Electronic supplementary information

High-throughput computational screening of 137953 metal-organic frameworks for membrane separation of $CO_2/N_2/CH_4$ mixture

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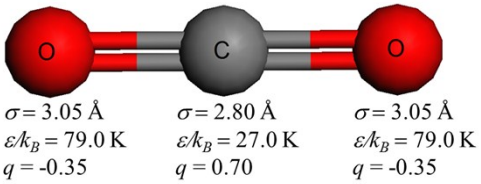
1. Molecular models

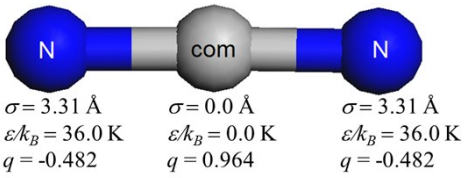
Table S1 Lennard Jones parameters of MOFs.

Atoms	C	O	H	N	F	Cl	Br	Zn	Cu	Zr	V
σ (Å)	3.43	3.12	2.57	3.26	2.997	3.517	3.73	2.46	3.114	2.783	2.80
ε/k_B (K)	52.83	30.19	22.14	34.72	25.16	114.23	126.3	62.40	2.516	34.72	8.05

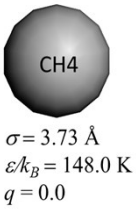
From A. K. Rappe, C. J. Casewit, K. S. Colwell, W. A. Goddard, W. M. Skiff, UFF, a Full Periodic Table Force Field for Molecular Mechanics and Molecular Dynamics Simulations. *J. Am. Chem. Soc.* 1992, **114**, 10024-10035.



Three-site model of CO_2



Three-site model of N_2



United-atom model of CH_4

Fig. S1 Lennard-Jones parameters and charges of CO_2, N_2 and CH_4.

From J. J. Potoff, J. I. Siepmann, Vapor–Liquid Equilibria of Mixtures Containing Alkanes, Carbon Dioxide and Nitrogen. *AIChE J.* 2001, **47**, 1676-1682.

2. Pore limiting diameters

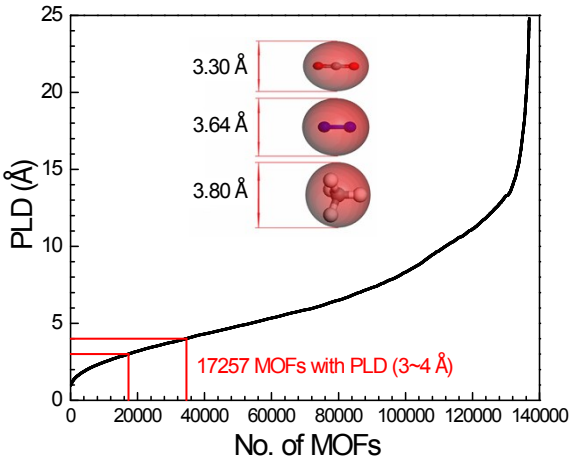


Fig. S2 PLDs of 137953 MOFs. There are 17257 MOFs with PLD between 3 ~ 4 Å.

3. Diffusion of CO_2, N_2 and CH_4 at infinite dilution in a MOF with PLD = 3.2 Å

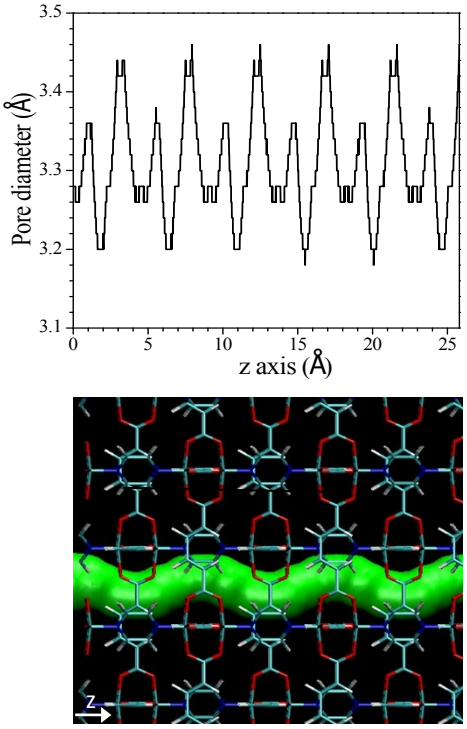


Fig. S3 Pore diameter and morphology along z-axis in a MOF (ID: 31136) with PLD of 3.2 Å.

MOF_31136+CO2.mp4, MOF_31136+N2.mp4 and MOF_31136+CH4.mp4 visualize the diffusion of CO_2, N_2 and CH_4 in a MOF (ID: 31136) at infinite dilution. In each video, the number of gas molecules is 30; however, there is no gas-gas intermolecular interaction, thus corresponding to infinite dilution.

4. Percentage of pore size distribution between d_1 and d_2

As illustrated in Fig. S4, the percentage of pore size distribution (PSD) between d_1 and d_2 is defined as

$$\mathrm{PSD}\%_{(d_1 \sim d_2)} = A_{12}/A_{\mathrm{total}} \times 100\%$$

where A_{12} is the area for pore size between d_1 and d_2, and A_{total} is the total area under the entire PSD curve.

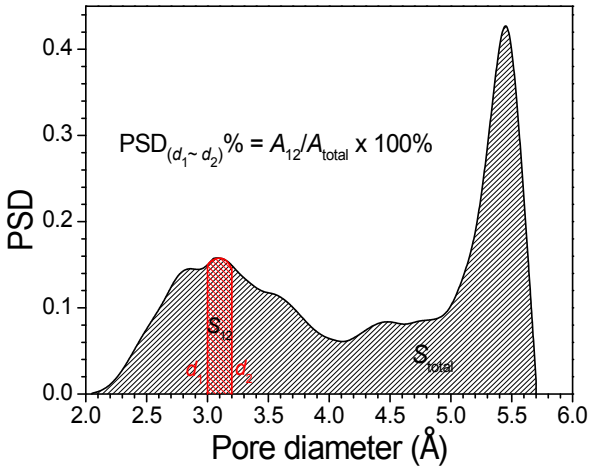


Fig. S4 Pore size distribution between d_1 and d_2.

5. Diffusivity and diffusion selectivity versus density, porosity and VSA

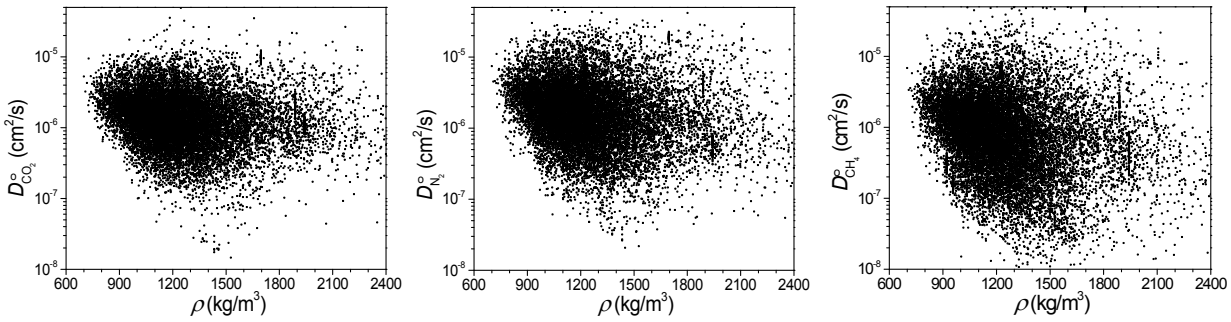


Fig. S5.1 Diffusivity versus density.

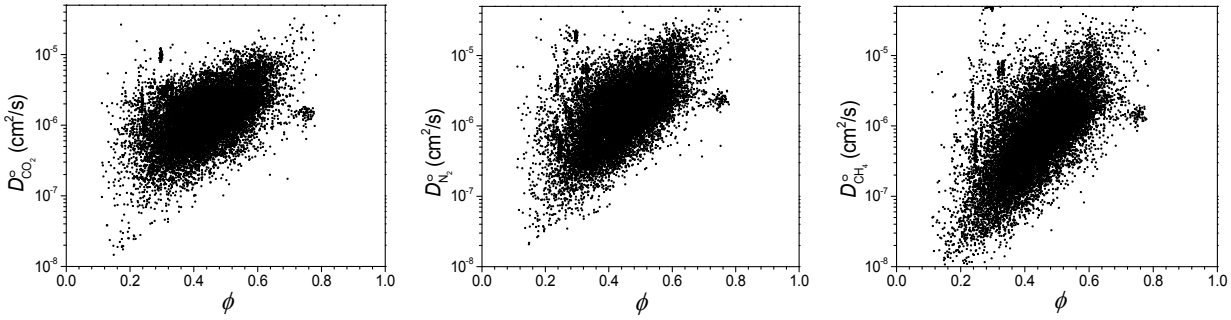


Fig. S5.2 Diffusivity versus porosity.

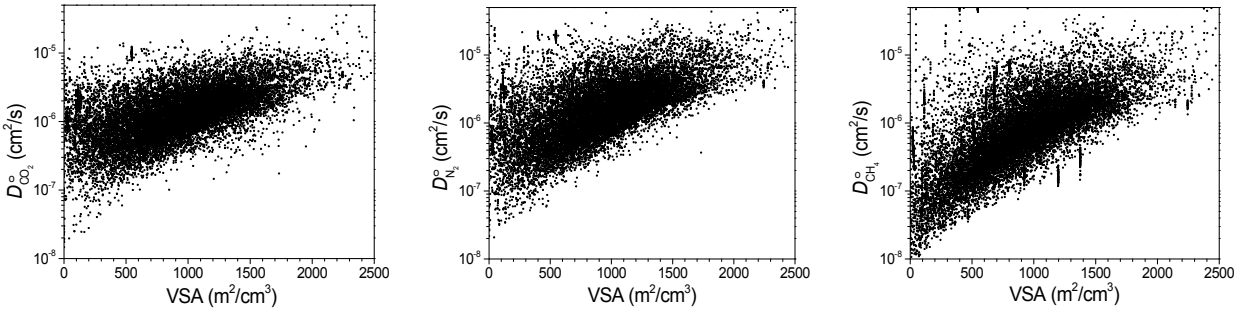


Fig. S5.3 Diffusivity versus VSA.

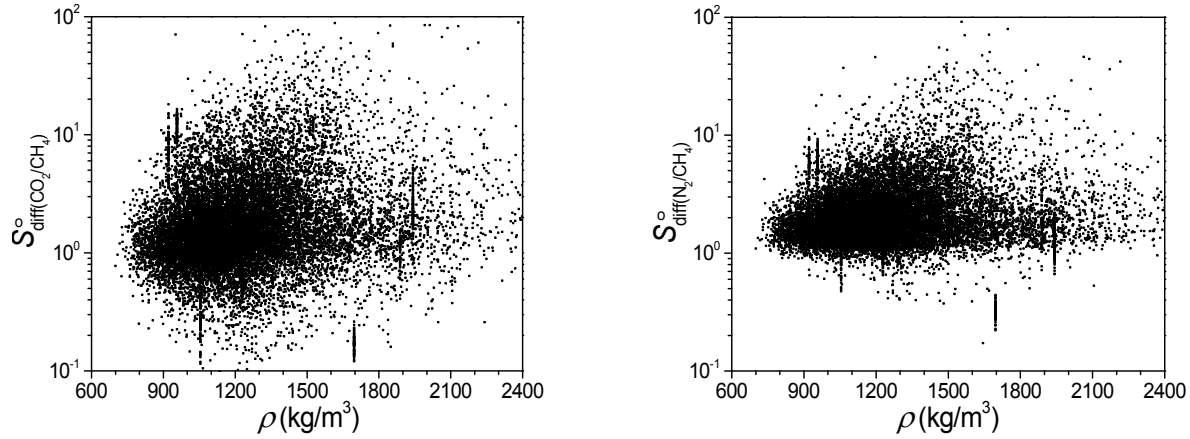


Fig. S6.1 Diffusion selectivity versus density for CO_2/CH_4 and N_2/CH_4.

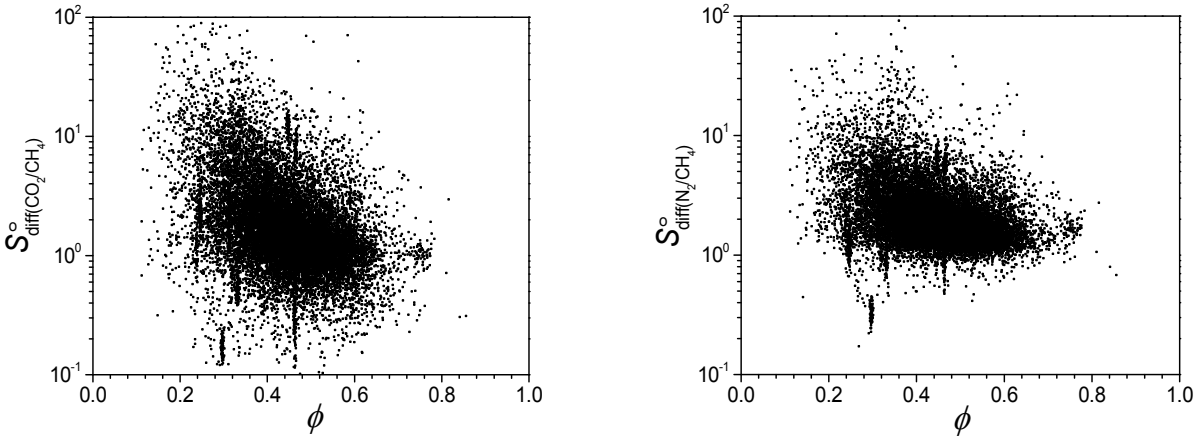


Fig. S6.2 Diffusion selectivity versus porosity for CO_2/CH_4 and N_2/CH_4.

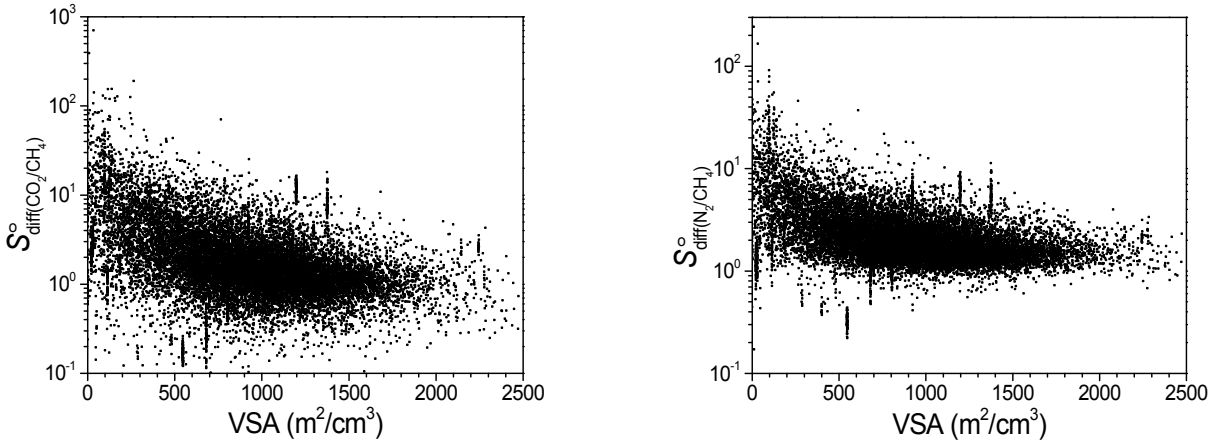


Fig. S6.3 Diffusion selectivity versus VSA for CO_2/CH_4 and N_2/CH_4.

6. Permeation and permselectivity versus density, porosity and VSA

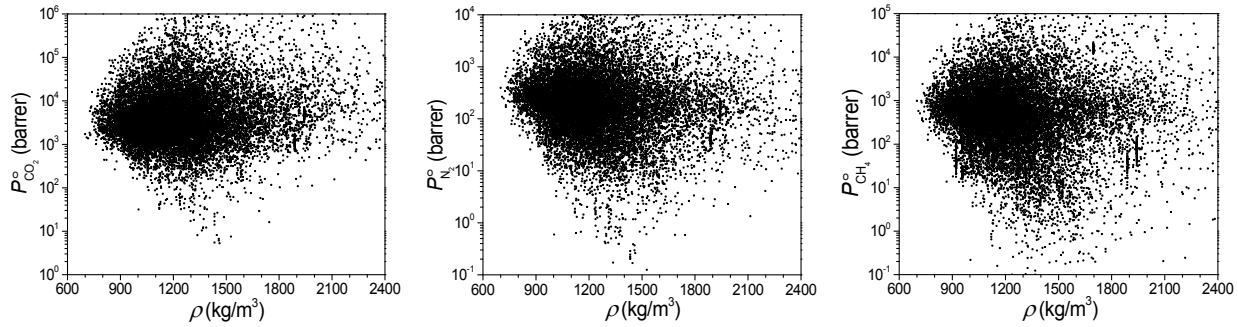


Fig. S7.1 Permeability versus density.

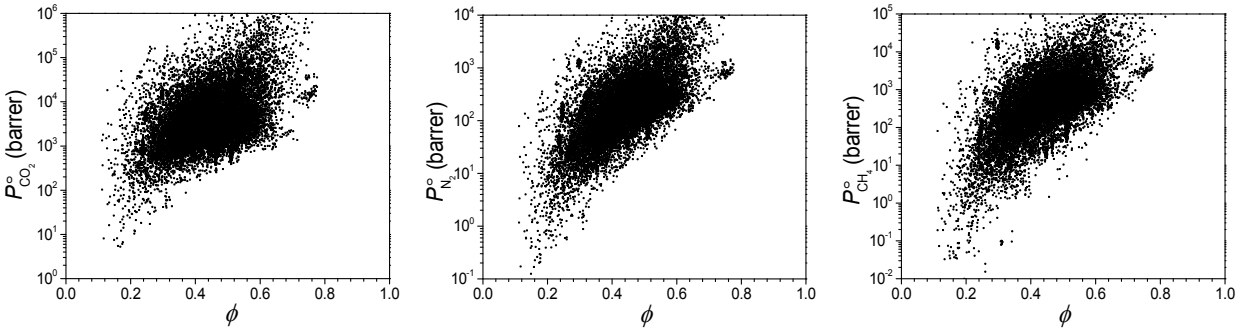


Fig. S7.2 Permeability versus porosity.

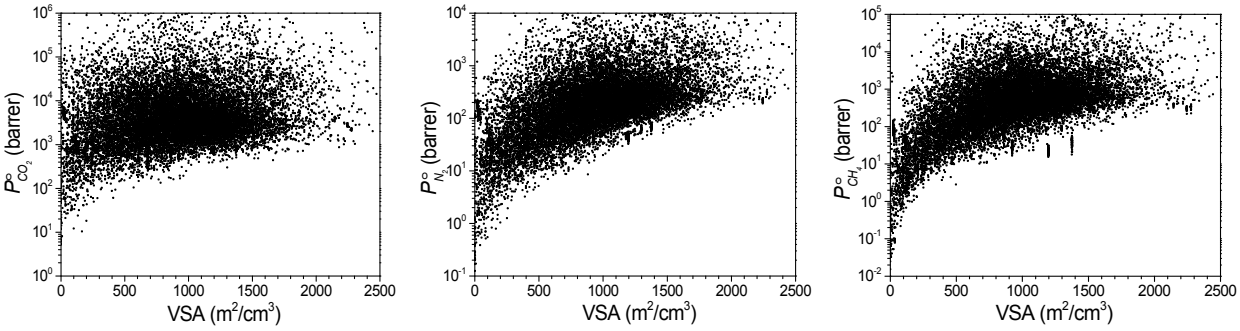


Fig. S7.3 Permeability versus VSA.

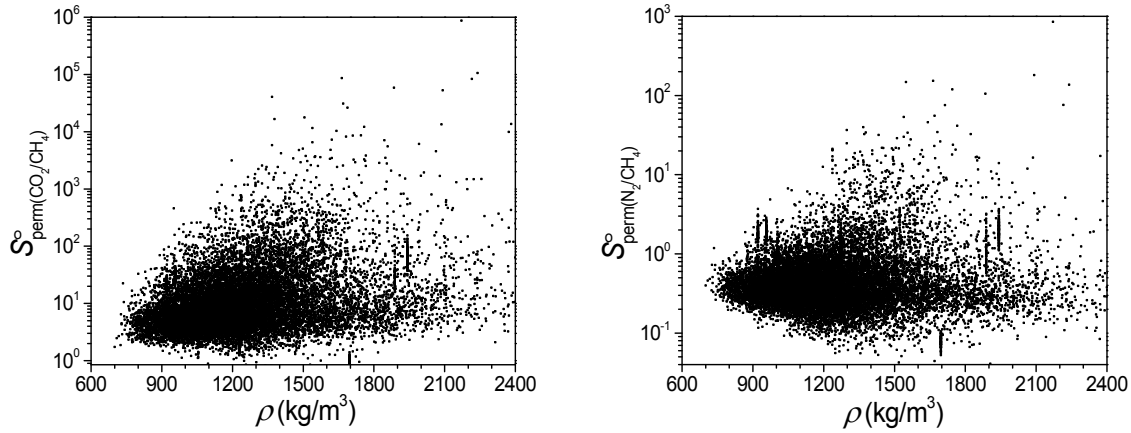


Fig. S8.1 Permselectivity versus density for CO_2/CH_4 and N_2/CH_4.

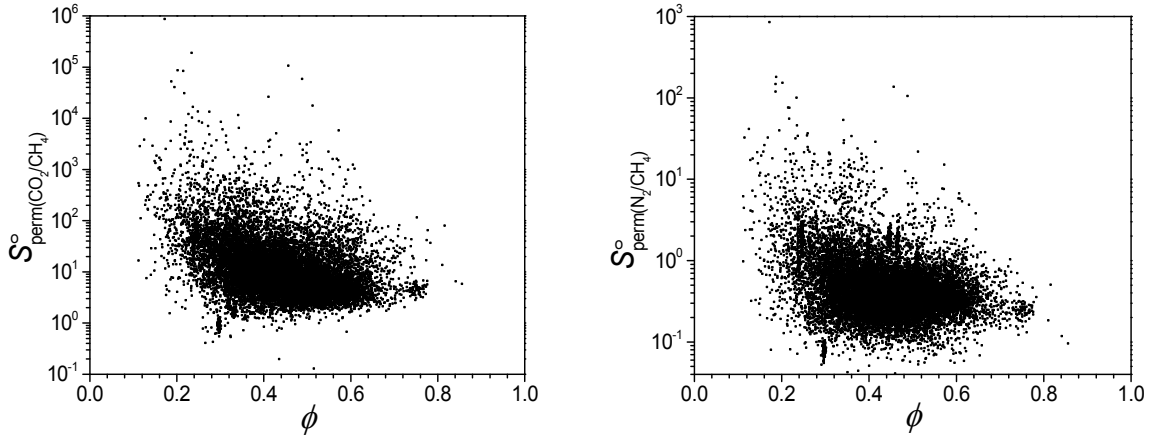


Fig. S8.2 Permselectivity versus porosity for CO_2/CH_4 and N_2/CH_4.

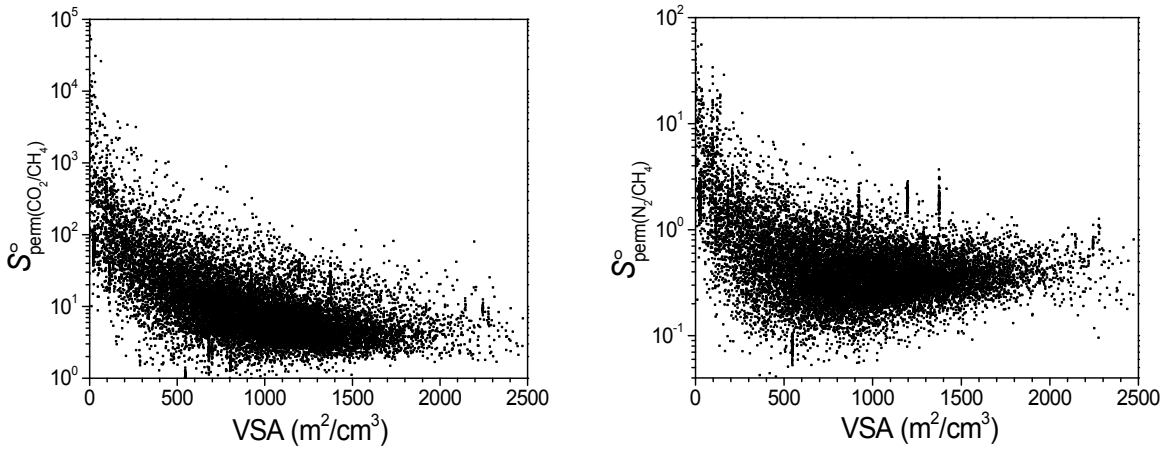


Fig. S8.3 Permselectivity versus VSA for CO_2/CH_4 and N_2/CH_4.

7. 24 Prescreened MOFs

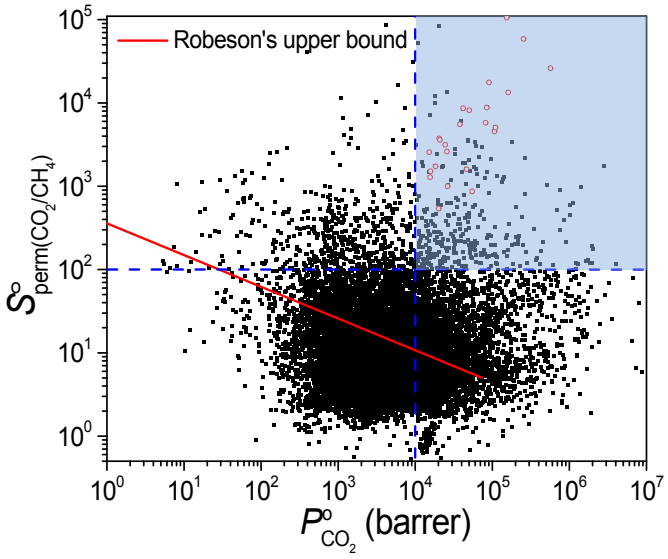


Fig. S9 Prescreened MOFs (red circles) for both CO_2/CH_4 and N_2/CH_4 separation.

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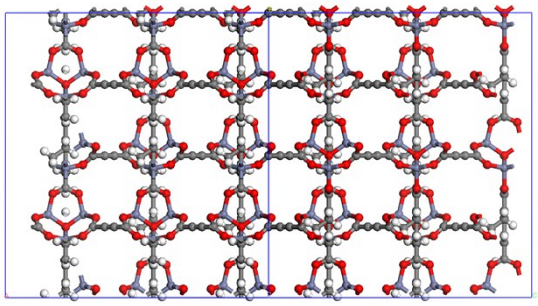

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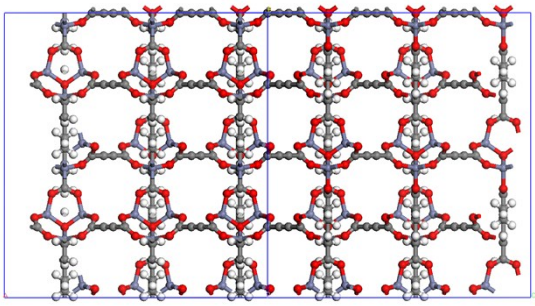

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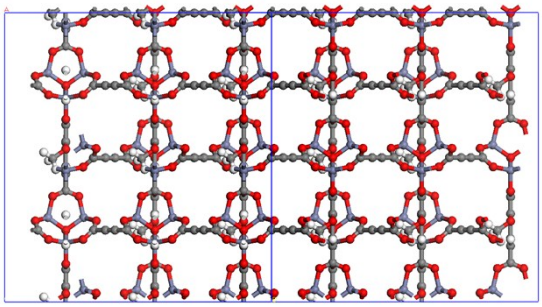

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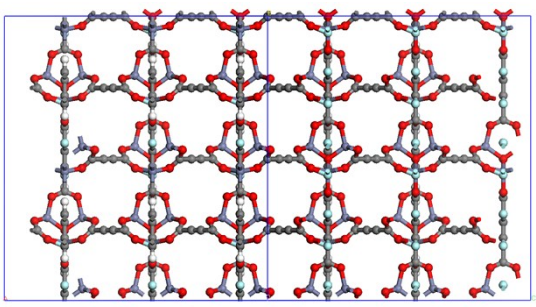

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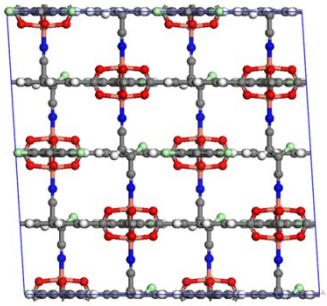

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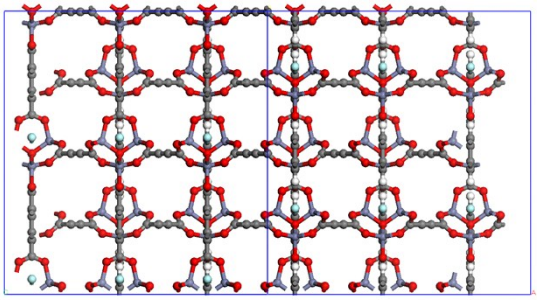

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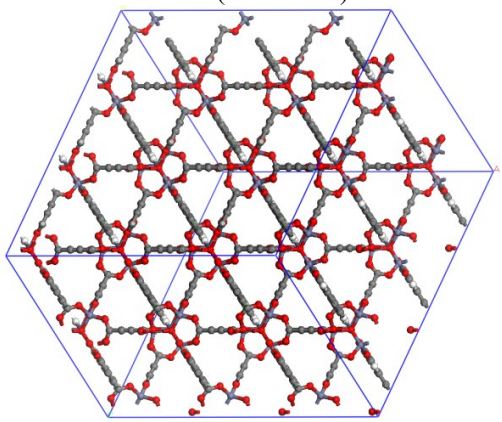

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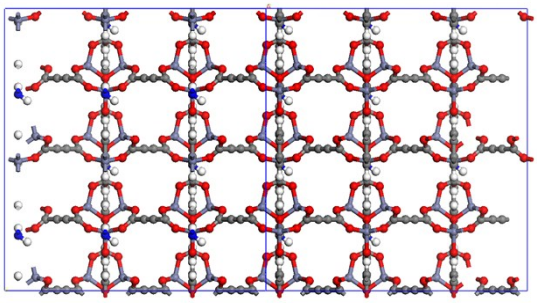

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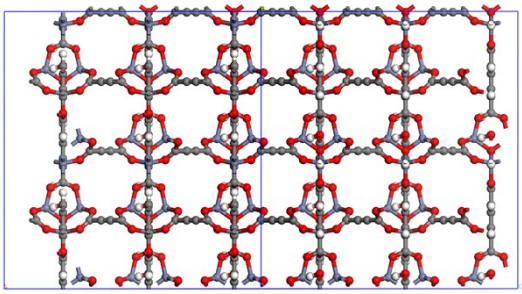

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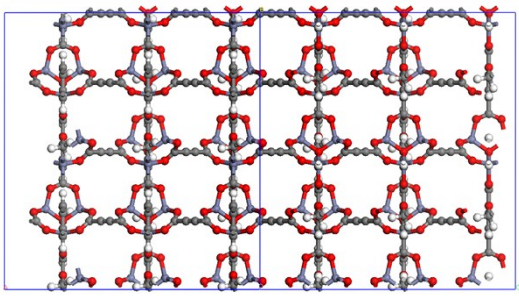

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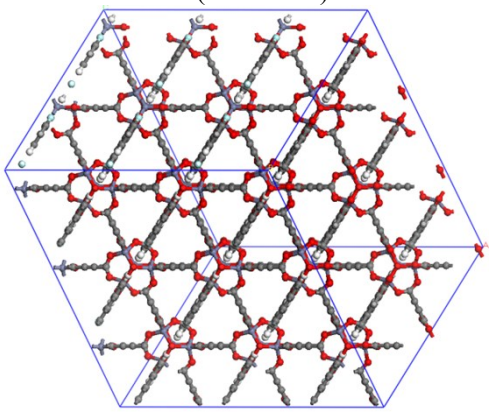

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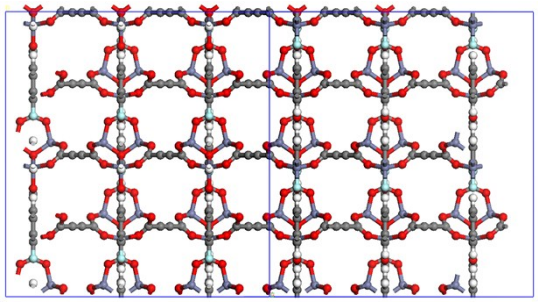

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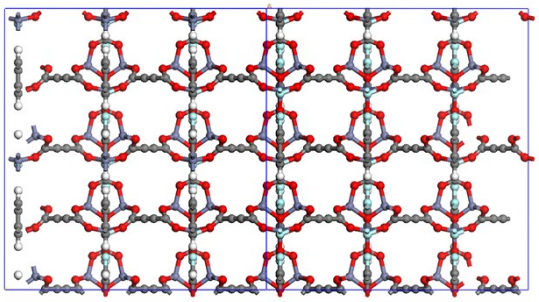

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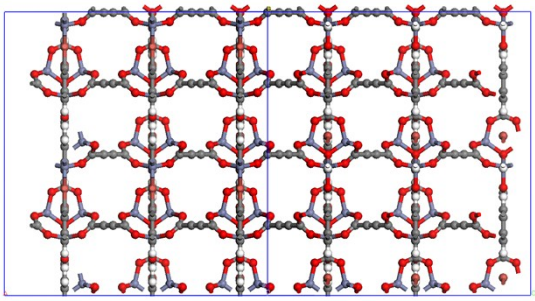

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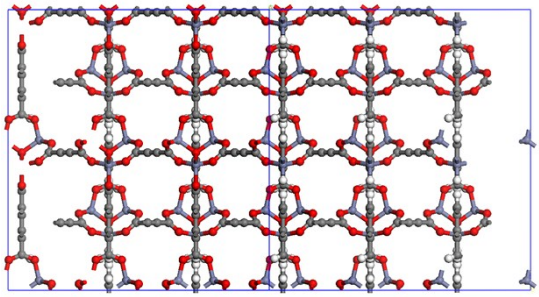

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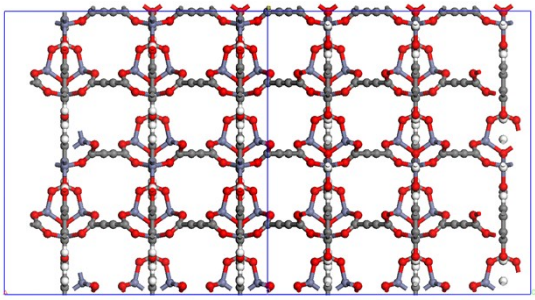

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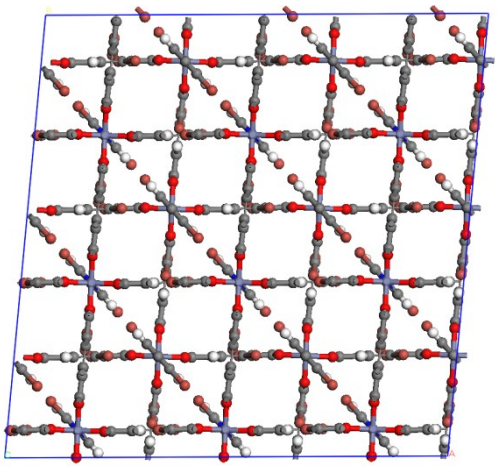

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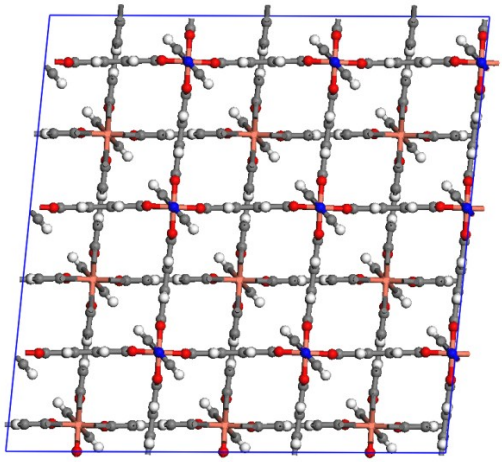

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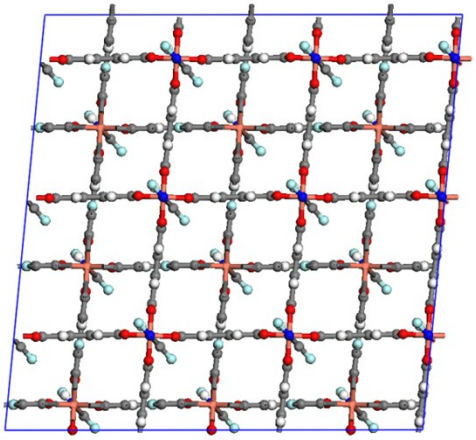

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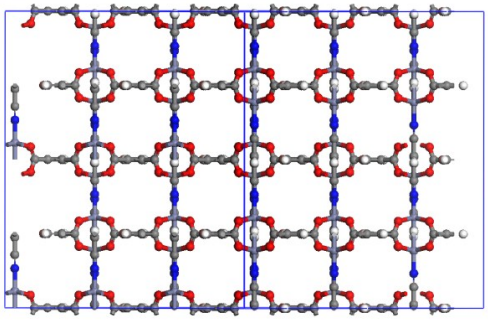

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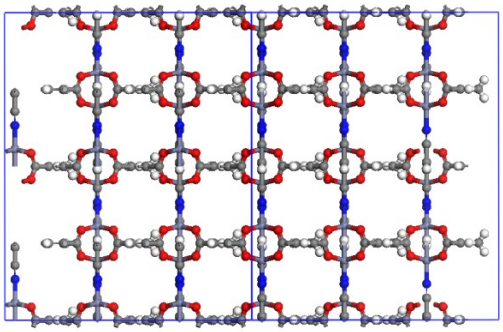

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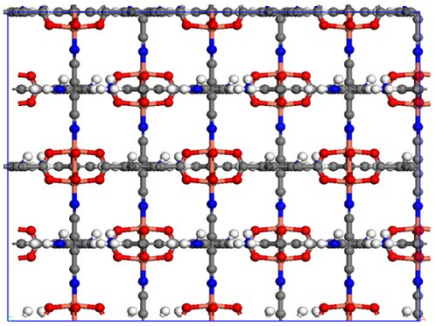

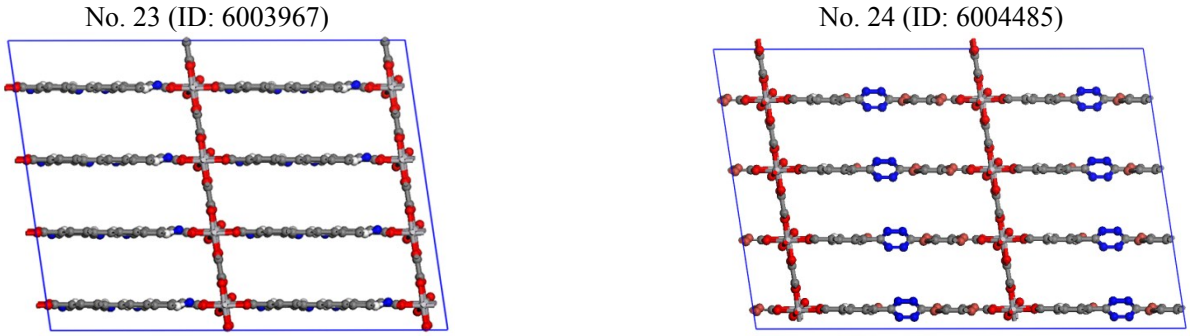

Fig. S10 Atomistic structures of 24 prescreened MOFs.

8. $CO_2/N_2/CH_4$ mixture in a MOF with PLD = 3.2 Å

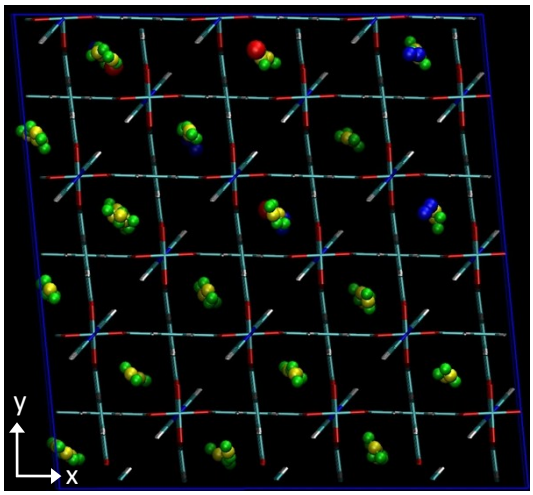

Fig. S11 Simulation snapshot for $CO_2/N_2/CH_4$ mixture in a MOF (ID: 31136)
CO_2: green-yellow-green balls, N_2: blue dumbbells, CH_4: red balls.