Supporting Information

for

Modelling of Gas Transport through Polymer/MOF Interfaces: A Microsecond Scale Concentration Gradient Driven Molecular Dynamics Study

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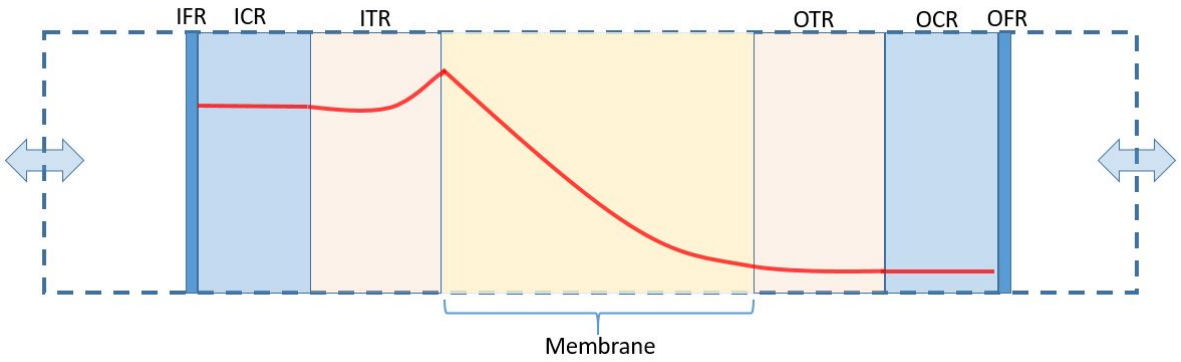


Figure S1: Schematic representation of CGD-MD simulation setup. IFR, ICR and ITR are force, control and transition regions for the inlet. OFR, OCR and OTR are force, control and transition regions for the outlet. A bell shaped force acts at the centre of the IFR and OFR. The red line is an illustration of the density profile of fluid molecules due to the concentration gradient. The blue dashed line represents the simulation box.

Table S1: CGD-MD specific parameters used in simulations. $\boldsymbol{Z_F}$ is the position where the centre of a bell shaped bias function is placed with respect to the z=0 (i.e. these correspond to the centre of the IFR and OFR shown in Figure S1), $\boldsymbol{w}$ is the width of the bias force function (i.e. width of IFR and OFR), and $\boldsymbol{k_i}$ is the bias force constant.

	Z_F (nm)	**w (nm)**	**k_i (kJ nm^3/mol)**		**ICR/OCR width (nm)**	**ITR/OTR width (nm)**
PIM-1 (Single Permeation)	5.125	0.25	Inlet	k_{CH4}=5,000 k_{H2}=5,000	2.5	2.5
	36.125	0.25	Outlet	k_{CH4}=500,000 k_{H2} =500,000	2.5	2.5
ZIF-8 (Single Permeation)	5.175	0.25	Inlet	k_{CH4}=5,000 k_{H2}=8,000	2.5	2.5
	25.125	0.25	Outlet	k_{CH4}=500,000 k_{H2} =500,000	2.5	2.5
MMM (Single Permeation)	15.125	0.25	Inlet	k_{CH4}=20,000 k_{H2}=20,000	2.5	2.5
	76.775	0.25	Outlet	k_{CH4}=500,000 k_{H2} =500,000	2.5	2.5
MMM (Mixture Permeation)	15.125	0.25	Inlet	k_{CH4}=20,000 k_{H2}=20,000	2.5	2.5
	76.775	0.25	Outlet	k_{CH4}=500,000 k_{H2} =500,000	2.5	2.5

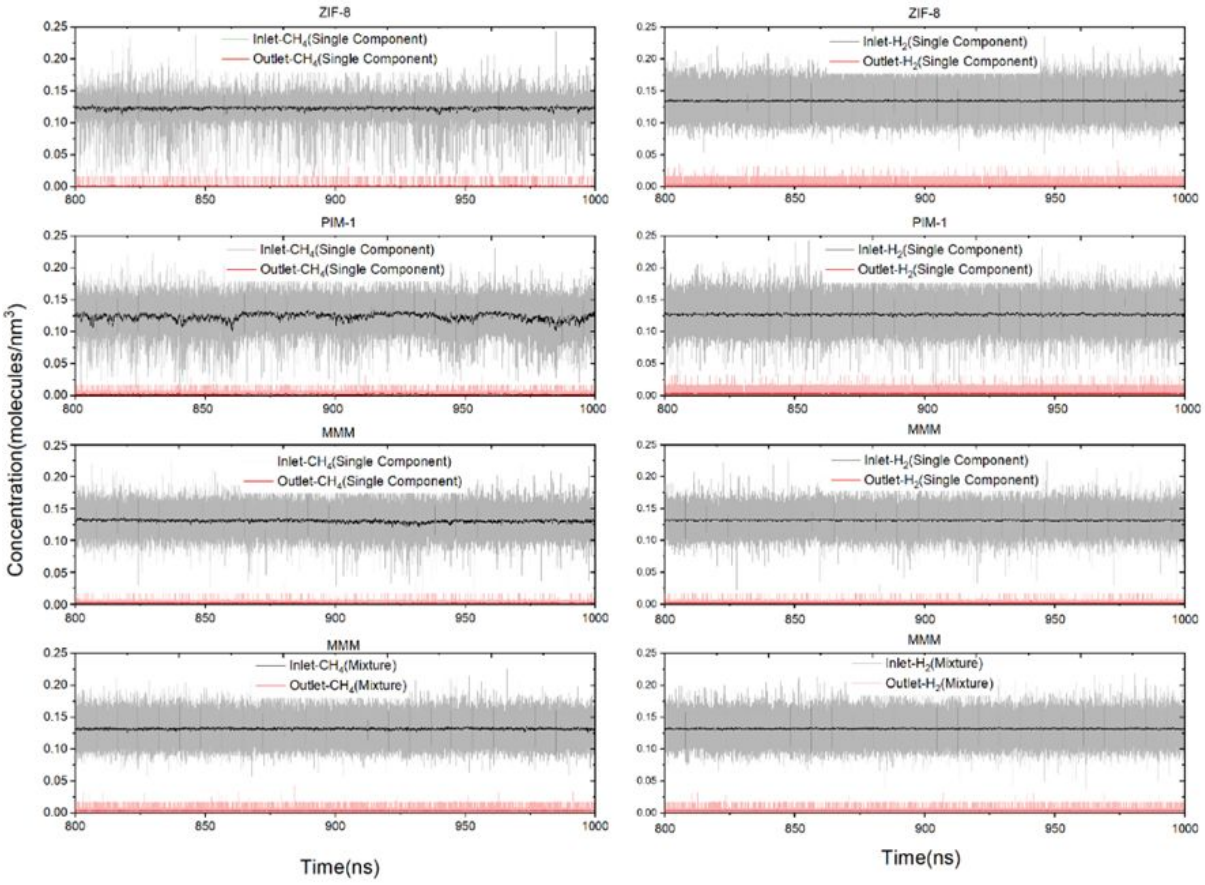
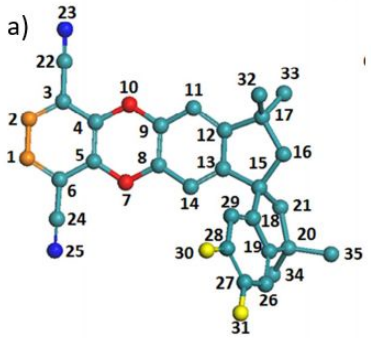
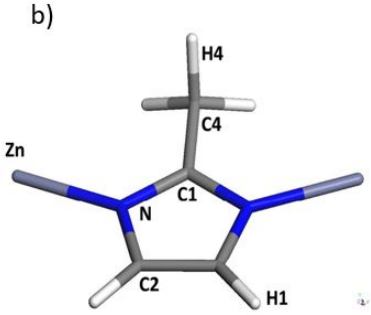


Figure S2: Variation of a) CH_4 and b) H_2 concentrations as a function of simulation time in the inlet (black lines) and outlet (red lines) control regions (ICR and OCR, respectively), for (from top to bottom), single component ZIF-8, PIM-1 and PIM-1/ZIF-8 and mixture PIM-1/ZIF-8 CGD-MD simulations. Dark lines are moving averages of the gas concentrations whereas transparent colours are instantaneous concentrations in the control regions.

Table S2: Tuned Lennard-Jones cross parameters between gas molecules and the atoms of a) PIM-1 and b) ZIF-8.





[nonbond_params]

; i j sigma(nm) epsilon(KJ/mole)

H_2	H1[ZIF-8 gate hydrogen]	0.20508	0.1414839
H_2	OS[7,10]	0.19453	0.430446008
H_2	CA1[4,5]	0.22953	0.284900364
H_2	CA2[3,6]	0.23933	0.235649165
H_2	CA3[11,14,26,29]	0.232848	0.366100728
H_2	CA4[12,13,18,19]	0.23933	0.235649165
H_2	CA5[27,28](terminal)	0.22953	0.284900364
H_2	C31[15,17,20]	0.32753	0.036426312
H_2	C32[16,21]	0.23968	0.367564207
H_2	C33[33,33,34,35]	0.23478	0.509420772
H_2	C[22,24]	0.22778	0.398226791
H_2	N1[23,25]	0.20678	0.398226791
H_2	CO[8,9,27,28]	0.22953	0.284900364
H_2	LCA[1,2](terminal)	0.22953	0.284900364
H_2	LOS[30,31](terminal)	0.20993	1.118952898
H_2	F(terminal)	0.20328	0.540185055
CH_4	H1[ZIF-8 gate hydrogen]	0.35390	0.2991060
CH_4	OS[7,10]	0.35540	0.864266552
CH_4	CA1[4,5]	0.41154	0.572034239
CH_4	CA2[3,6]	0.42726	0.473145735
CH_4	CA3[11,14,26,29]	0.41688	0.735071548
CH_4	CA4[12,13,18,19]	0.427269	0.473145735
CH_4	CA5[27,28](terminal)	0.411548	0.572034239
CH_4	C31[15,17,20]	0.568757	0.073138192
CH_4	C32[16,21]	0.427831	0.738009976
CH_4	C33[33,33,34,35]	0.419970	1.022835212
CH_4	C[22,24]	0.408741	0.799575532
CH_4	N1[23,25]	0.375054	0.799575532
CH_4	CO[8,9,27,28]	0.411548	0.572034239
CH_4	LCA[1,2](terminal)	0.411548	0.572034239
CH_4	LOS[30,31](terminal)	0.380107	2.246677968
CH_4	F(terminal)	0.369439	1.084604956

Table S3: Permeabilities of H_2 and CH_4 obtained from CGD-MD simulations.

Membrane	Permeability ($x10^{13}$ mol.m/m^2sPa)	
	CH_4	H_2
ZIF-8	62.5	320.2
PIM-1	128.0	803.2
PIM-1/ZIF-8 (single component)	136.4	628.4
PIM-1/ZIF-8 (mixture)	139.8	470.0