Supplementary Information

for

Computer Simulations of 4,240 MOF Membranes for H_2/CH_4 Separations: Insights into Structure-Performance Relations

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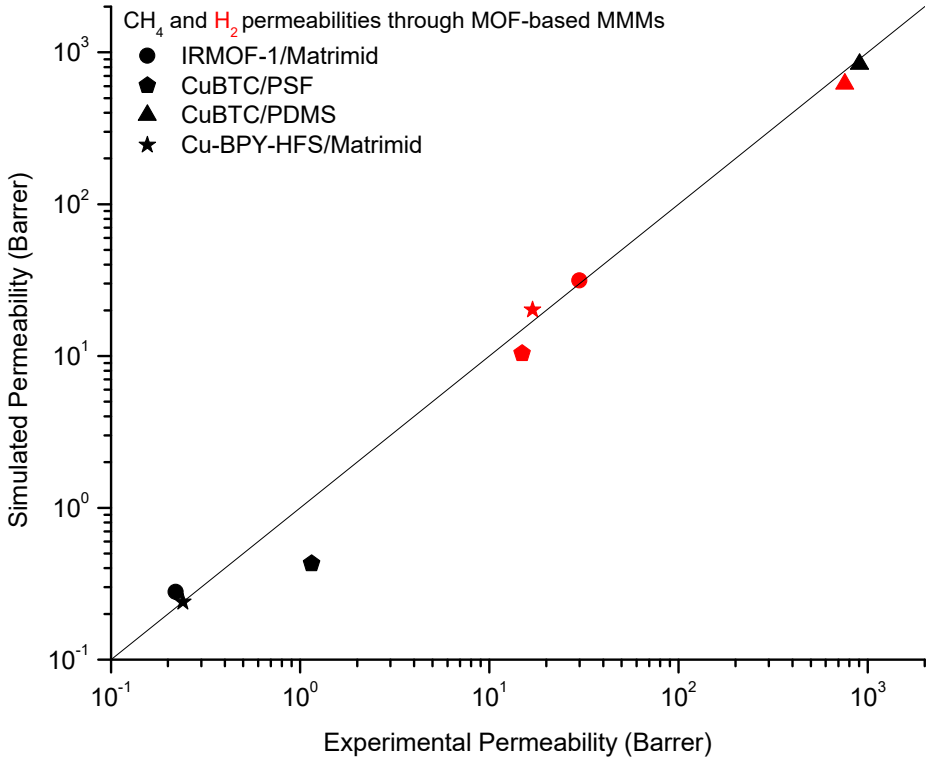

Figure S1. Comparison of our predictions (using modified Felske model)* and experiments for permeabilities of CH_4 and H_2 in IRMOF-1/Matrimid, CuBTC/PSF, CuBTC/PDMS and Cu-BPY-HFS/Matrimid mixed matrix membranes. The loading of the MOFs in the MMMs is 10%. The operating conditions of the membranes are listed in Table 1. *J.D. Felske, *Int. J. Heat Mass Transfer*. 2004, 47, 3453–3461.

Table S1. Properties and permeance measurement conditions of pure MOF membranes and MOF-based mixed matrix membranes

MOF membranes	Measured gas permeances	P_{feed} (bar)	$P_{permeate}$ (bar)	T (°C)	Membrane thickness (μm)	Ref.
IRMOF-1	CH_4, H_2,	1.06	Vacuum	25	25	1
Ni-MOF-74	CH_4	1.8	1	25	25	2
ZIF-69	H_2	1	Vacuum	25	50	3
ZIF-78	H_2	2.5	1	25	25	4
	H_2/CH_4:50/50	2				
ZIF-90	CH_4, H_2	1	Vacuum	200	20	5
	H_2/CH_4:50/50					
ZIF-95	CH_4	1	Vacuum	325	30	4
	H_2/CH_4:50/50					
ZIF-8	H_2	1	Vacuum	25	12	6
	H_2/CH_4:50/50					
	H_2	1	Vacuum	25	15	7
	H_2	1	Vacuum	25	11	8
	H_2	2.76	1.76	25	10	9
IRMOF-1/Matrimid MMM	CH_4, H_2	2	Vacuum	35	35	10
CuBTC/PSF MMM CuBTC/PDMS MMM	CH_4, H_2	-	-	-	-	11
Cu-BPY-HFS/Matrimid MMM	CH_4, H_2	4	Vacuum	35	-	12

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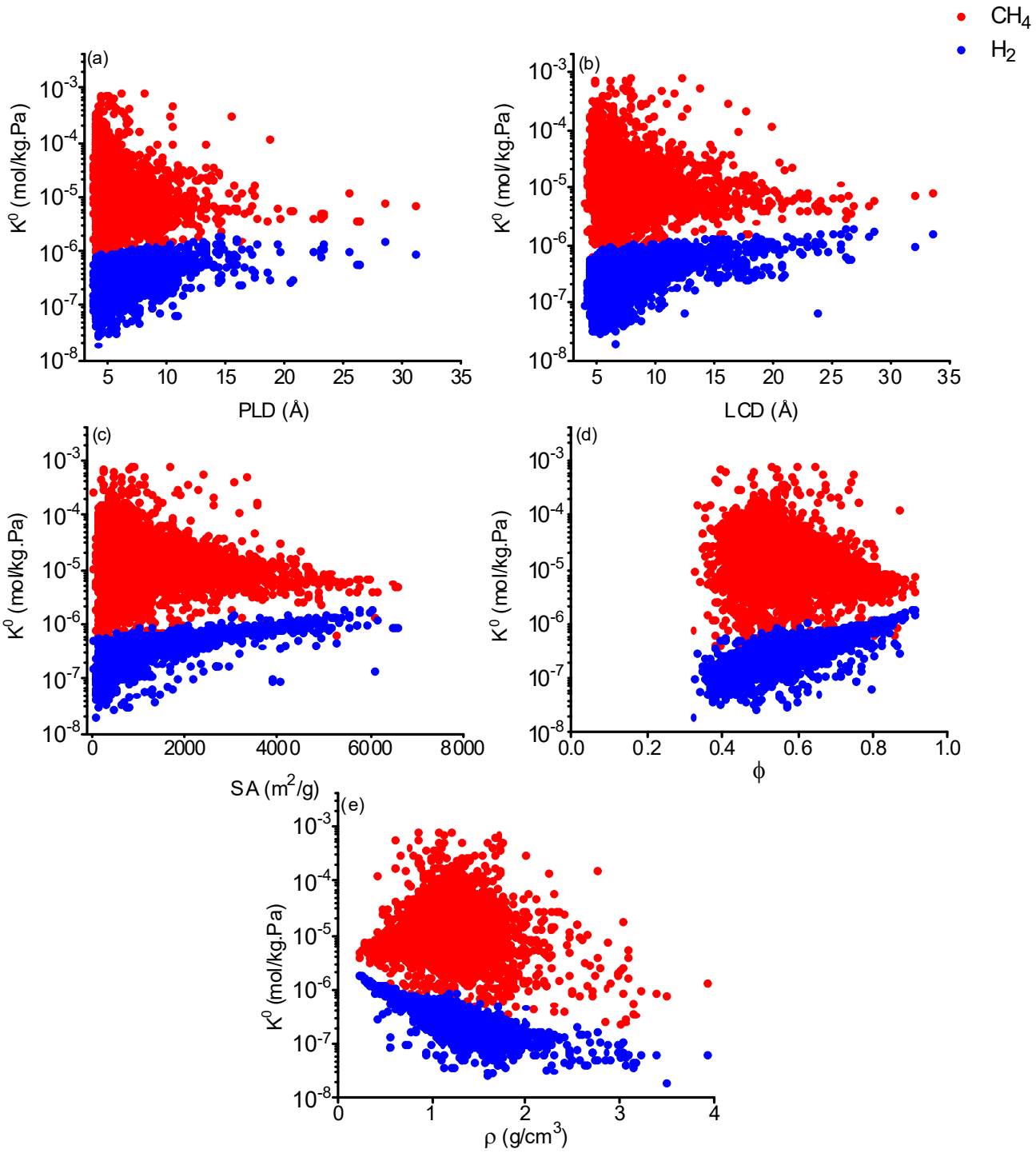


Figure S2. Henry's constant (K^0) versus (a)PLD (b)LCD (c)SA (d)ϕ (e)ρ for H_2 and CH_4 in MOFs.

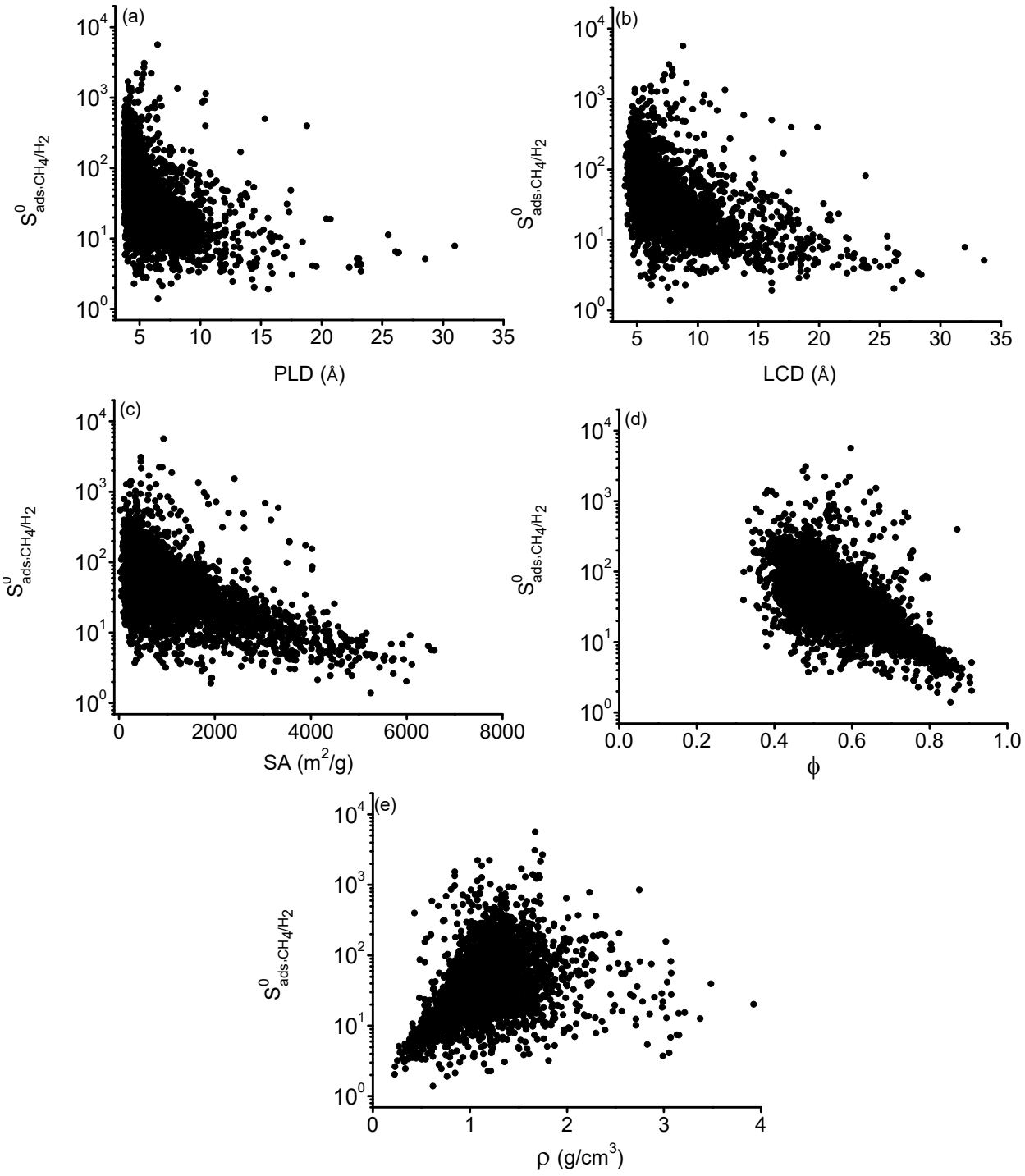


Figure S3. Adsorption selectivity (CH_4/H_2) versus (a)PLD (b)LCD (c)SA (d)ϕ (e)ρ of MOFs.

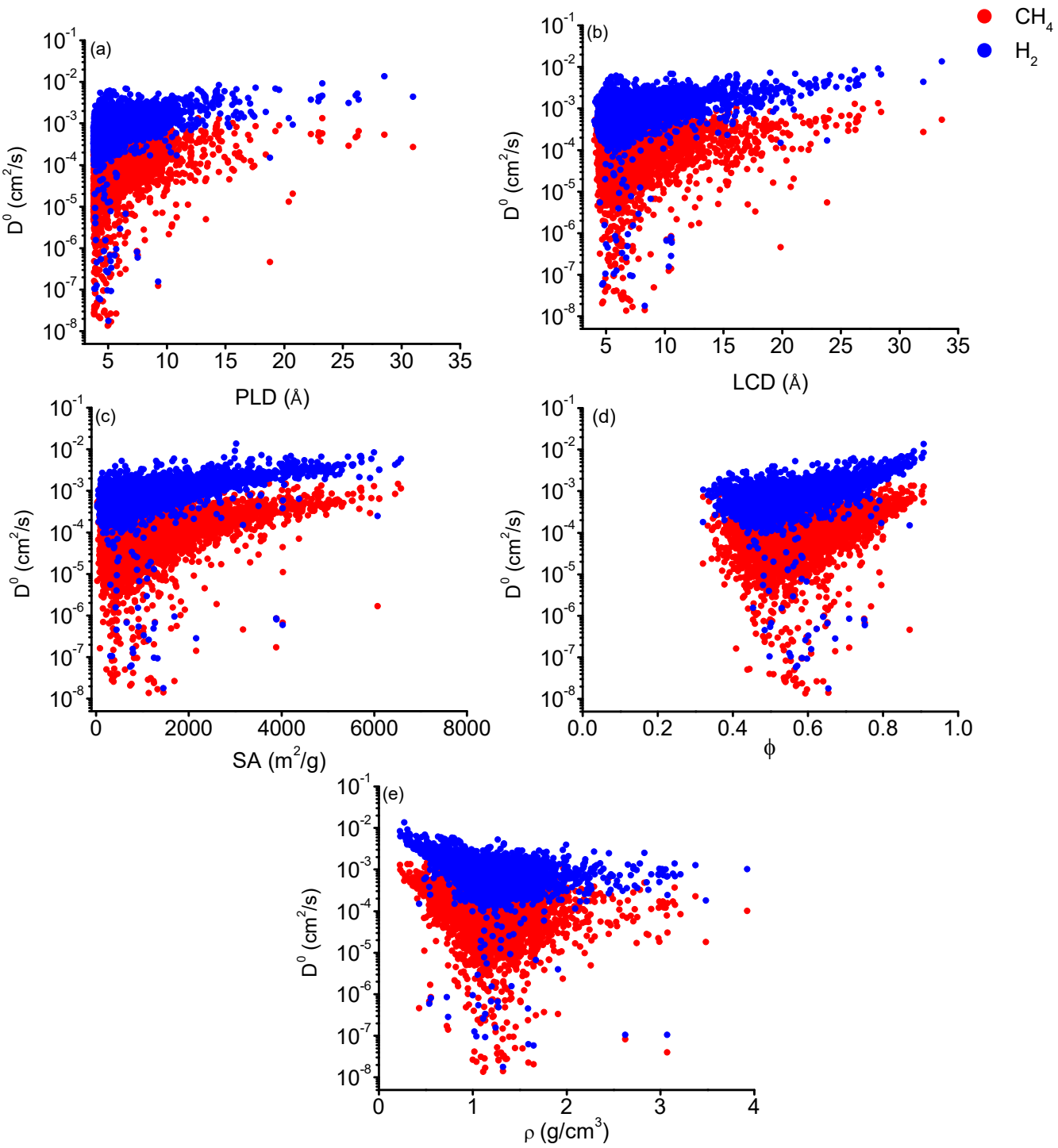


Figure S4. Diffusivities of gases versus (a)PLD (b)LCD (c)SA (d)ϕ (e)ρ in MOFs.

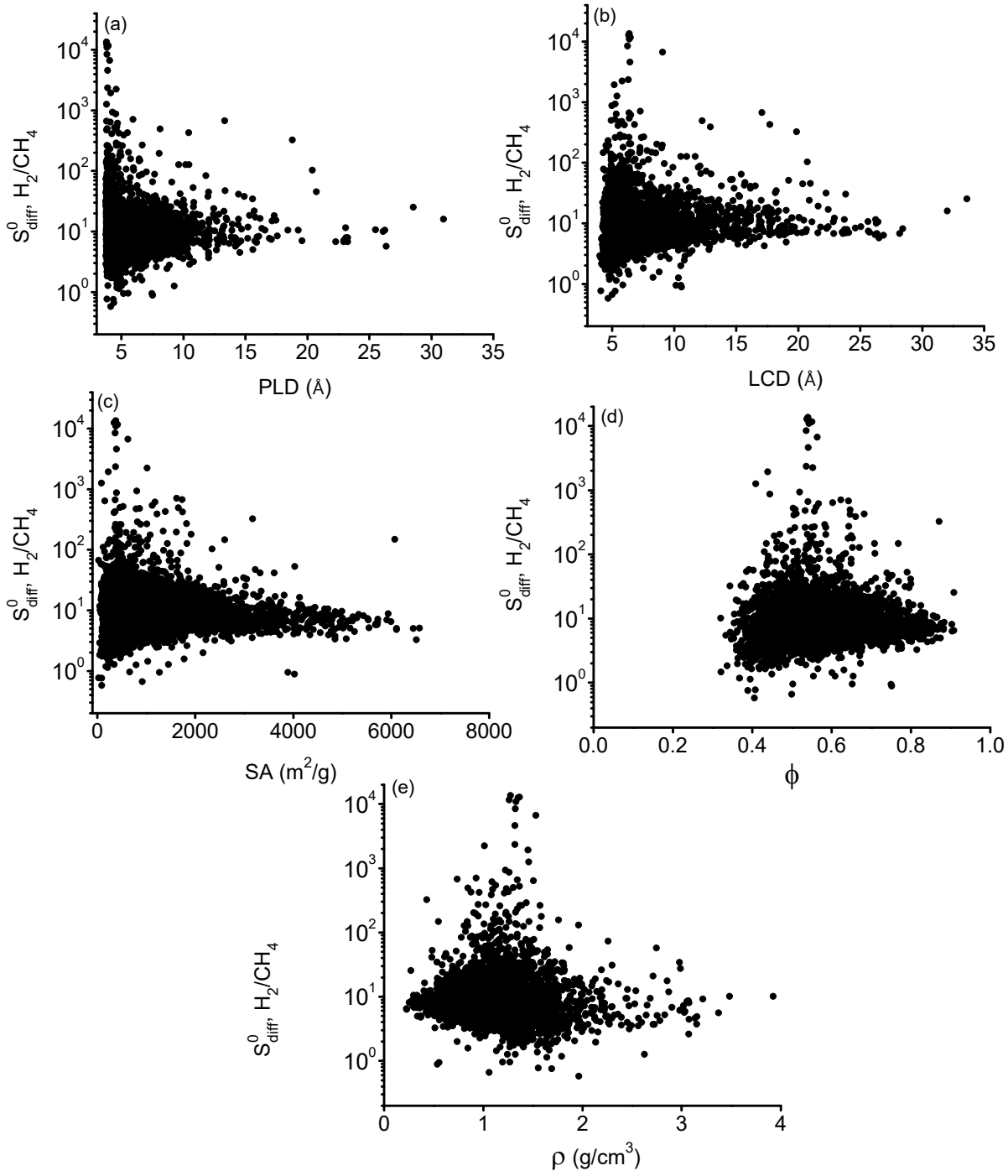


Figure S5. Diffusion selectivity (H_2/CH_4) versus (a)PLD (b)LCD (c)SA (d)ϕ (e)ρ of MOFs.

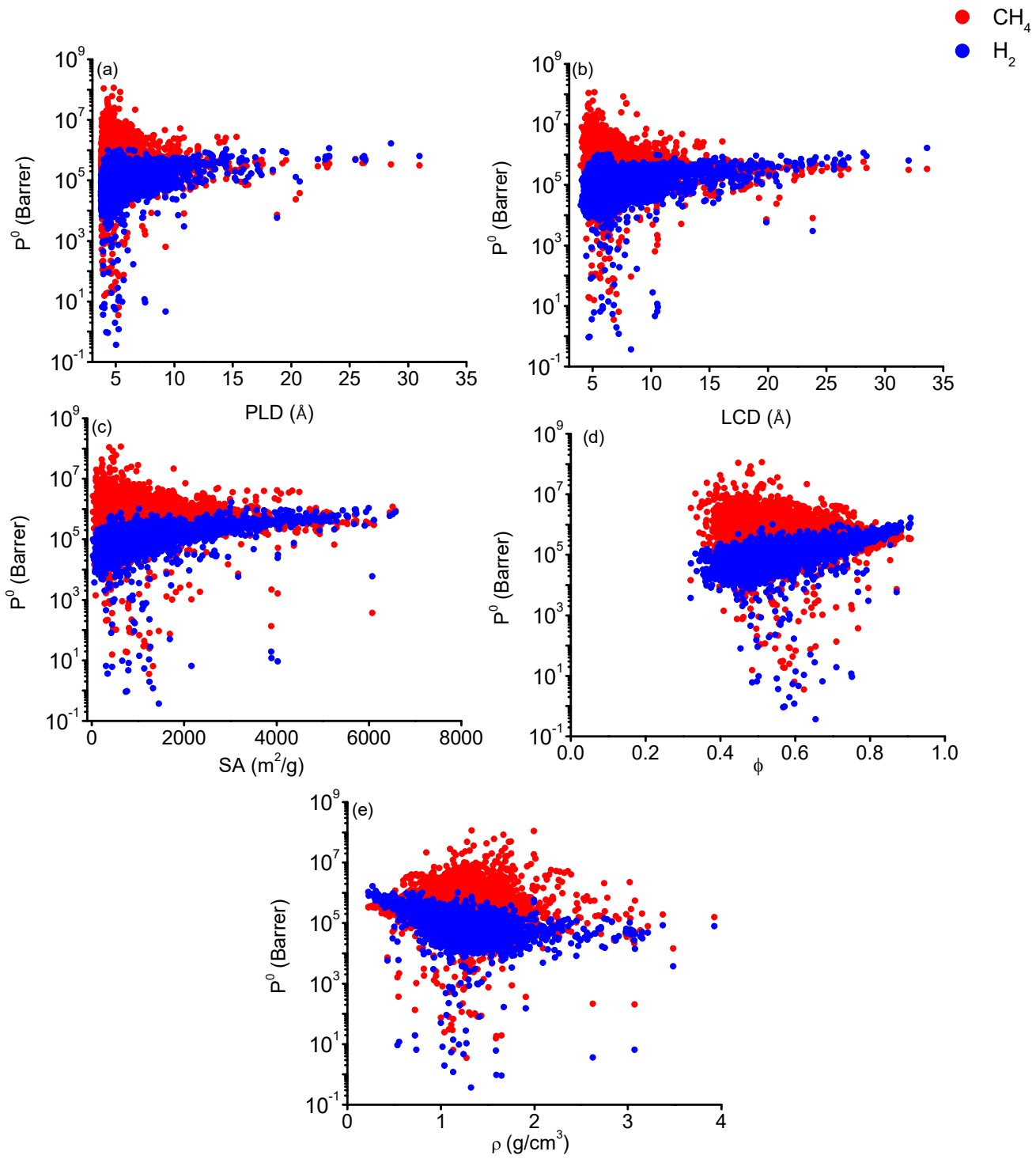


Figure S6. Permeabilities of gases versus (a)PLD (b)LCD (c)SA (d)ϕ (e)ρ in MOFs.

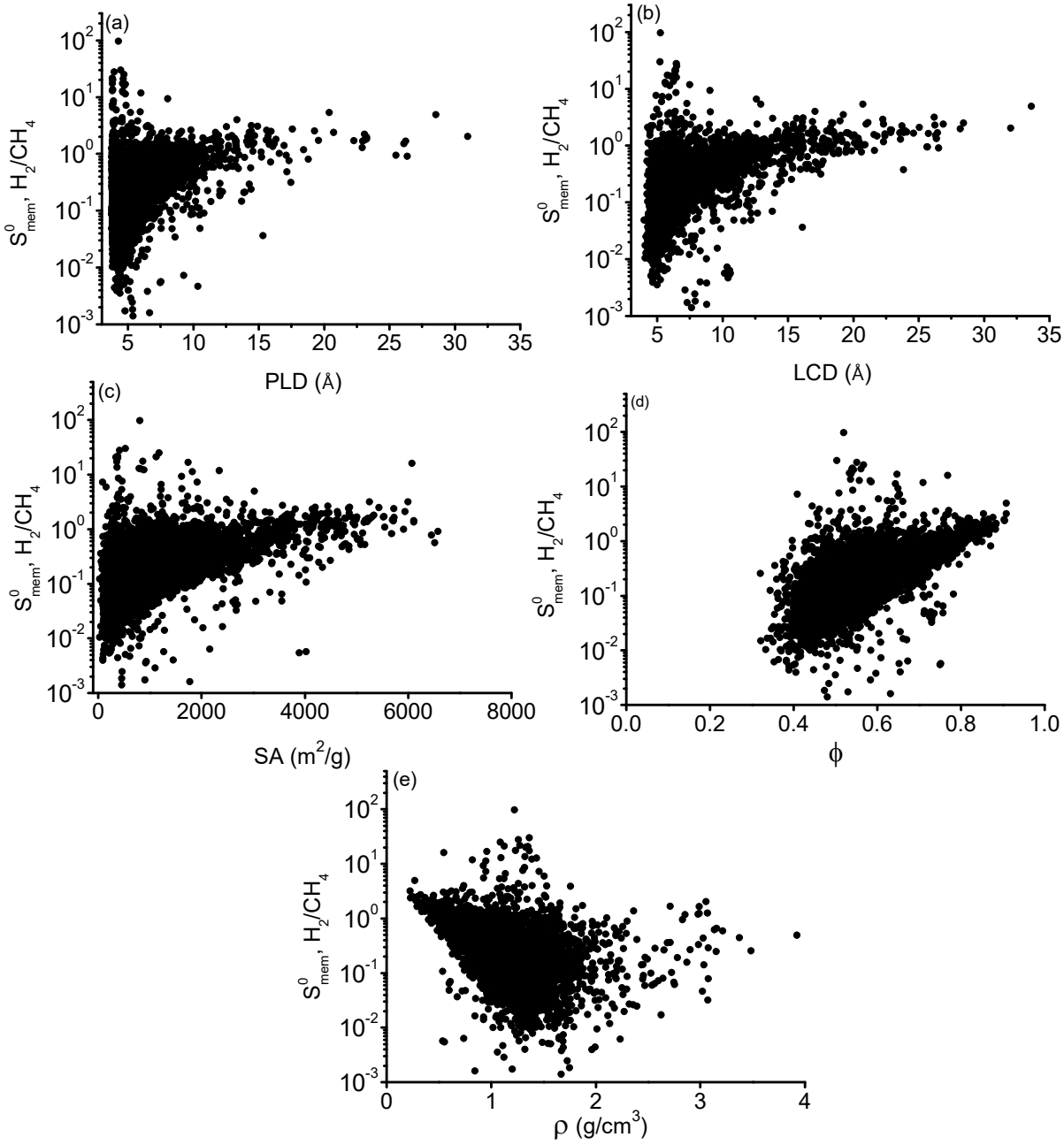
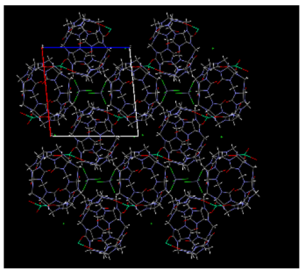
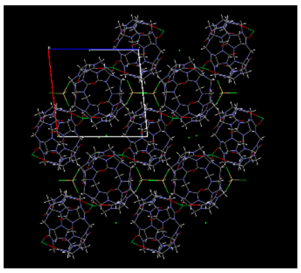


Figure S7. Membrane selectivity (H_2/CH_4) versus (a)PLD (b)LCD (c)SA (d)ϕ (e)ρ of MOFs.

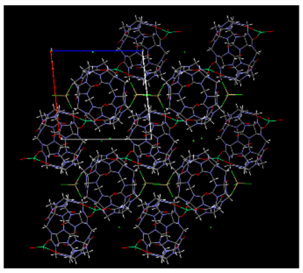
	MEFMEQ	OGAJEN	OGAMOA	OGALEP	OGALUF	OGAKUE	PIZHOX	OGAJAJ	HIFVUO	QONKOV
MEFMEQ	1.00	0.83	0.84	0.84	0.84	0.84	0.67	0.84	0.49	0.66
OGAJEN	0.83	1.00	0.85	0.86	0.86	0.86	0.70	0.86	0.50	0.68
OGAMOA	0.84	0.85	1.00	0.88	0.87	0.88	0.68	0.87	0.49	0.65
OGALEP	0.84	0.86	0.88	1.00	0.87	0.89	0.68	0.87	0.49	0.66
OGALUF	0.84	0.86	0.87	0.87	1.00	0.88	0.68	0.86	0.49	0.67
OGAKUE	0.84	0.86	0.88	0.89	0.88	1.00	0.68	0.88	0.49	0.65
PIZHOX	0.67	0.70	0.68	0.68	0.68	0.68	1.00	0.67	0.63	0.83
OGAJAJ	0.84	0.86	0.87	0.87	0.86	0.88	0.67	1.00	0.48	0.65
HIFVUO	0.49	0.50	0.49	0.49	0.49	0.49	0.63	0.48	1.00	0.65
QONKOV	0.66	0.68	0.65	0.66	0.67	0.65	0.83	0.65	0.65	1.00



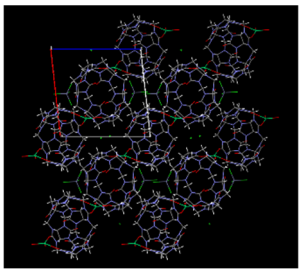
OGAJEN



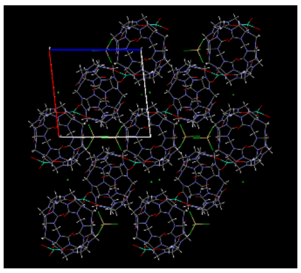
OGAKUE



OGALEP



OGAJAJ



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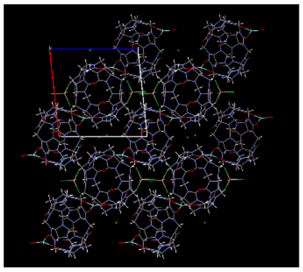

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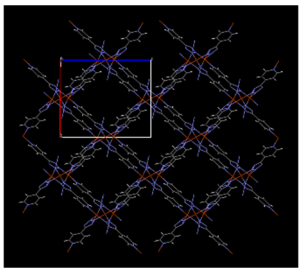
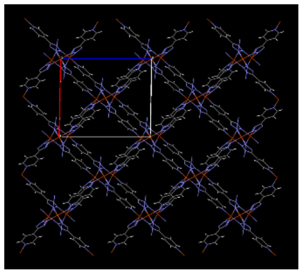
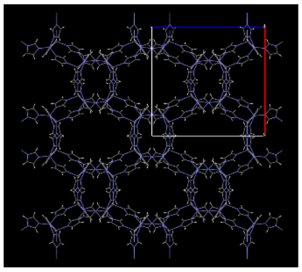


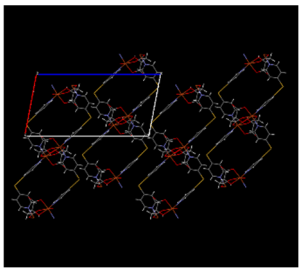
PIZHOX



QONKOV



HIFVUO



MEFMEQ

Figure S8. (top)Structural similarity indexes of the top ten most promising MOF membranes. Green, light green, yellow and orange colors correspond to the most similar materials, highly similar, lightly similar and dissimilar materials having similarity index of 0.8-1.0, 0.7-0.8, 0.5-0.7 and 0.3-0.5. (bottom)2×2×2 unit cell representations of the top ten most promising MOFs in (010) direction.

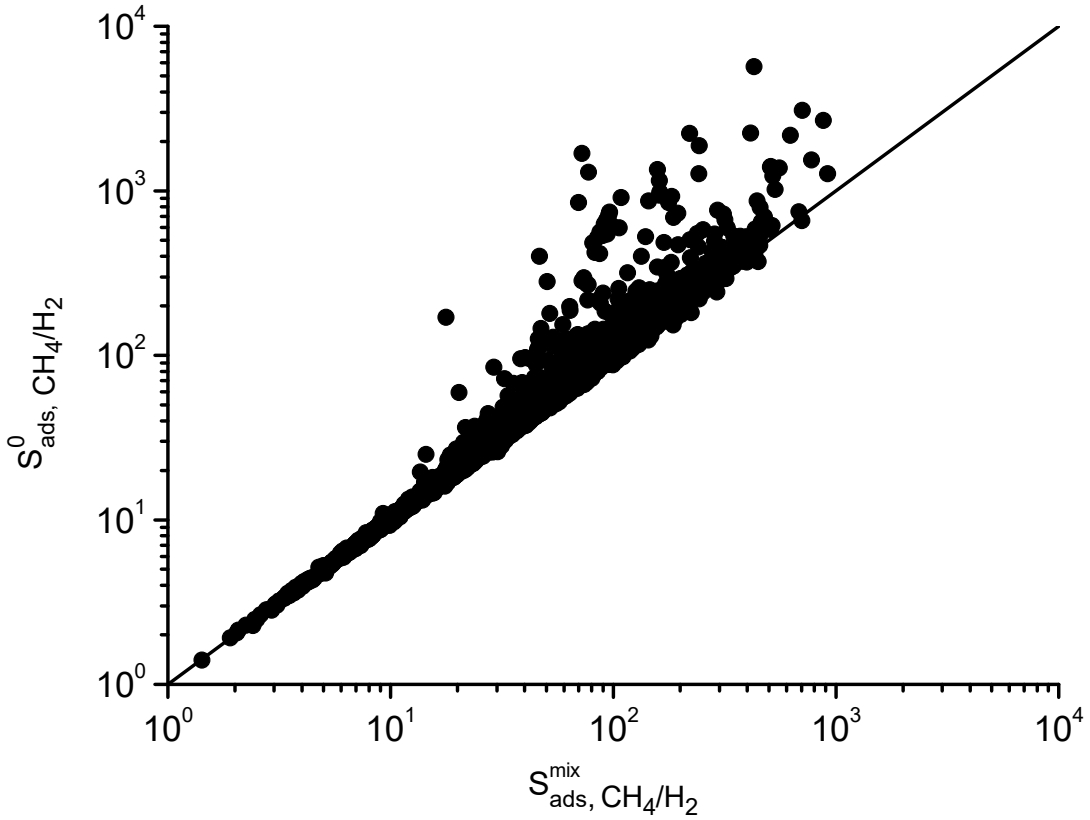


Figure S9. Comparison of $S^0_{ads,CH_4/H_2}$ with $S^{mix}_{ads,CH_4/H_2}$ computed at 1 bar for 4,240 MOFs.

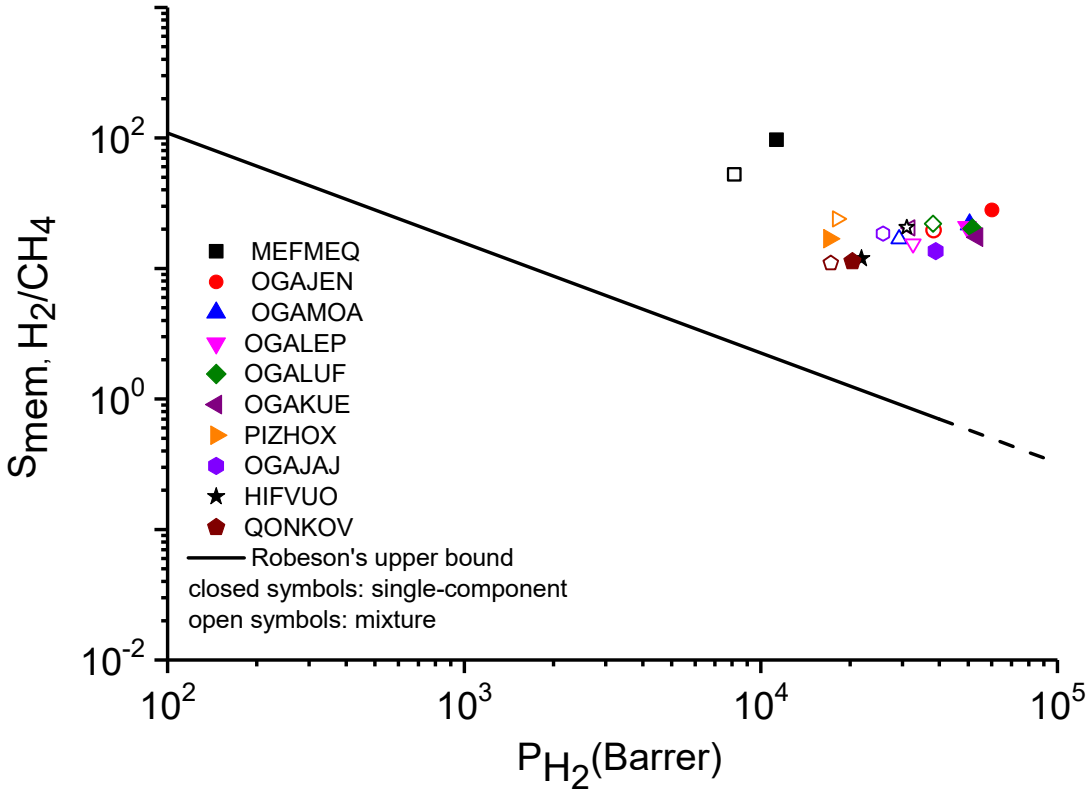


Figure S10. Comparison of the single-component and mixture calculations for permeabilities and selectivities of the top ten promising MOF membranes.