Electronic Supplementary Material (ESI) for Molecular Systems Design & Engineering.

Supporting Information

Role of Partial Charge Assignment Methods in High-Throughput Screening of MOF Adsorbents and Membranes for CO_2/CH_4 Separations

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Table S1. Potentials used for gas molecules in molecular simulations.

Gas molecule	Atom	ε (K)	σ (Å)	q(e^-)
CO_2	C	27.0	2.80	0.70
	O	79.0	3.05	-0.35
CH_4	single sphere	148.0	3.73	0.0

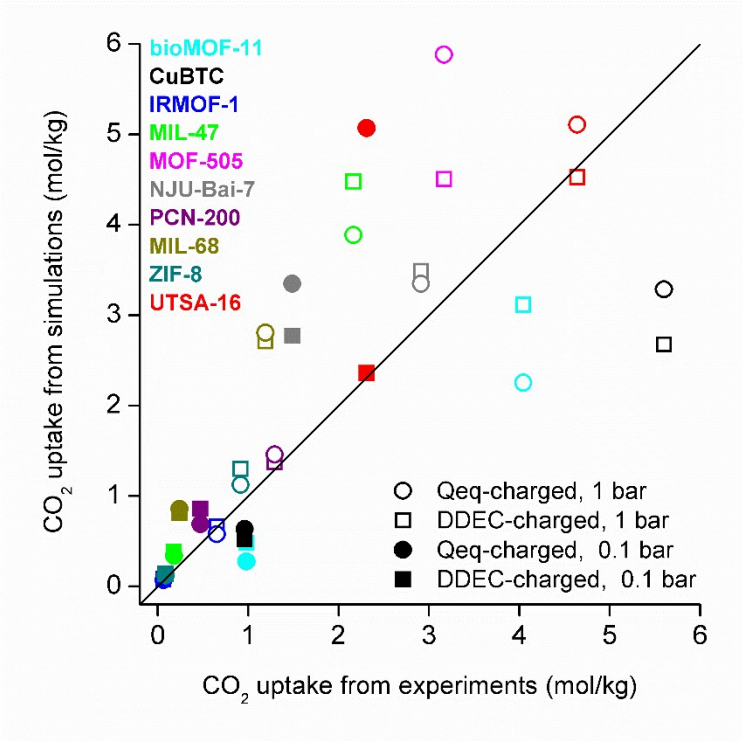

Figure S1. Comparison of the experimental CO_2 uptakes[1-7] with our simulation results obtained using different charge assignment methods. Squares (circles) represent the simulation results of DDEC (Qeq)-charged MOFs while filled (empty) symbols are the results for 0.1 (1) bar.

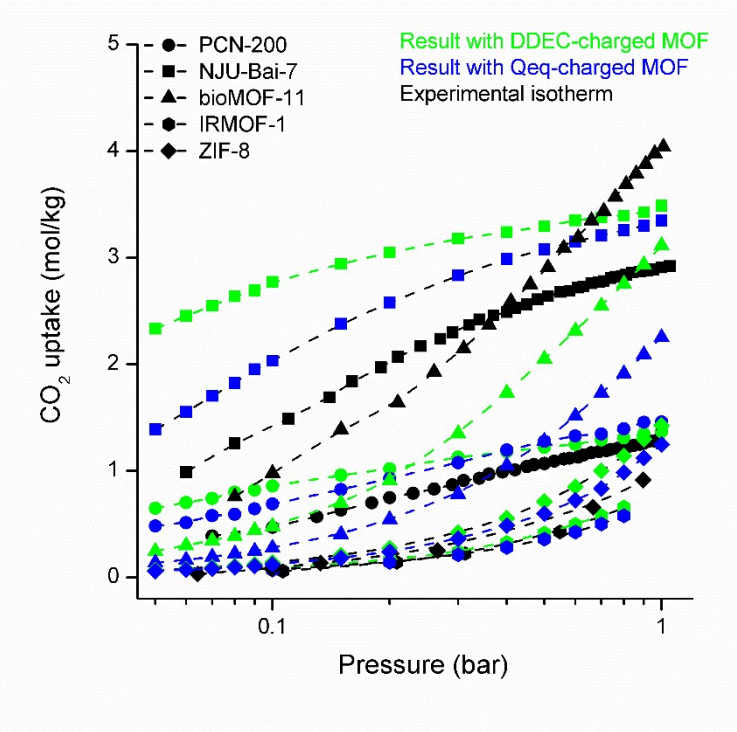

Figure S2. Comparison of CO_2 adsorption isotherms obtained for DDEC-charged and Qeq-charged MOFs at 298 K.

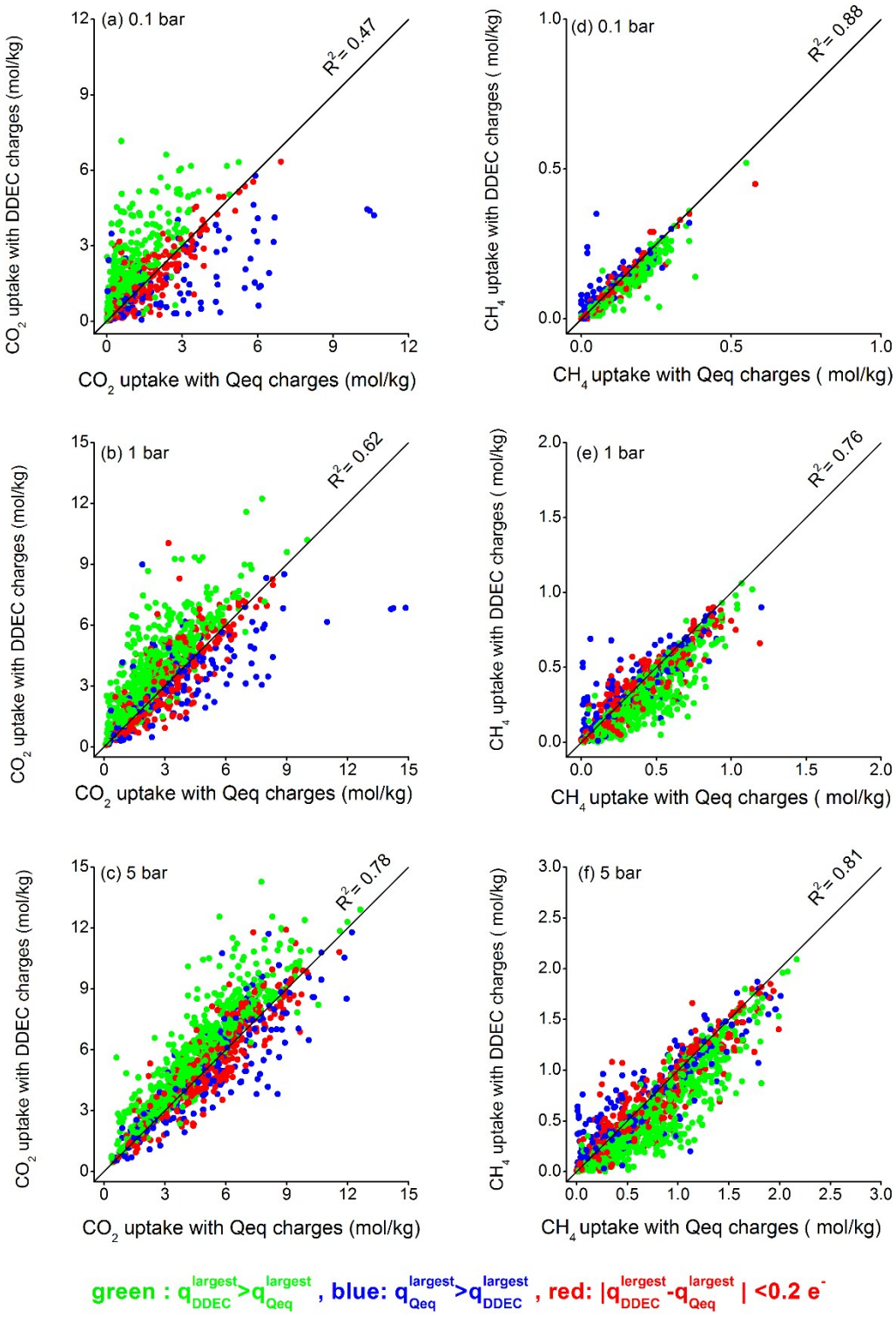

Figure S3. CO_2 uptakes (a-c) and CH_4 uptakes (b-f) obtained from simulations using Qeq and DDEC charges at 0.1, 1, 5 bar for CO_2/CH_4: 50/50 mixture.

Table S2. Range of calculated adsorbent evaluation metrics.

Composition	Adsorbent metric	$Range_{DDEC}$ ($Range_{Qeq}$) P_{ads} :1 bar, P_{des}: 0.1 bar	$Range_{DDEC}$ ($Range_{Qeq}$) P_{ads} :5 bar, P_{des}:1 bar
CO_2/CH_4: 10/90	ΔN_{CO_2} (mol/kg)	0.02-5.64 (0.01-5.51)	0.02-8.07 (0.02-5.43)
	S_{ads}	1.62-1633.80 (1.23-2477.74)	1.54-1068.04 (1.14-1329.03)
	APS (mol/kg)	0.03-2179.55 (0.02-2715.96)	0.03-846.23 (0.04-1910.94)
	R%	9.14-92.22 (3.93- 90.58)	2.52-82.07 (1.77-81.05)
CO_2/CH_4: 50/50	ΔN_{CO_2}(mol/kg)	0.09-10.17 (0.06-6.97)	0.03-8.30 (0.10-10.26)
	S_{ads}	1.66-1068.28 (1.02-1215.28)	1.54-883.75 (0.97-1009.64)
	APS (mol/kg)	0.17-1282.70 (0.11-3026)	0.20-706.53 (0.13-1315.55)
	R%	4.49-93.31 (3.65-91.02)	1.43-85.19 (2.42-83.74)

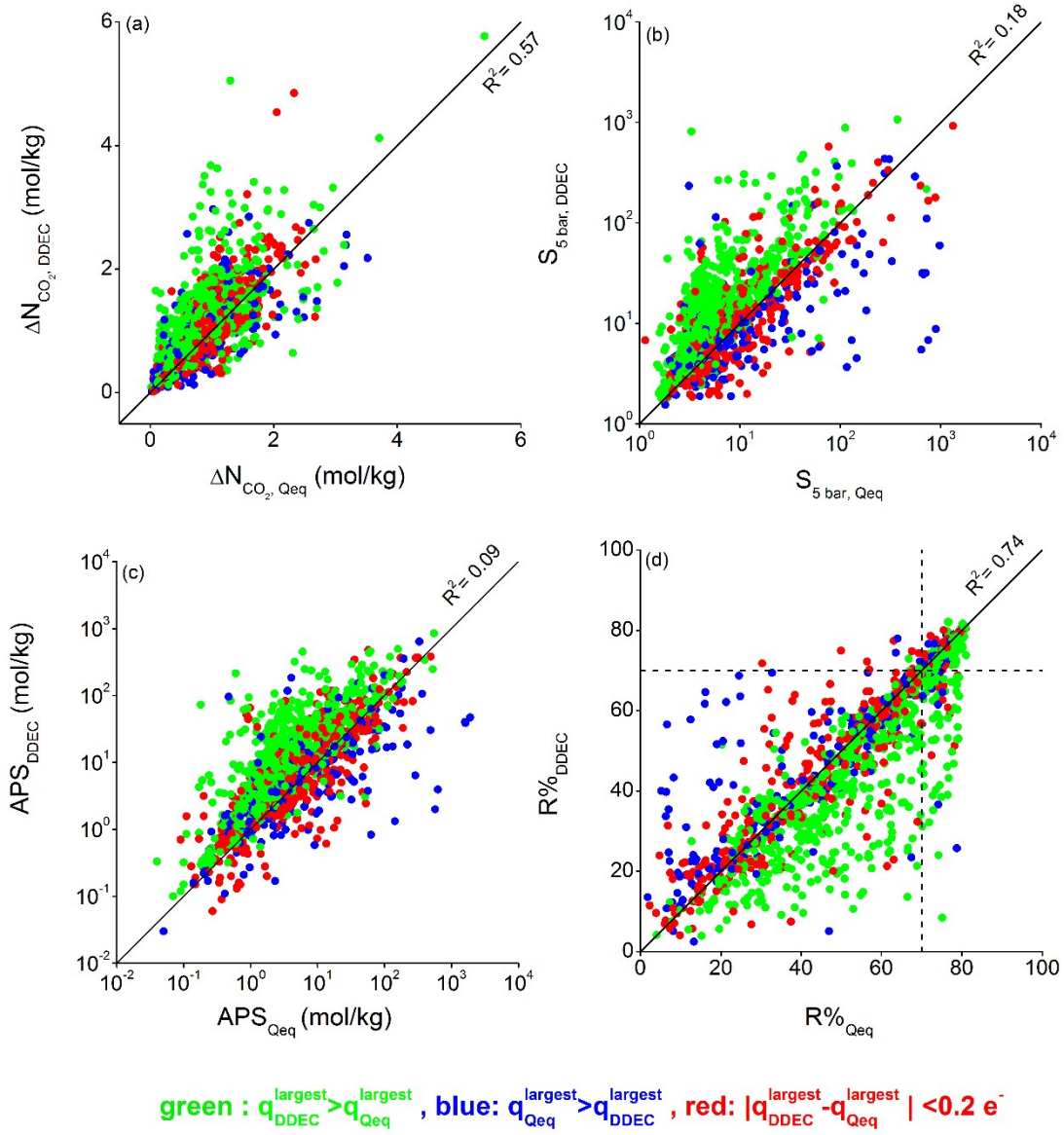

Figure S4. Calculated adsorbent performance evaluation metrics using Qeq and DDEC charges at PSA condition, 298 K for CO_2/CH_4:10/90 mixture; (a) $^{\Delta N_{CO_2}}$, (b) S_{ads}, (c) APS, (d) R%. Dashed lines in (d) show R% =70%.

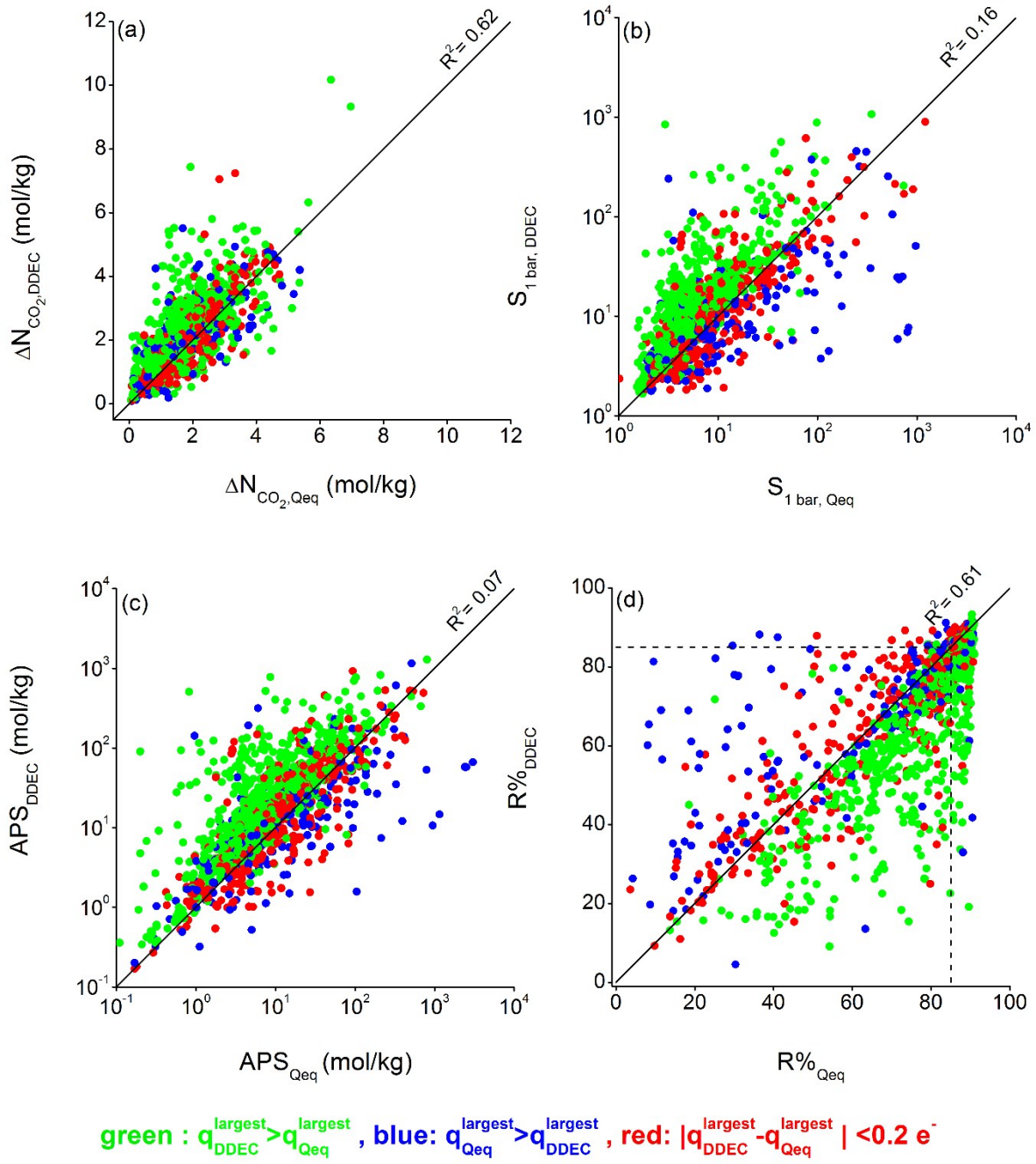

Figure S5. Calculated adsorbent performance evaluation metrics using Qeq and DDEC charges at VSA condition, 298 K for CO_2/CH_4:50/50 mixture; (a) $^{\Delta N_{CO_2}}$, (b) S_{ads}, (c) APS, (d) R%. Dashed lines in (d) show R% =85%.

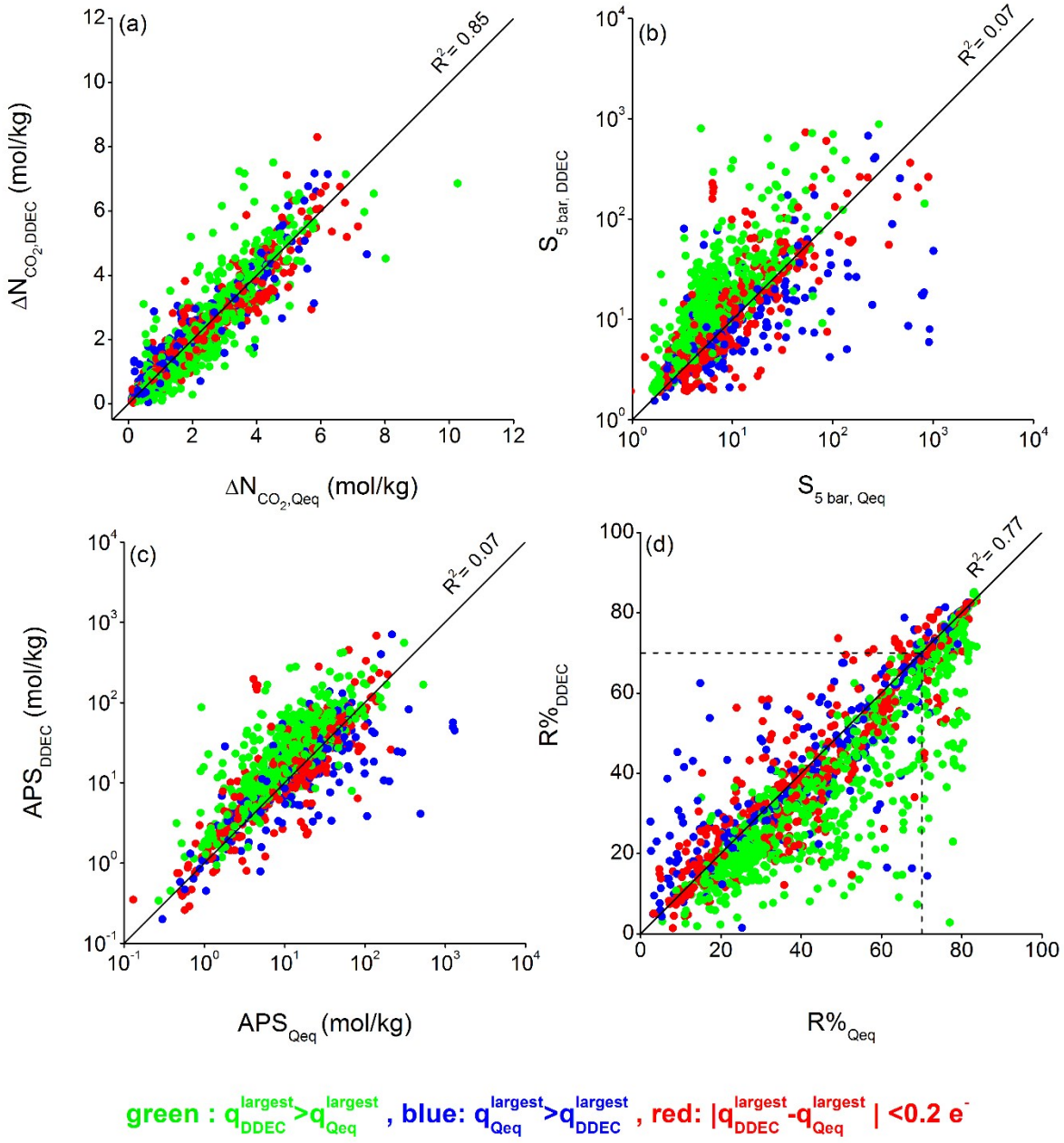

Figure S6. Calculated adsorbent performance evaluation metrics using Qeq and DDEC charges at PSA condition, 298 K for CO_2/CH_4:50/50 mixture; (a) $^{\Delta N_{CO_2}}$, (b) S_{ads}, (c) APS, (d) R%. Dashed lines in (d) show R% =70%.

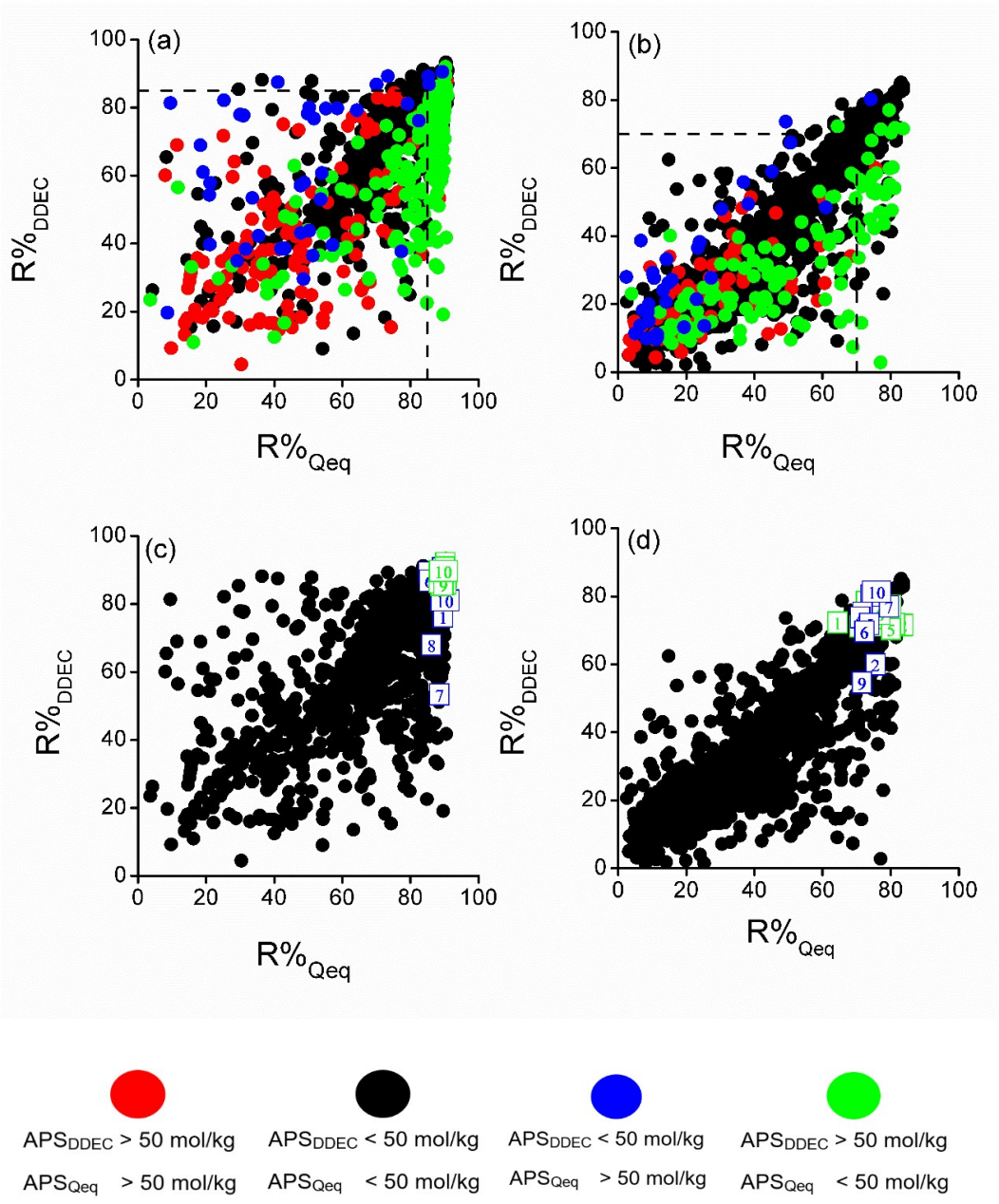

Figure S7. Comparison of R% of DDEC and Qeq-charged MOFs at (a, c) VSA (b, d) PSA conditions for CO_2/CH_4: 50/50 mixture. Dashed lines show R%=85% in (a), R%=70% in (b). The top 10 MOFs identified by DDEC and Qeq methods are shown with green and blue, respectively.

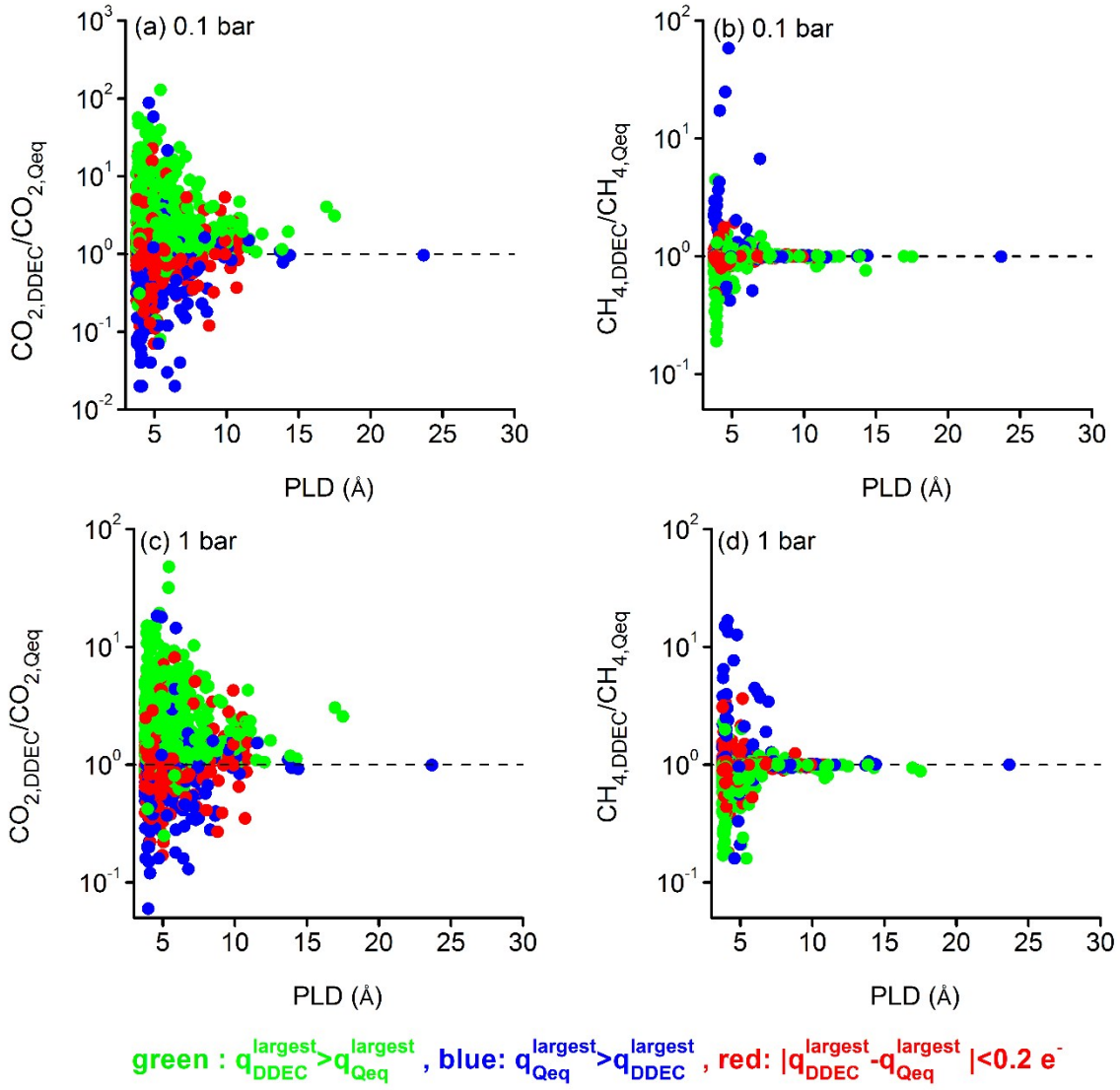

Figure S8. The ratio of (a, c) CO_2 uptakes (b, d) CH_4 uptakes computed by DDEC-charged to the ones computed by Qeq-charged MOFs as a function of PLD.

Table S3. Calculated uptakes and diffusivities of CO_2 and CH_4 in DDEC-charged and Qeq-charged MOFs; adsorption, diffusion and membrane selectivities of MOFs for separation of CO_2/CH_4 mixture.

MOFs	PLD (Å)	N_{CO_2} (mol/kg)	N_{CH_4} (mol/kg)	$D_{self,\ CO_2}$ (cm^2/s)	$D_{self,\ CH_4}$ (cm^2/s)	S_{ads}	S_{diff}	S_{mem}
		DDEC (Qeq)	DDEC (Qeq)	DDEC (Qeq)	DDEC (Qeq)	DDEC (Qeq)	DDEC (Qeq)	DDEC (Qeq)
REFTUT	5.82	1.70 (0.21)	0.19 (0.35)	4.55×10^{-6} (3.50×10^{-5})	9.83×10^{-6} (3.74×10^{-5})	81.96 (5.34)	0.47 (0.94)	38.71 (5.03)
GIQYIP	3.91	4.69 (6.88)	0.13 (0.17)	4.17×10^{-7} (4.96×10^{-7})	5.20×10^{-6} (2.13×10^{-6})	318.39 (362.88)	0.09 (0.25)	29.74 (91.66)
LIFWEE	3.87	1.18 (1.39)	0.71 (0.69)	6.80×10^{-6} (6.17×10^{-6})	5.93×10^{-6} (3.49×10^{-6})	14.83 (18.20)	1.15 (1.82)	17.13 (33.18)
CAZGIT	3.98	0.50 (1.68)	0.85 (0.25)	2.53×10^{-6} (4.78×10^{-7})	2.08×10^{-6} (2.69×10^{-7})	5.31 (59.52)	1.27 (2.12)	6.75 (126.11)
ATAFIK	5.70	2.82 (0.32)	0.74 (0.88)	2.04×10^{-5} (1.41×10^{-4})	1.08×10^{-4} (2.98×10^{-4})	34.45 (3.26)	0.19 (0.48)	6.55 (1.55)
ACODED	3.79	1.28 (0.83)	1.70 (1.86)	6.23×10^{-6} (9.90×10^{-6})	8.31×10^{-6} (1.08×10^{-5})	6.81 (4.03)	0.76 (0.91)	5.17 (3.68)
PEPBAO	3.96	0.55 (0.31)	0.97 (1.10)	1.87×10^{-6} (2.75×10^{-6})	2.42×10^{-6} (3.35×10^{-6})	5.09 (2.57)	0.77 (0.82)	3.93 (2.12)
Co26NDP	10.60	0.48 (0.36)	1.11 (1.11)	4.51×10^{-5} (6.82×10^{-5})	1.07×10^{-4} (1.06×10^{-4})	3.87 (2.92)	0.42 (0.64)	1.64 (1.87)

LOQLIN	13.93	1.00 (1.06)	1.26 (1.18)	2.42×10^{-5} (3.06×10^{-5})	1.28×10^{-4} (1.40×10^{-4})	7.11 (8.08)	0.19 (0.22)	1.35 (1.75)
KIGCEK	10.28	0.17 (0.26)	0.59 (0.59)	4.67×10^{-5} (3.80×10^{-5})	1.16×10^{-4} (1.08×10^{-4})	2.54 (3.93)	0.40 (0.35)	1.02 (1.38)

References

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