Supporting Information

Microporous Crystalline Membranes for Kr/Xe Separation: Comparison Between AlPO-18, SAPO-34, and ZIF-8.

Ting Wu[1], Jolie Lucero [1], Zhaowang Zong[1], Sameh K. Elsaidi[2], Praveen K. Thallapally[2]*, Moises A. Carreon[1]*

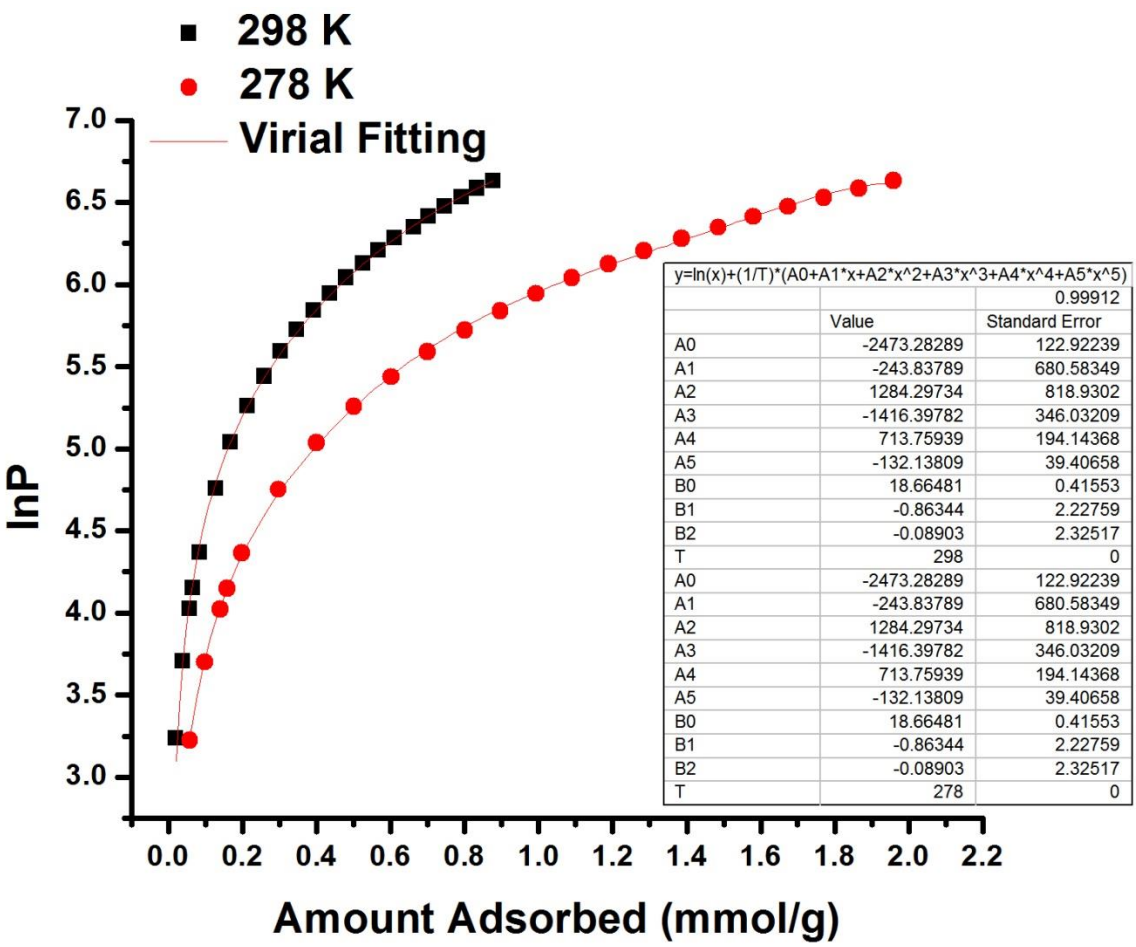
[1] Chemical and Biological Engineering Department, Colorado School of Mines, Golden, CO 80401 United States

[2] Pacific Northwest National Laboratory, Richland, WA 99352 United States

*Corresponding authors: mcarreon@mines.edu; Praveen.Thallapally@pnnl.gov

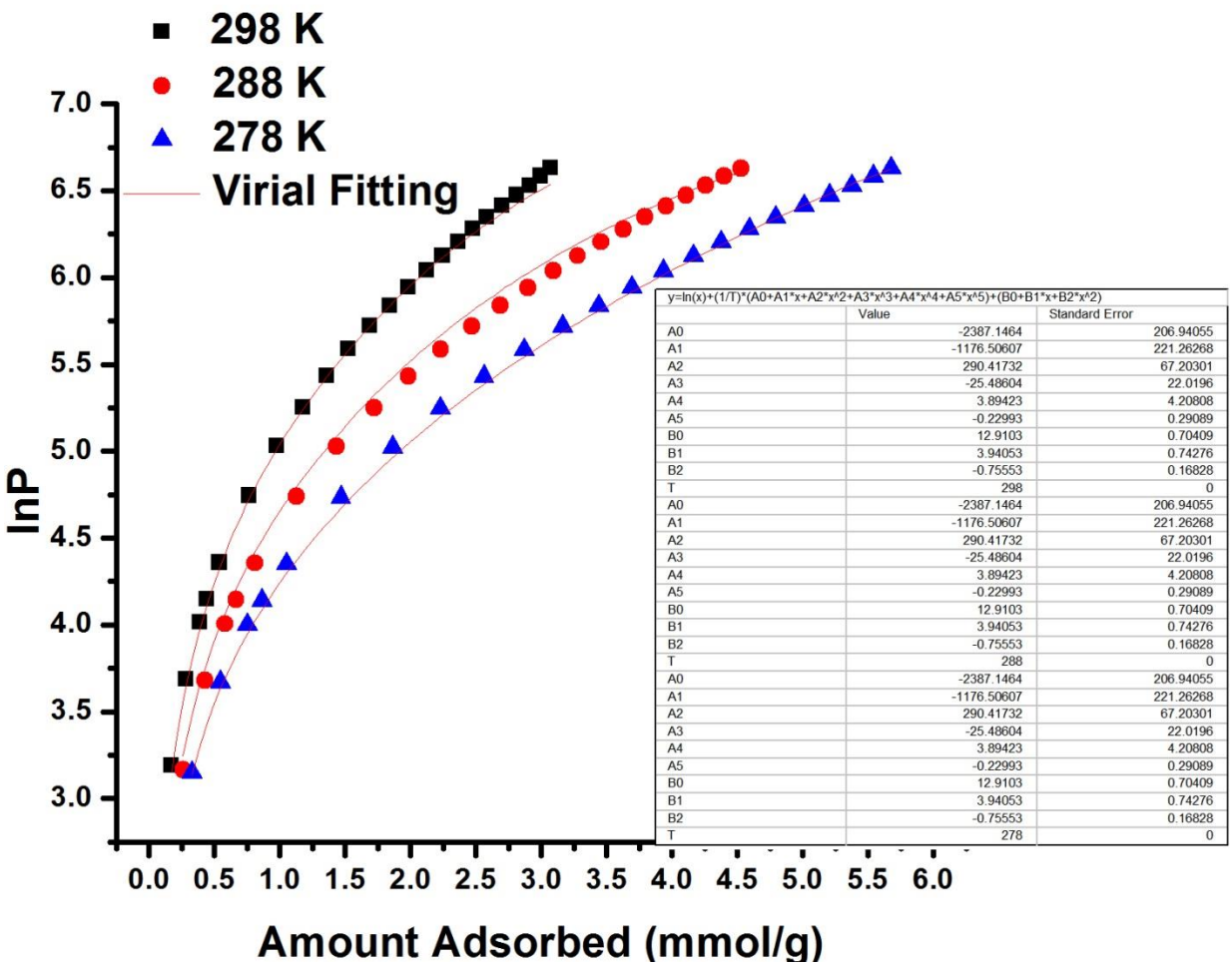
Calculation of Isosteric heat of Adsorption (Q_{st}):

The Q_{st} values for Xe and Kr gases over AlPO-18 crystals were calculated using Virial method as shown in Figures S1 and S2



y=ln(x)+(1/T)*(A0+A1*x+A2*x^2+A3*x^3+A4*x^4+A5*x^5)		
		0.99912
	Value	Standard Error
A0	-2473.28289	122.92239
A1	-243.83789	680.58349
A2	1284.29734	818.9302
A3	-1416.39782	346.03209
A4	713.75939	194.14368
A5	-132.13809	39.40658
B0	18.66481	0.41553
B1	-0.86344	2.22759
B2	-0.08903	2.32517
T	298	0
A0	-2473.28289	122.92239
A1	-243.83789	680.58349
A2	1284.29734	818.9302
A3	-1416.39782	346.03209
A4	713.75939	194.14368
A5	-132.13809	39.40658
B0	18.66481	0.41553
B1	-0.86344	2.22759
B2	-0.08903	2.32517
T	278	0

Figure S1. Kr adsorption isotherm of AlPO-18 at 298 K and 278 K fitted using Virial equation.



y=ln(x)+(1/T)*(A0+A1*x+A2*x^2+A3*x^3+A4*x^4+A5*x^5)+(B0+B1*x+B2*x^2)

	Value	Standard Error
A0	-2387.1464	206.94055
A1	-1176.50607	221.26268
A2	290.41732	67.20301
A3	-25.48604	22.0196
A4	3.89423	4.20808
A5	-0.22993	0.29089
B0	12.9103	0.70409
B1	3.94053	0.74276
B2	-0.75553	0.16828
T	298	0
A0	-2387.1464	206.94055
A1	-1176.50607	221.26268
A2	290.41732	67.20301
A3	-25.48604	22.0196
A4	3.89423	4.20808
A5	-0.22993	0.29089
B0	12.9103	0.70409
B1	3.94053	0.74276
B2	-0.75553	0.16828
T	288	0
A0	-2387.1464	206.94055
A1	-1176.50607	221.26268
A2	290.41732	67.20301
A3	-25.48604	22.0196
A4	3.89423	4.20808
A5	-0.22993	0.29089
B0	12.9103	0.70409
B1	3.94053	0.74276
B2	-0.75553	0.16828
T	278	0

Figure S2. Xe adsorption isotherm of AlPO-18 at 298 K and 278 K fitted using Virial equation.

Table S1. Breakthrough times for AlPO-18, SAPO-34 and ZIF-8 crystals

Porous crystal (bulk material)	Column Breakthrough time
AlPO-18	Kr: 3 min; Xe: 10 min
SAPO-34	Kr: 22 min; Xe: 39 min
ZIF-8	Kr: 2.6 min; Xe: 6 min

Estimation of Fickian Diffusivities.

The experimental diffusivities for Kr and Xe over AlPO-18, SAPO-34 [10] and ZIF-8 [12] membranes are shown in **Tables S2-S4**. Fick's first law: was used to fit collected experimental data for these membranes under the assumptions of steady state, ideal gases (Kr and Xe), one-dimension gas transport, and constant membrane thicknesses. J (mol/m^{2}s), represents the gas diffusion flux estimated by multiplying the permeance of each gas component and the partial pressure of the gas in the permeate side. (mol/m^3) represents the molar gas concentration gradient, which is the difference between the gas concentration in the feed side and that in the permeate side. is membrane thickness. The diffusivities were derived from experimental results and thus incorporate adsorption effects.

Table S2. Estimated diffusivities for Kr/Xe gas mixture (feed composition: 8% Kr; 92 % Xe) over AlPO-18 membranes.

Membrane ID	D (Kr) ($x10^{-10}$ m^2/s)	D (Xe) ($x10^{-10}$ m^2/s)	D(Kr)/D(Xe)
M1	6.08	1.38	4.44
M2	6.73	0.79	8.51
M3	7.68	3.17	4.65
M4	5.68	0.78	7.26
M5	3.41	0.68	5.04

Table S3. Diffusivities for Kr/Xe gas mixture (feed composition: 8% Kr; 92 % Xe) over SAPO-34 membranes [1]. Membrane IDs same as reported elsewhere [1]

Membrane ID [10]	)	D(Kr) (10^{-10} m^2/s)	D(Xe) (10^{-10} m^2/s)	D(Kr)/D(Xe)
A (150)	8.7	3.95	0.31	12.7
B (200)	6.0	11.00	0.56	19.4
C (250)	4.3	10.50	0.33	32.3
D (300)	3.5	9.39	0.49	19.1
E (350)	3.0	8.86	0.32	27.3

Table S4. Estimated diffusivities for Kr/Xe gas mixture (feed composition: 8% Kr; 92 % Xe) over ZIF-8 membranes [2]. Membrane IDs same as reported elsewhere [2]

Membrane ID [12]	(μm)	D(Kr) (10^{-10} m^2/s)	D(Xe) (10^{-10} m^2/s)	D(Kr)/D(Xe)
1A	21.5	1.76	0.32	5.5
1B	22.0	0.95	0.08	12.5
2A	22.0	9.33	0.68	13.7
2B	25.0	8.11	0.51	16.0
C	33.0	3.74	0.41	9.0

References

[1] Feng, X.; Zong, Z.; Elsaidi, S. K.; Jasinski, J. B.; Krishna, R.; Thallapally, P. K.; Carreon, M. A. Kr/Xe Separation over a Chabazite Zeolite Membrane. Krypton- Xenon Separation Properties of SAPO- 34 Zeolite Materials and Membranes. *J. Am. Chem. Soc*. **2016**, *138*, 9791-9794.

[2] Wu, T.; Feng, X.; Elsaidi, S. K.; Thallapally, P. K.; Carreon, M. A. Zeolitic Imidazolate Framework-8 (ZIF-8) Membranes for Kr/Xe Separation. *Ind. Eng. Chem. Res*. **2017**, *56*, 1682-1686.