gas constant, and A is the effective area of the sample. The $(dp_1/dt)_{ss}$ and $(dp_1/dt)_{leak}$ are the pressure increase of steady-state permeation and leaking test in the downstream volume. The gases were tested with the sequence of N₂, H₂, CH₄, and O₂.

Lab 10

The PSf film from Goodfellow was stored in a room temperature drawer and its packaging material until permeation testing. Once ready for testing, a piece of PSf of 25 mm² was cut from the commercial film. The thickness was measured in triplicate using a Mitutoyo micrometer and the film was adhered to a brass disk using Devon 5-min Epoxy and filter paper as backing. Once the epoxy solidified, the area was measured using ImageJ software and the sample was loaded into an in-line stainless-steel permeation cell using new O-rings with a small amount of vacuum grease. Pure-gas H₂, O₂, N₂, and CH₄ permeation tests were performed on a constant-volume variable-pressure system provided by Maxwell robotics. The system temperature was controlled using a ThermoFischer circulator in a water bath. In a typical test, the film is loaded to the cell and de-gassed for about 30 minutes. A routine leak test is conducted by pressurizing the upstream to 15 psia and flowing gas to the film to confirm that the film is not damaged. Once the film passes the preliminary leak test, the sample is de-gassed for 8 h, the leak is tested again, and the testing begins. To switch between gases, a Helium flush at 15 psia followed by evacuation for longer than six times the time lag of the previous gas was applied. The gases were tested in the following order: H₂, O₂, N₂, and CH₄. The leak rate of the system was calculated through a 30 min long leak rate at the beginning of the test. Finally, the steady state transport rate for each test was used to calculate permeability using an automated code in MATLAB.

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