Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Optimisation of the Fischer-Tropsch process using zeolites for tail gas separation

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Electronic Supporting Information

Table S1.- Henry coefficients and isosteric heats of adsorption in FAU-type structures.

FAU	CO_2		CO		CH_4		N_2		H_2	
	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]
T_{Si}	2.81	-29.90	0.24	-17.74	1.11	-23.96	0.10	-11.97	0.03	-5.82
$T_{1,12}$	4.85	-29.85	0.21	-17.82	1.16	-22.95	0.10	-12.26	0.03	-6.02
$T_{1,24}$	4.78	-28.34	0.10	-12.98	0.56	-17.78	0.07	-10.42	0.03	-6.06
$T_{1,32}$	6.77	-29.28	0.08	-11.06	0.62	-16.71	0.07	-10.38	0.03	-6.18
$T_{1,48}$	15.50	-31.97	0.08	-11.30	0.83	-17.33	0.07	-11.04	0.04	-6.05
$T_{1,54}$	19.00	-32.13	0.08	-11.46	0.91	-17.86	0.08	-11.28	0.04	-5.83

Table S2.- Henry coefficients and isosteric heats of adsorption in MFI-type structures.

MFI	CO_2		CO		CH_4		N_2		H_2	
	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]
T_{Si}	4.20	-27.07	0.22	-17.01	0.86	-20.19	0.09	-13.02	0.02	-7.16
$T_{5,8}$	46.59	-38.93	0.14	-17.51	1.68	-23.61	0.07	-14.49	0.02	-7.62
$T_{8,8}$	86.50	-42.51	0.15	-17.94	1.98	-24.29	0.08	-14.92	0.02	-7.86
$T_{11,8}$	51.47	-38.78	0.17	-18.34	2.21	-24.10	0.09	-15.11	0.02	-7.86

Table S3.- Henry coefficients and isosteric heats of adsorption in MOR-type structures.

MOR	CO_2		CO		CH_4		N_2		H_2	
	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]
T_{Si}	1.03	-22.17	0.13	-16.22	0.61	-21.66	0.08	-14.09	0.02	-7.23
$T_{1,4}$	6.42	-33.36	0.10	-16.80	1.31	-24.30	0.07	-14.87	0.02	-7.58
$T_{2,4}$	9.67	-34.61	0.10	-17.15	1.65	-24.53	0.08	-15.37	0.02	-7.56
$T_{3,4}$	2.39	-27.11	0.09	-15.88	1.20	-25.72	0.06	-13.91	0.02	-7.70
$T_{4,4}$	14.00	-34.48	0.10	-16.65	1.65	-24.11	0.08	-14.88	0.02	-7.29
$T_{1,8}$	28.30	-39.31	0.07	-16.69	2.39	-25.49	0.06	-15.32	0.03	-7.64
$T_{2,8}$	29.80	-40.08	0.05	-15.95	2.45	-25.90	0.04	-15.02	0.03	-7.53

Table S4.- Henry coefficients and isosteric heats of adsorption in DDR-type structures.

DDR	CO_2		CO		CH_4		N_2		H_2	
	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]	K_H [mol/bar/kg]	Q_{ST} [kJ/mol]
T_{Si}	3.50	-25.11	0.29	-18.88	2.08	-26.25	0.16	-16.42	0.02	-8.24
$T_{1,5}$	8.30	-32.58	0.14	-16.66	1.24	-22.09	0.06	-13.56	0.03	-8.26
$T_{2,5}$	5.94	-30.57	0.13	-16.35	0.93	-21.21	0.06	-13.11	0.02	-8.13
$T_{4,5}$	6.53	-30.04	0.15	-16.54	1.17	-21.94	0.06	-13.25	0.02	-8.03
$T_{5,5}$	14.90	-33.32	0.17	-16.85	1.44	-22.28	0.07	-13.70	0.03	-8.33

Figure S1.- Selectivity at zero loading of CO2/CH4 (red), CH4/CO (blue), CO/H2 (green), and N2/H2 (orange) in: (a) FAU-type structures, (b) MFI-type structures, (c) MOR-type structures, and (d) DDR-type structures.

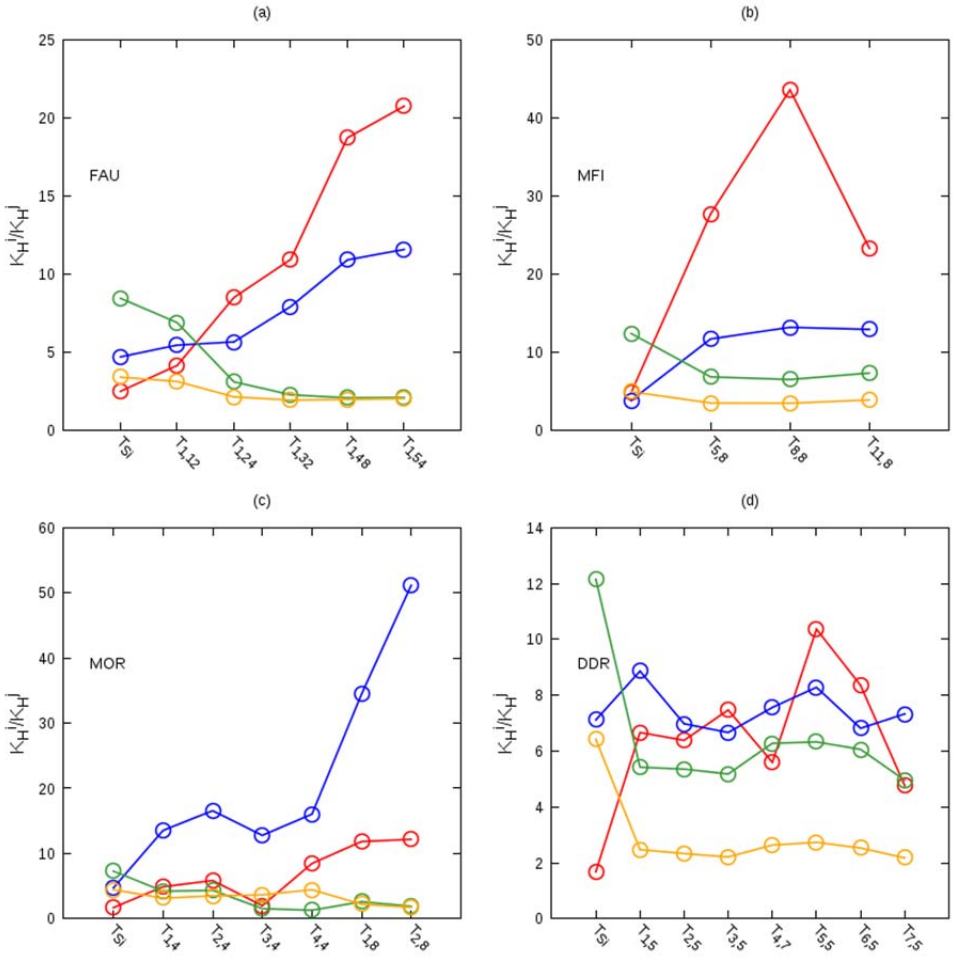

Figure S2.- Pure component adsorption isotherms in FAU-type structures: T_{Si} (grey), $T_{1,12}$ (red), $T_{1,24}$ (blue), $T_{1,32}$ (green), $T_{1,48}$ (orange), and $T_{1,54}$ (violet). Circles at top left (a) are for CO_2, squares at top right (b) are for CO, diamonds at centre left (c) are for CH_4, up-pointing triangles at centre right (d) are for N_2, and down-pointing triangles at bottom (e) are for H_2.

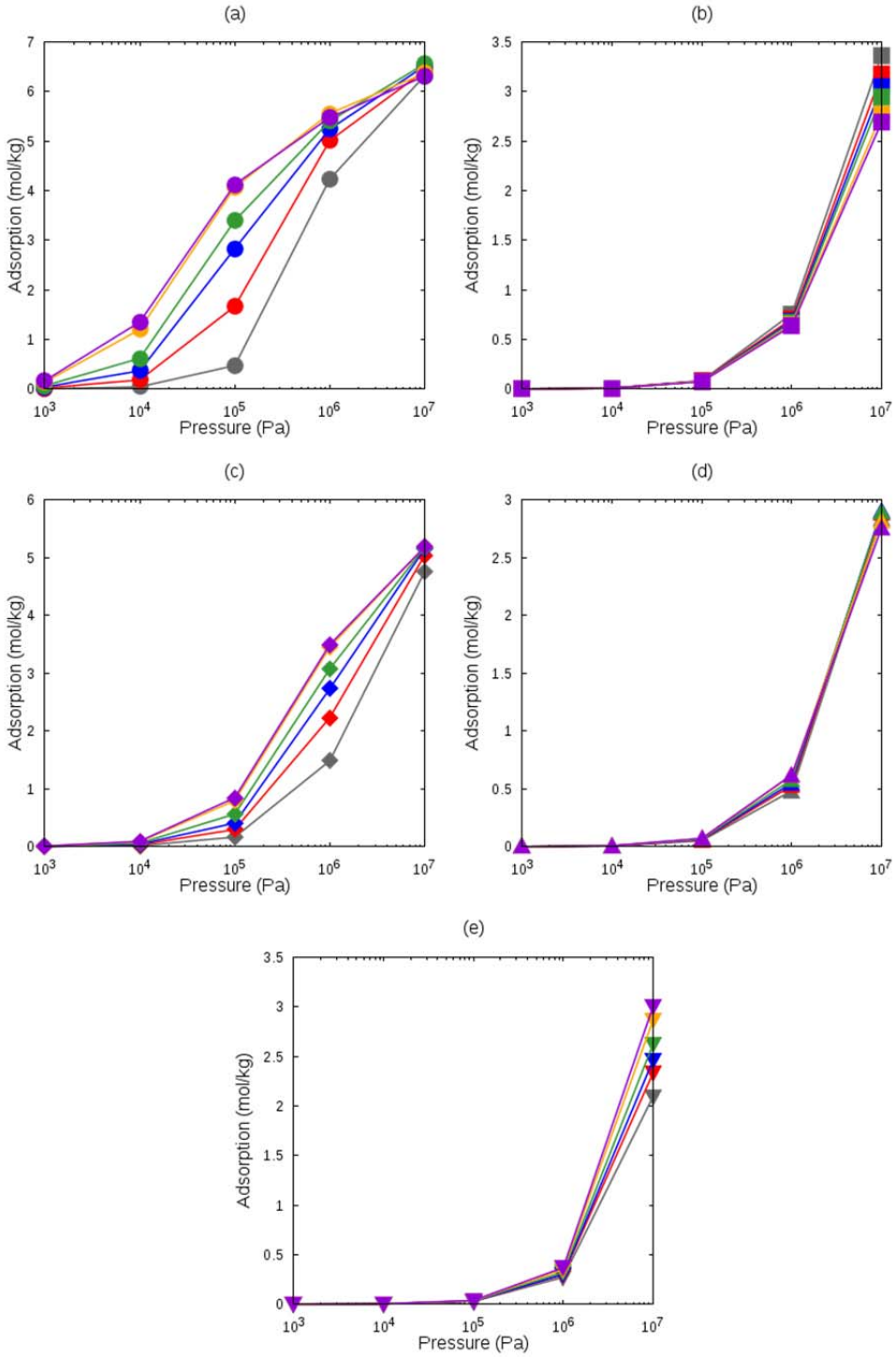

Figure S3.- Pure component adsorption isotherms in MFI-type structures: T_{Si} (grey), $T_{5,8}$ (red), $T_{8,8}$ (blue), and $T_{11,8}$ (green). Circles at top left (a) are for CO_2, squares at top right (b) are for CO, diamonds at centre left (c) are for CH_4, up-pointing triangles at centre right (d) are for N_2, and down-pointing triangles at bottom (e) are for H_2.

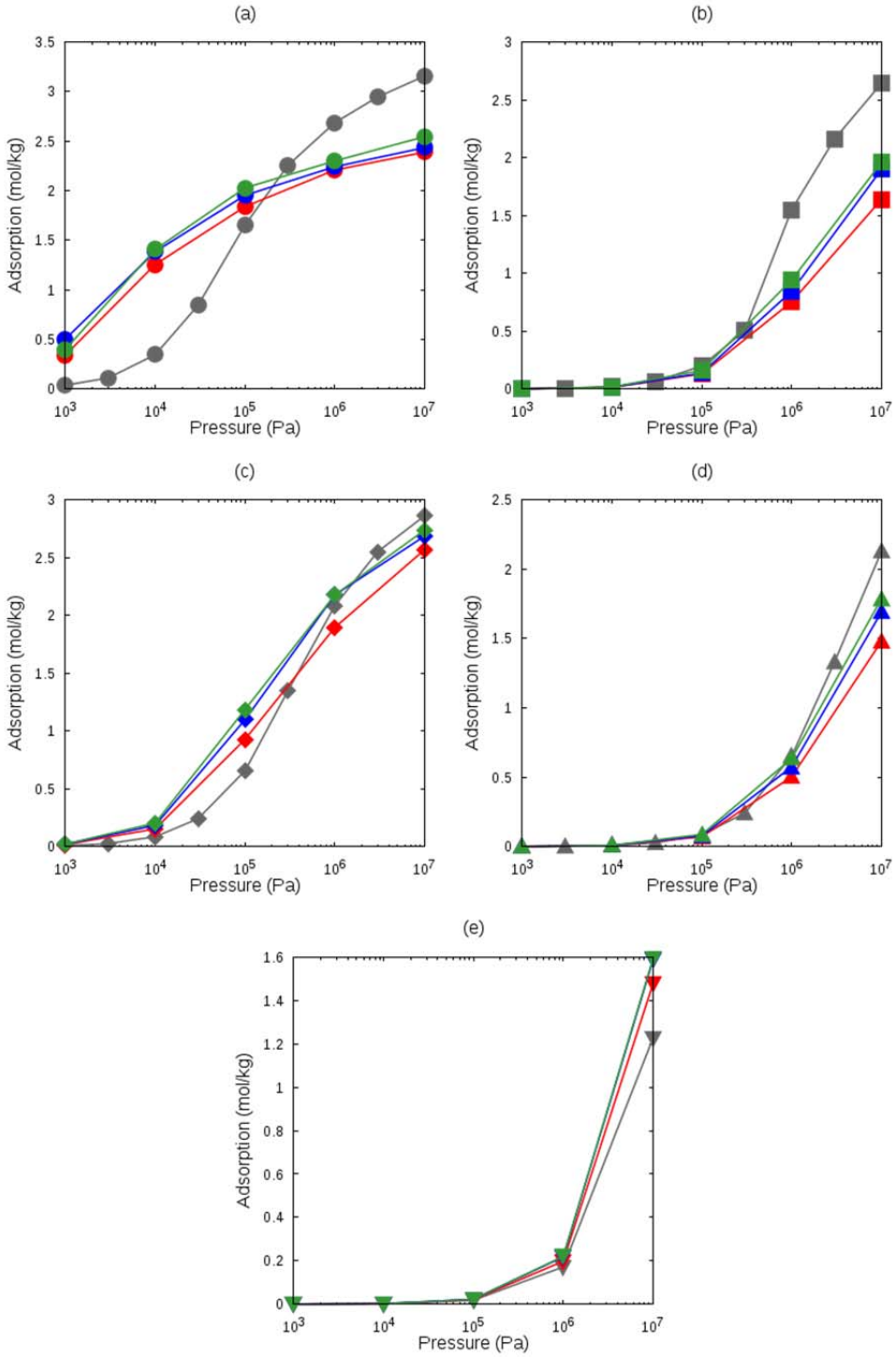

Figure S4.- Pure component adsorption isotherms in MOR-type structures: T_{Si} (grey), $T_{1,4}$ (red), $T_{2,4}$ (blue), $T_{1,8}$ (green), and $T_{2,8}$ (orange). Circles at top left (a) are for CO_2, squares at top right (b) are for CO, diamonds at centre left (c) are for CH_4, up-pointing triangles at centre right (d) are for N_2, and down-pointing triangles at bottom (e) are for H_2.

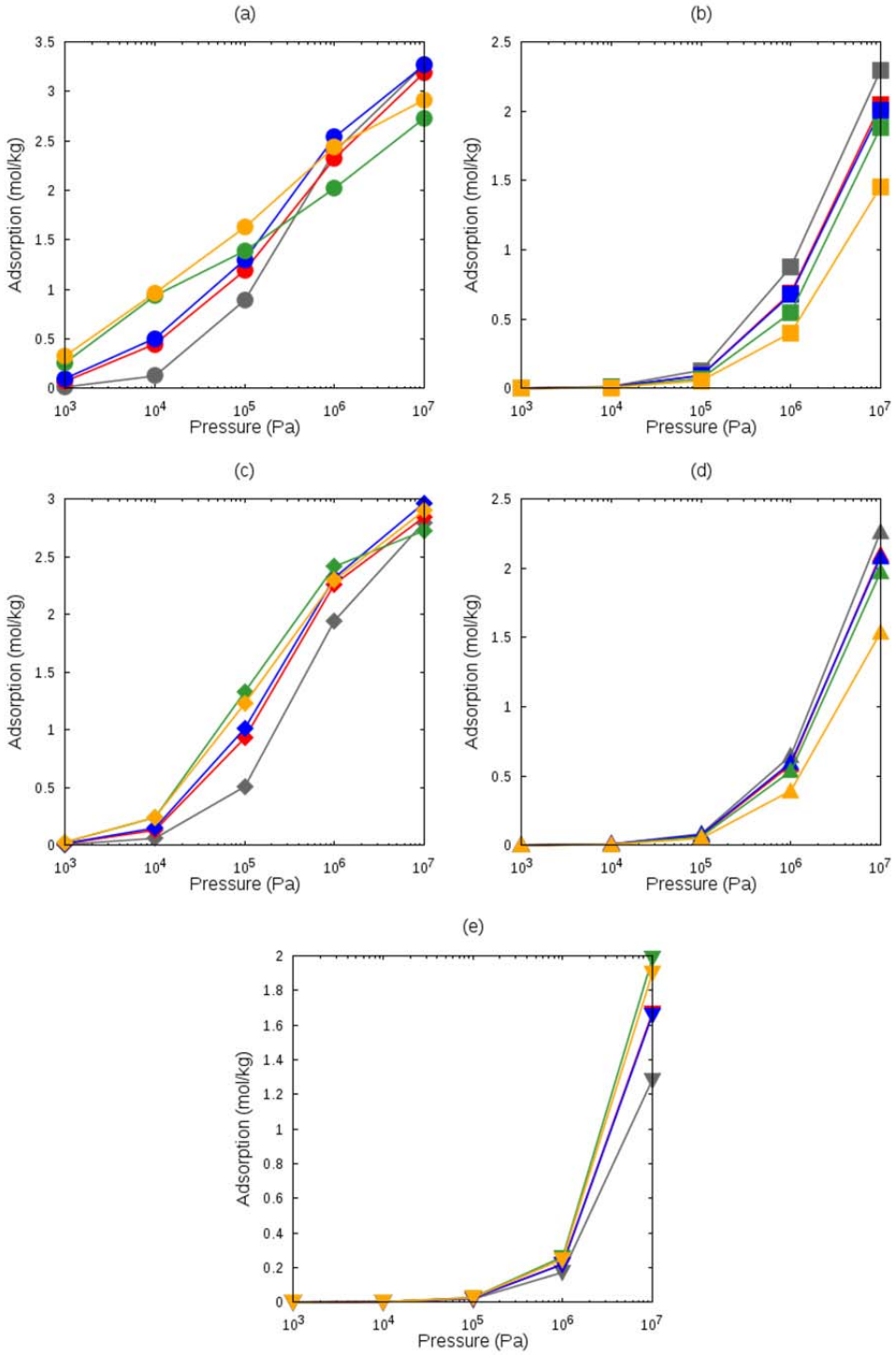

Figure S5.- Pure component adsorption isotherms in DDR-type structures: T_{Si} (grey), $T_{1,5}$ (red), $T_{2,5}$ (blue), $T_{4,5}$ (green), and $T_{5,5}$ (orange). Circles at top left (a) are for CO_2, squares at top right (b) are for CO, diamonds at centre left (c) are for CH_4, up-pointing triangles at centre right (d) are for N_2, and down-pointing triangles at bottom (e) are for H_2.

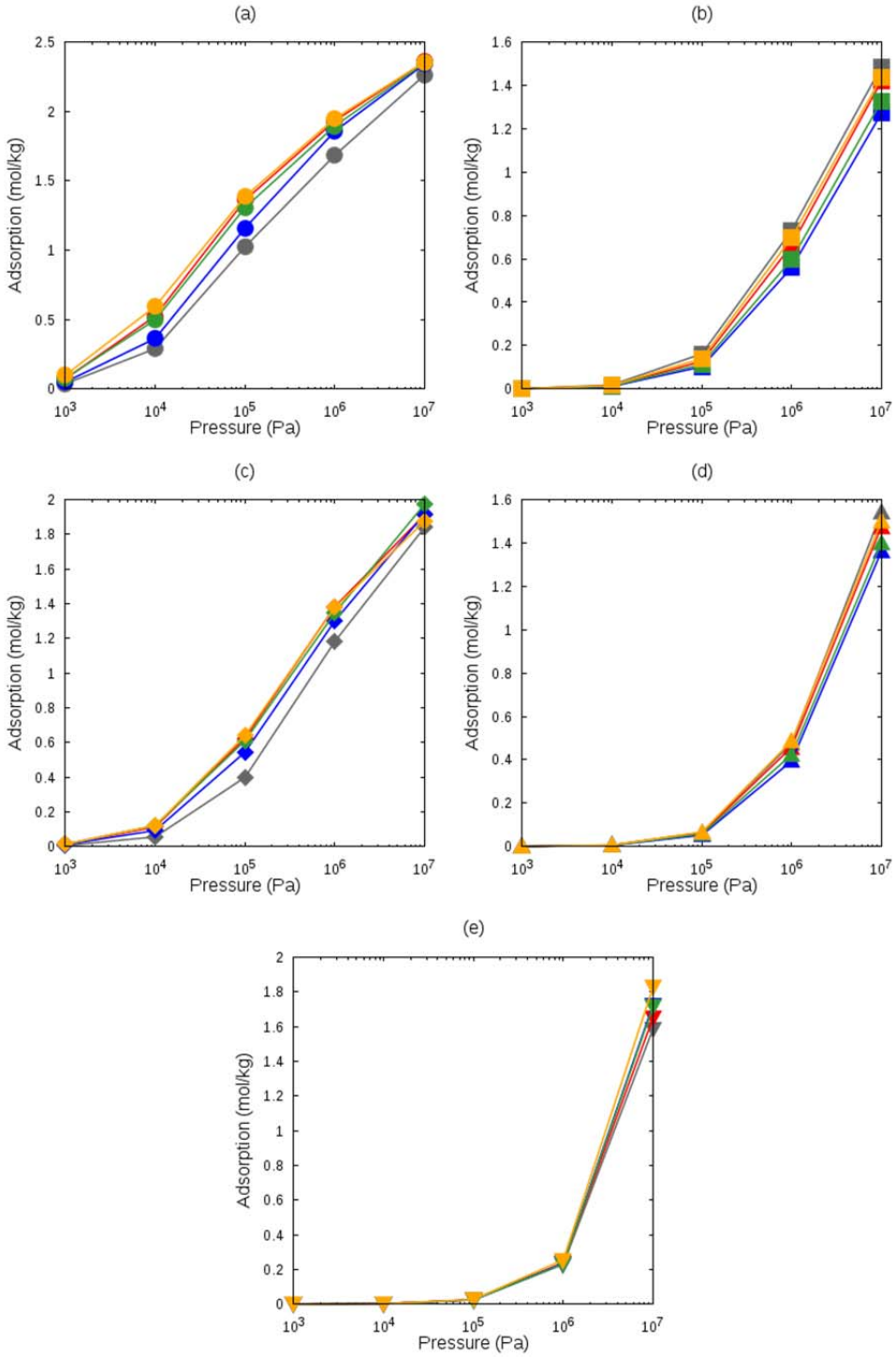

Figure S6.- Pure component adsorption isotherms in MFI-type structures: TSi (grey), T7,4 (red), T9,4 (blue), T10,4 (green), and T12,4 (orange). Circles at top left (a) are for CO2, squares at top right (b) are for CO, diamonds at centre left (c) are for CH4, up-pointing triangles at centre right (d) are for N2, and down-pointing triangles at bottom (e) are for H2.

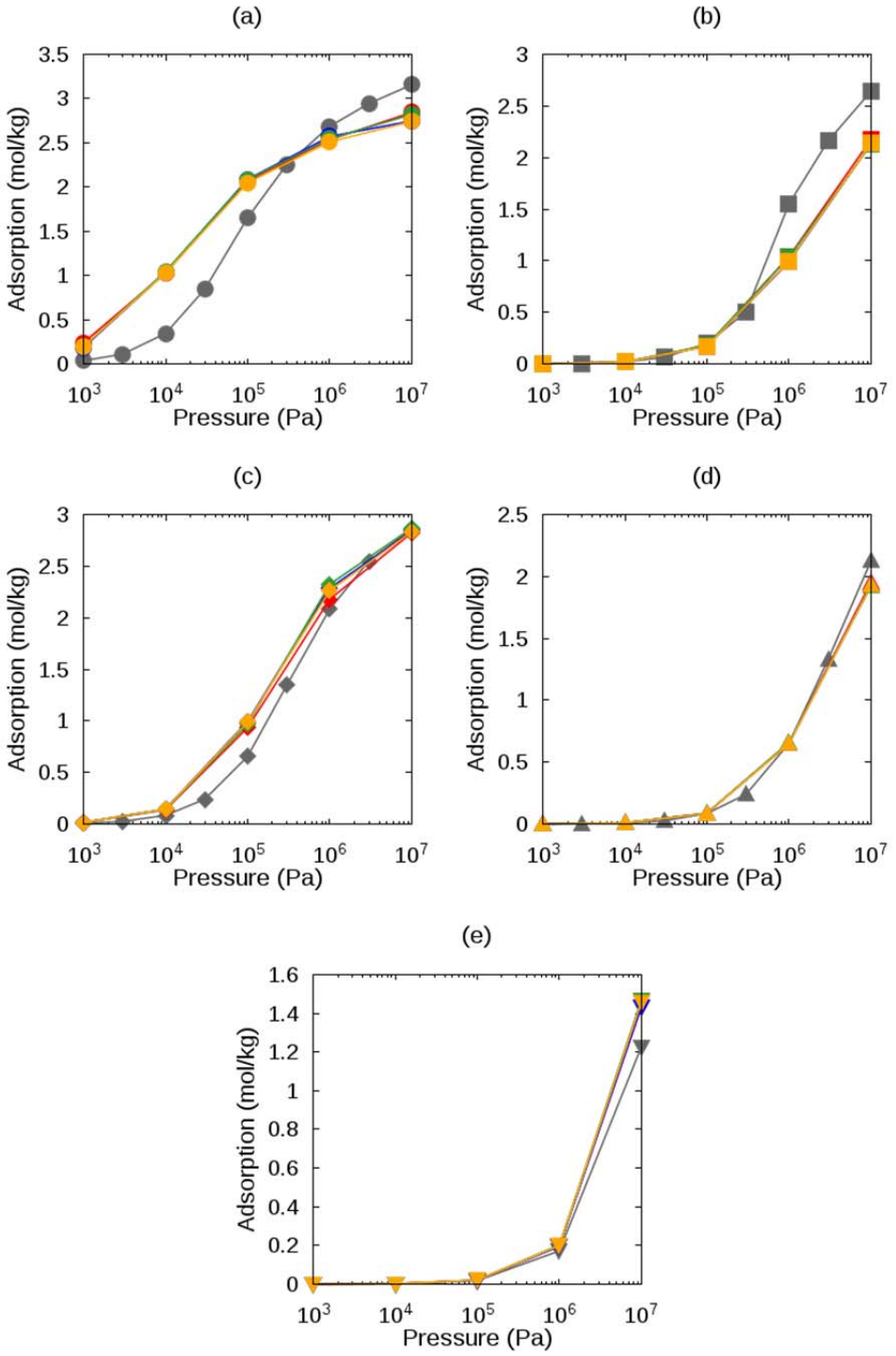

Figure S7.- Computed adsorption isotherms of the five-component mixture (red circles for CO_2, blue squares for CO, green diamonds for CH_4, orange up-pointing triangles for N_2, and violet down-pointing triangles for H_2) in FAU-type structures: $T_{1,12}$ at top left (a), $T_{1,24}$ at top right (b), $T_{1,32}$ at centre left (c), $T_{1,48}$ at centre right (d), and $T_{1,54}$ at bottom (e).

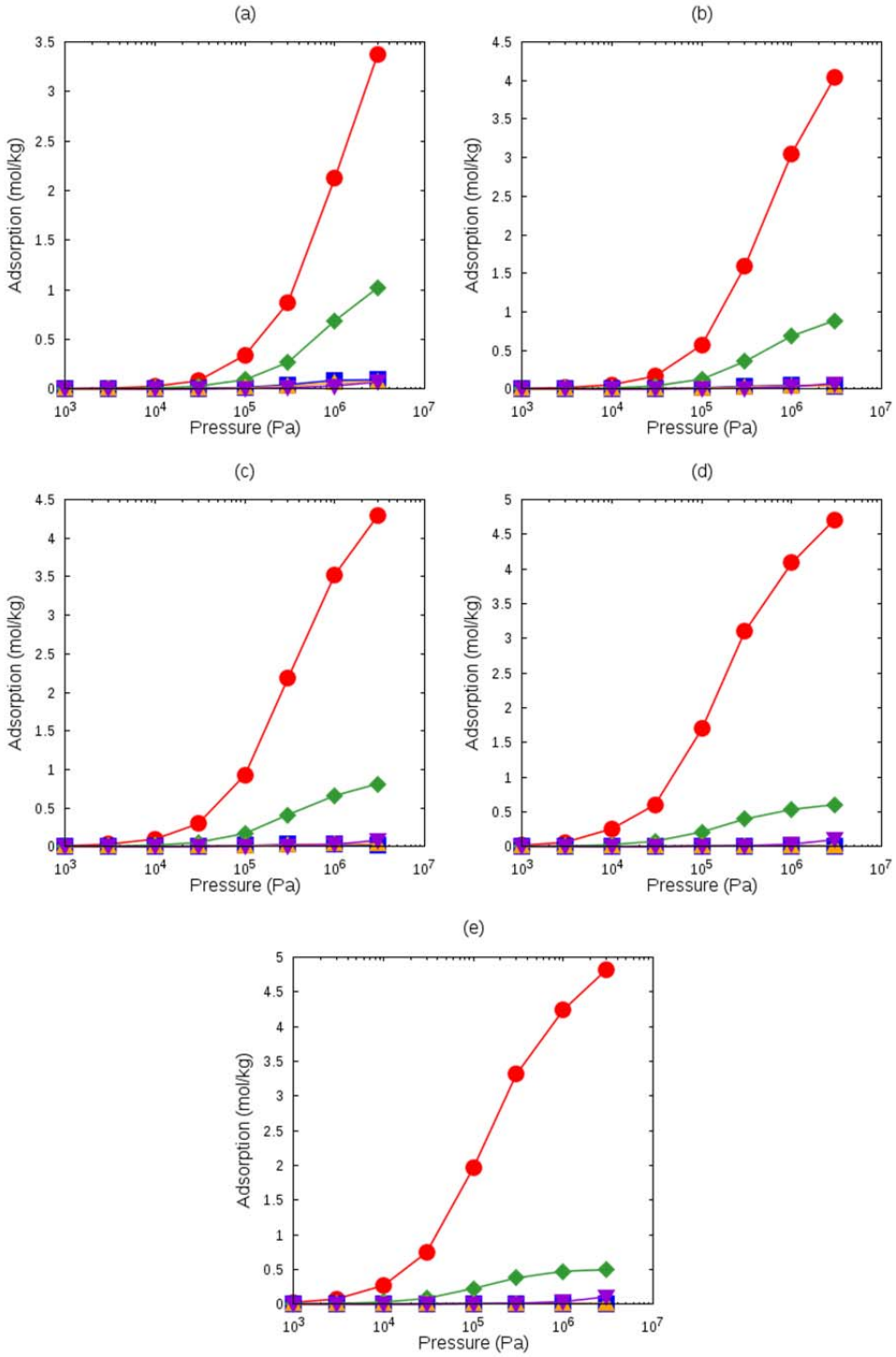

Figure S8.- Computed adsorption isotherms of the five-component mixture (red circles for CO_2, blue squares for CO, green diamonds for CH_4, orange up-pointing triangles for N_2, and violet down-pointing triangles for H_2) in MFI-type structures: $T_{5,8}$ at top left (a), $T_{8,8}$ at top right (b), and $T_{11,8}$ at bottom (c).

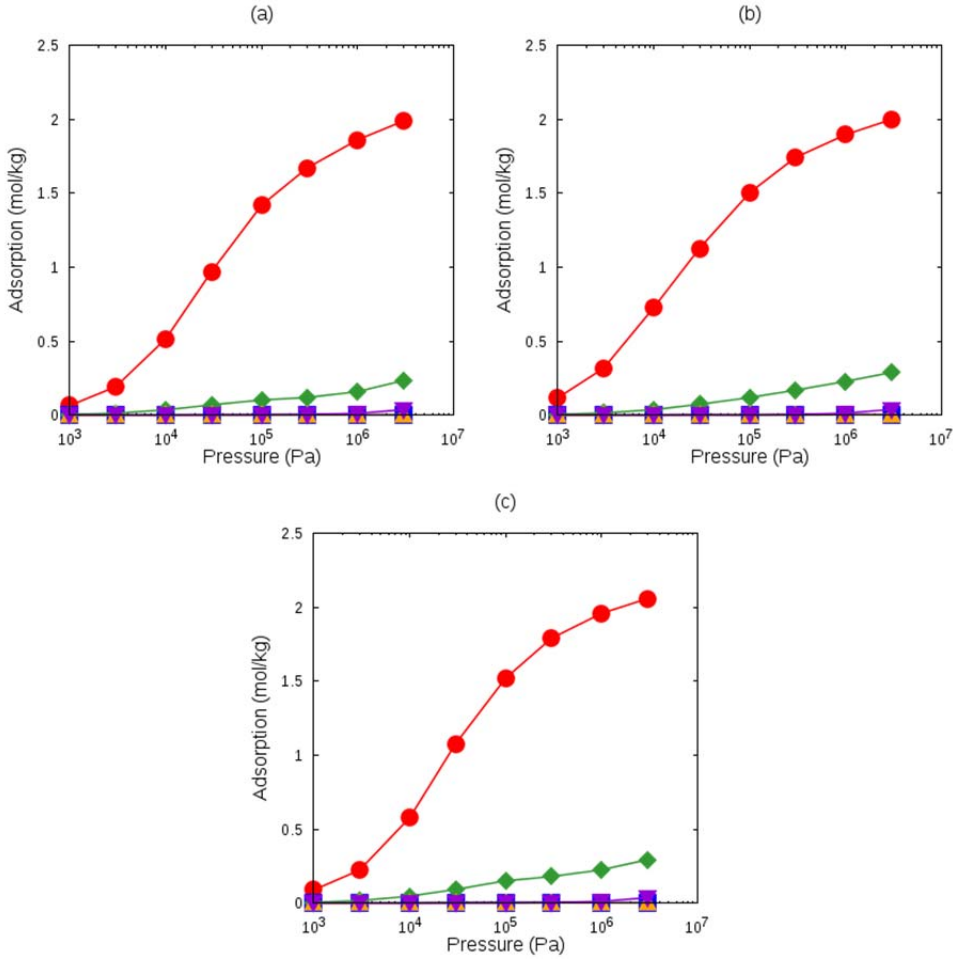

Figure S9.- Computed adsorption isotherms of the five-component mixture (red circles for CO_2, blue squares for CO, green diamonds for CH_4, orange up-pointing triangles for N_2, and violet down-pointing triangles for H_2) in MOR-type structures: $T_{1,4}$ at top left (a), $T_{2,4}$ at top right (b), $T_{1,8}$ at bottom left (c), and $T_{2,8}$ at bottom right (d).

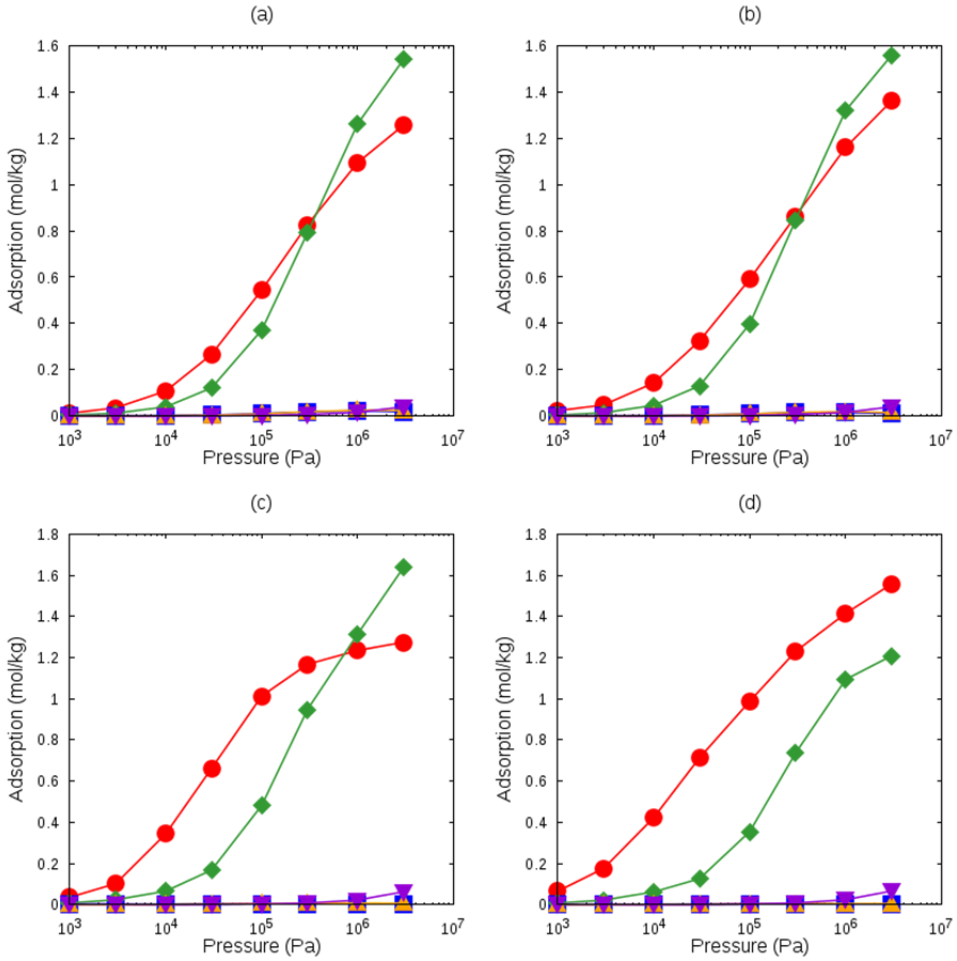

Figure S10.- Computed adsorption isotherms of the five-component mixture (red circles for CO_2, blue squares for CO, green diamonds for CH_4, orange up-pointing triangles for N_2, and violet down-pointing triangles for H_2) in DDR-type structures: $T_{1,5}$ at top left (a), $T_{2,5}$ at top right (b), $T_{4,5}$ at bottom left (c), and $T_{5,5}$ at bottom right (d).

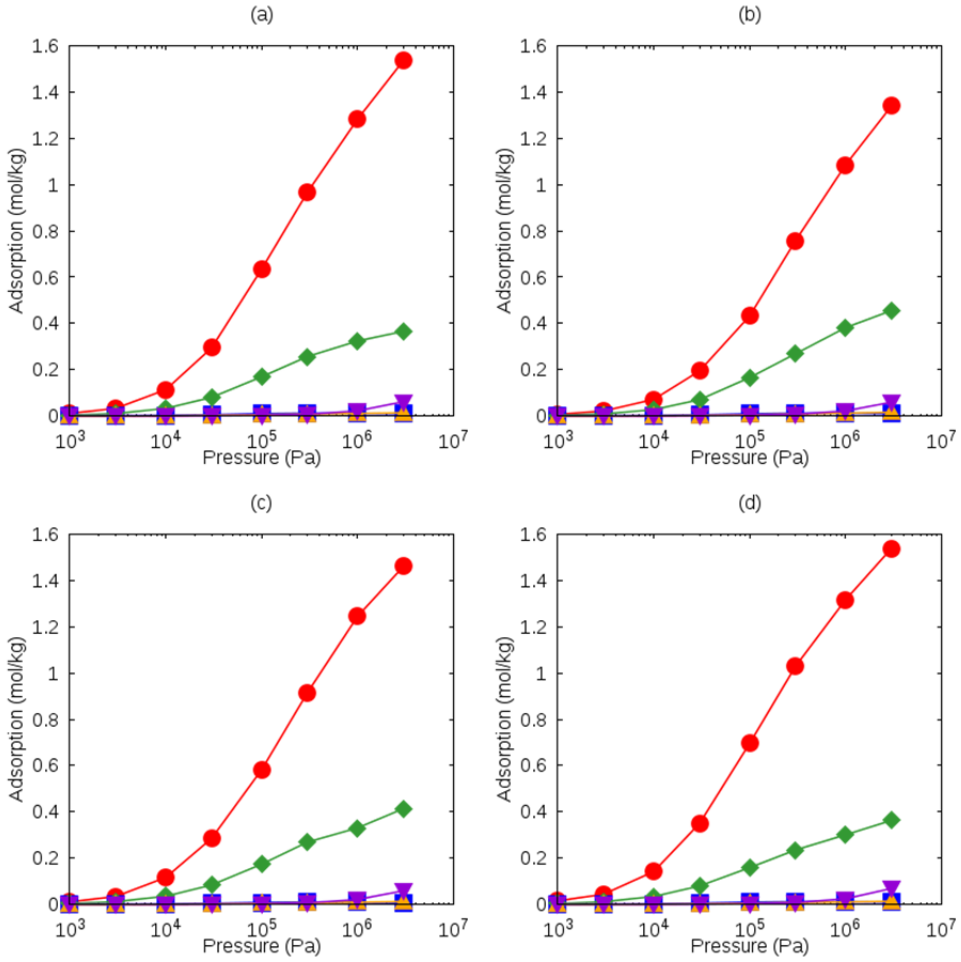

Figure S11.- Computed adsorption isotherms of the five-component mixture (red circles for CO2, blue squares for CO, green diamonds for CH4, orange up-pointing triangles for N2, and violet down-pointing triangles for H2) in MFI-type structures: T7,4 at top left (a), T9,4 at top right (b), T10,4 at bottom left (c), and T12,4 at bottom right (d).

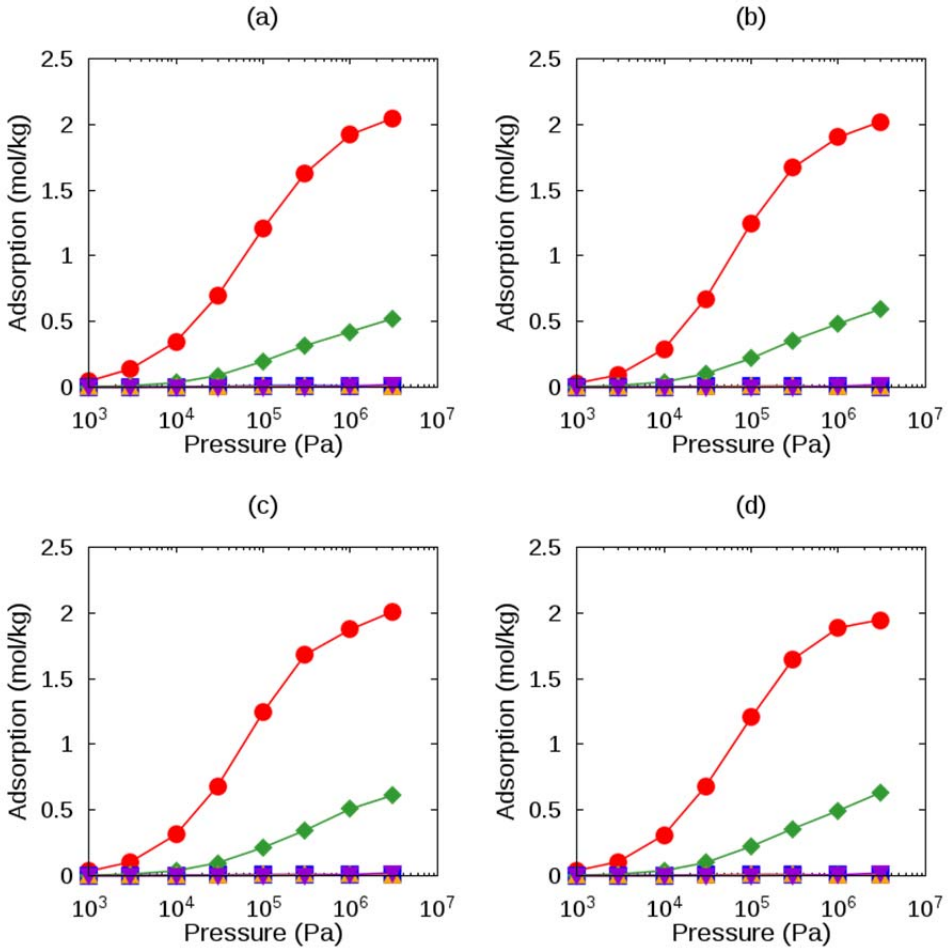

Figure S12.- Computed CO2/CH4 (left) and CH4/CO (right) adsorption selectivity in MFI structures: TSi (grey), T7,4 (red), T9,4 (blue), and T10,4 (green), and T12,4 (orange).

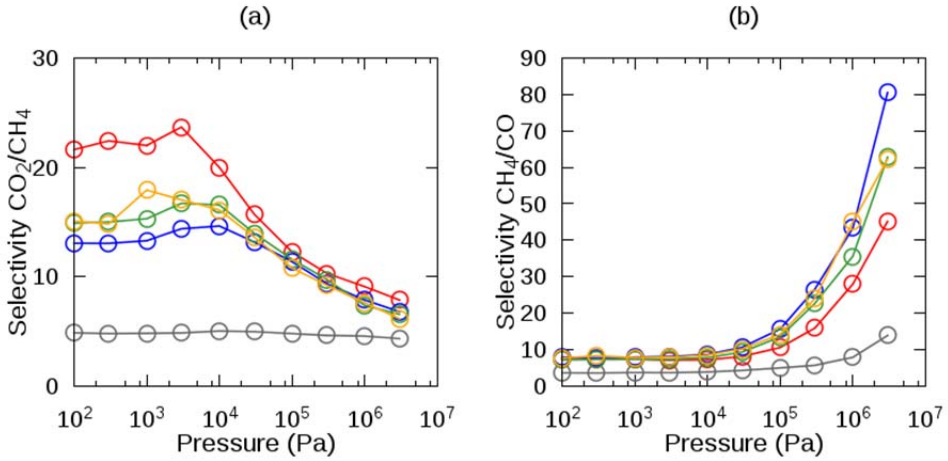

Figure S13.- Computed adsorption isotherms (circles for CO_2 and diamonds for CH_4) of five-component mixture and IAST calculations (continuous lines for CO_2 and dashed lines for CH_4) in $T_{2,8}$ MOR (orange) and $T_{2,8}$ MOR with the *side pockets* artificially blocked (violet).

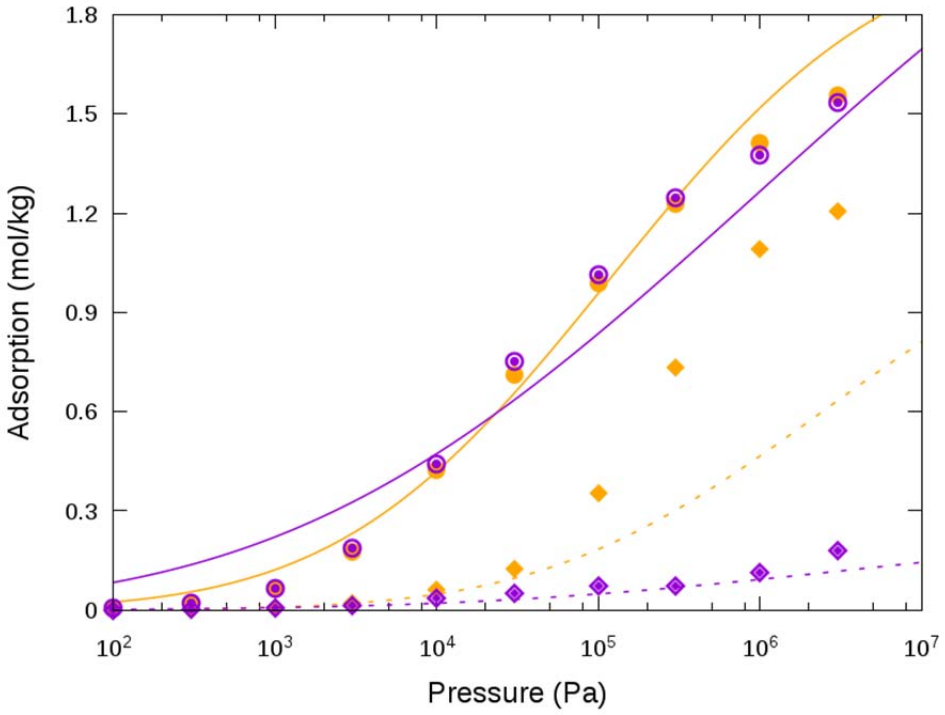

Figure S14.- Mean square displacement for CO_2 in T1,48 (orange) and T1,54 (violet) FAU, and T5,8 MFI (red). In grey, the MSD for T5,8 MFI for each diffusion direction: X axe (x symbol), Y axe (+ symbol), and Z axe (* symbol).

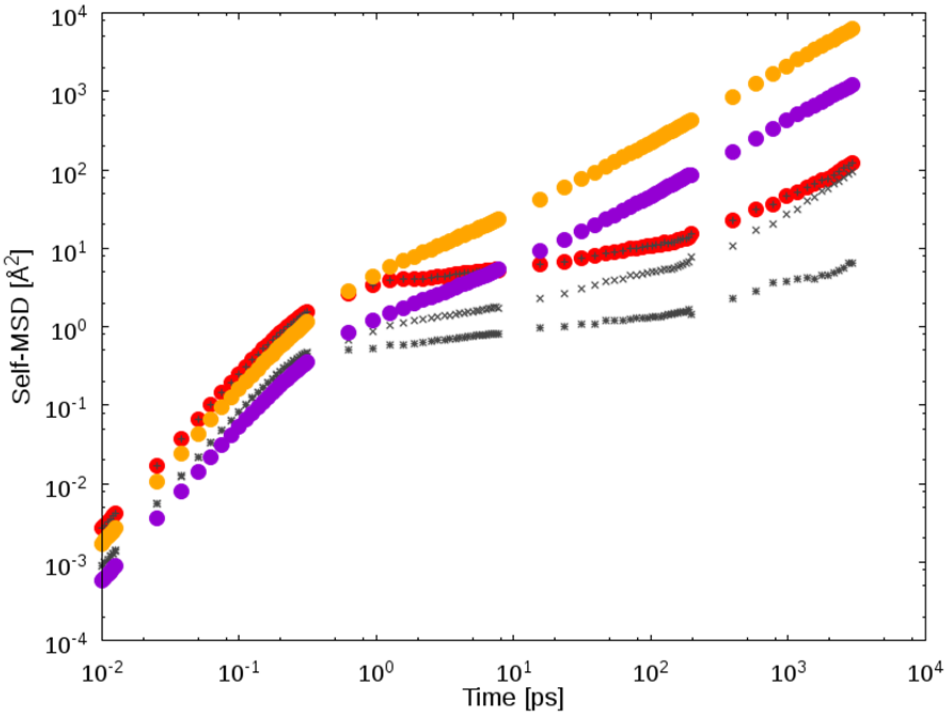

Figure S15.- XY view of the average occupation profiles of CO_2 (up) and CH_4 (bottom) on pure silica MFI (left end), $T_{5,8}$ MFI (left), $T_{8,8}$ (right), and $T_{11,8}$ (right end). The atomic structure has been included for reference, where aluminium atoms are highlighted in orange. The relation between colour and probability density (from black to yellow) is shown in the colour ramp on the right side of the figure.

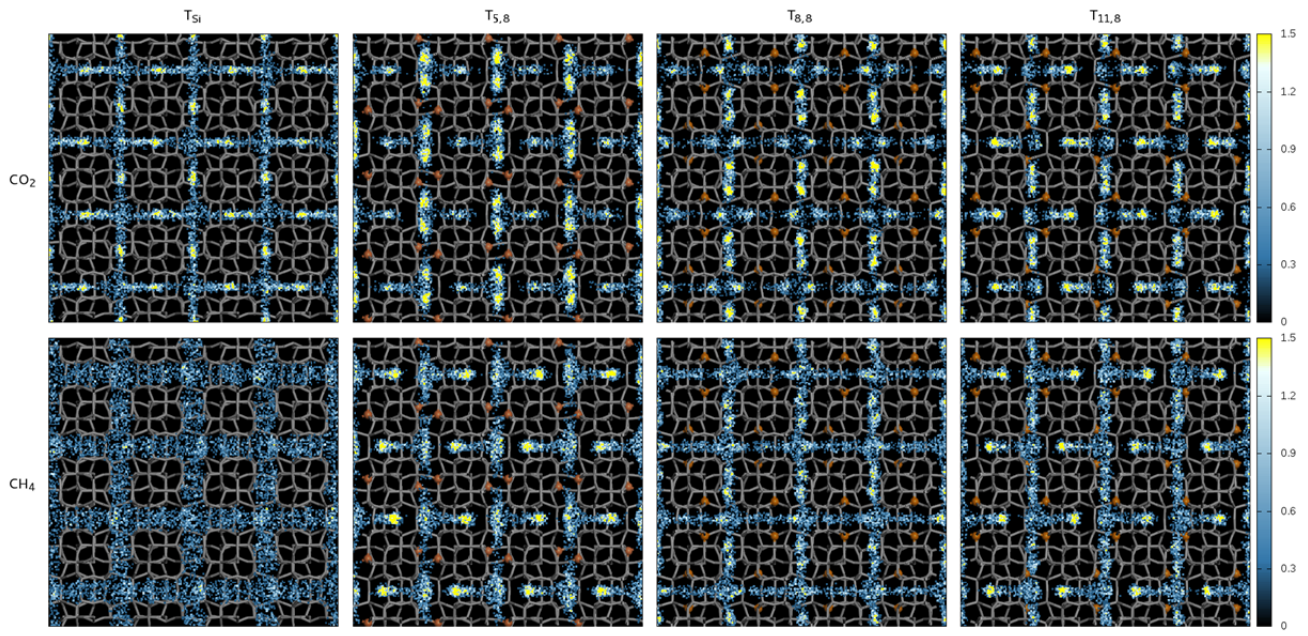

Figure S16.- YZ view of the average occupation profiles of CO_2 (up) and CH_4 (bottom) on pure silica MFI (left end), $T_{5,8}$ MFI (left), $T_{8,8}$ (right), and $T_{11,8}$ (right end). The atomic structure has been included for reference, where aluminium atoms are highlighted in orange. The relation between colour and probability density (from black to yellow) is shown in the colour ramp on the right side of the figure.

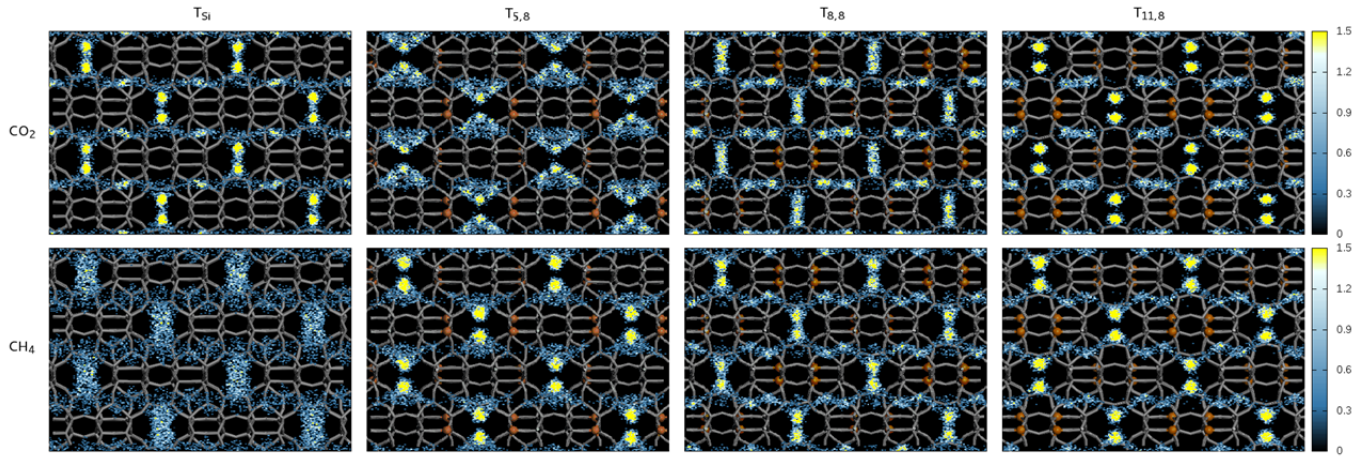

Figure S17.- ZX view of the average occupation profiles of CO_2 (up) and CH_4 (bottom) on pure silica MFI (left end), $T_{5,8}$ MFI (left), $T_{8,8}$ (right), and $T_{11,8}$ (right end). The atomic structure has been included for reference, where aluminium atoms are highlighted in orange. The relation between colour and probability density (from black to yellow) is shown in the colour ramp on the right side of the figure.