SUPPORTING INFORMATION

Use of a New Size Weighted Combining Rule to Predict Adsorption in Siliceous Zeolites

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CONTENT LIST

1- NUMERICAL RESULTS OF GCMC SIMULATIONS IN GAS PHASE

GCMC simulations were performed for the adsorbates at gas phase in order to establish the relationship between chemical potential and pressure at a set temperature prior to perform GCMC

simulations in adsorbed phase.

1.1- Pure components

The values of fixed variables (μ,V,T) and the output variables: number of molecules (N), pressure (P), mass density (ρ), internal energy (U) and enthalpy (H) for the performed GCMC simulations of methane, ethane, propane, n-butane, n-pentane, 2.methylpentane, n-hexane, carbon dioxide, and methanol are presented in Table S1, S2, S3, S4, S5, S6, S7, S8, and S9, respectively. In all simulations a cubic box was used, with *L* being the length of the box edge.

Table S1. Summary of set up and results for GCMC simulations of methane in gas phase.

Set up		**<Average values> on production run**				
$-\mu_x10^{-3}$ [J/mol]	**L_{box} [Å]**	**N [molecules]**	**P_x10^{-5} [Pa]**	**ρ [kg/m3]**	**U_x10^{-3} [J/mol]**	**H_x10^{-3} [J/mol]**
				T=298K		
45.0	500	100.6663	0.0331	0.0215	-0.0003	2.4774
42.0	400	174.5105	0.1122	0.0726	-0.0012	2.4770
40.0	300	162.9797	0.2482	0.1608	-0.0026	2.4739
39.0	300	247.5038	0.3771	0.2442	-0.0040	2.4736
38.0	200	109.7349	0.5641	0.3654	-0.0059	2.4705
37.0	180	119.3357	0.8414	0.5451	-0.0089	2.4675
36.0	170	151.3730	1.2657	0.8208	-0.0133	2.4606
35.0	170	227.0942	1.8968	1.2314	-0.0203	2.4509
34.0	120	119.7949	2.8423	1.8468	-0.0299	2.4392
33.0	100	103.8415	4.2449	2.7663	-0.0449	2.4169
32.0	80	80.4136	6.3933	4.1840	-0.0677	2.3837
31.0	70	81.7569	9.6859	6.3499	-0.1025	2.3447
28.0	50	107.3067	33.4347	22.869	-0.3715	1.9739
				T=300K		
60.0	4000	143.3729	0.0001	0.0001	$-9x10^{-8}$	2.4943
55.0	2000	132.2525	0.0007	0.0004	$-5x10^{-7}$	2.4943
50.0	1200	213.5462	0.0051	0.0033	$-5x10^{-5}$	2.4943
45.0	700	313.3899	0.0378	0.0243	-0.0004	2.4937
43.0	450	186.8460	0.0849	0.0546	-0.0009	2.4932
40.0	350	292.1065	0.2821	0.1815	-0.0030	2.4902
38.0	250	238.0454	0.6302	0.4059	-0.0068	2.4843
36.0	210	315.6780	1.4092	0.9081	-0.0147	2.4749
35.0	180	295.8178	2.0937	1.3513	-0.0220	2.4637
33.67	150	295.2102	3.6005	2.3302	-0.0381	2.4407
31.0	100	259.4049	10.5753	6.9106	-0.1119	2.3431
29.0	70	207.1018	24.1283	16.0851	-0.2597	2.1468
27.0	50	186.6091	56.5803	39.7702	-0.6372	1.6452
				T=303K		

45.0	450	101.5174	0.0466	0.0297	-0.0004	2.5188
42.0	350	156.5354	0.1527	0.0973	-0.0018	2.5169
40.0	250	126.7188	0.3391	0.2161	-0.0035	2.5144
39.0	250	188.1343	0.5033	0.3208	-0.0052	2.5121
38.0	180	105.0648	0.7522	0.4799	-0.0079	2.5067
37.0	160	109.4038	1.1158	0.7116	-0.0114	2.5045
36.0	150	134.4508	1.6613	1.0613	-0.0177	2.4937
				T=309K		
50.0	800	117.9591	0.0098	0.0061	-0.0001	2.5691
45.0	500	202.9121	0.0692	0.0432	-0.0007	2.5684
42.0	300	141.1198	0.2229	0.1392	-0.0022	2.5662
40.0	230	138.0310	0.4836	0.3022	-0.0049	2.5621
38.0	180	144.3665	1.0545	0.6595	-0.0109	2.5546
36.0	150	182.4199	2.2992	1.4399	-0.0235	2.5382
				T=338K		
55.0	800	121.6091	0.0111	0.0063	-0.0001	2.8101
50.0	500	176.3709	0.0658	0.0376	-0.0006	2.8095
47.0	350	176.5876	0.1922	0.1097	-0.0016	2.8088
45.0	300	225.0579	0.3888	0.2221	-0.0036	2.8050
44.0	270	234.9326	0.5567	0.3180	-0.0049	2.8039
43.5	200	113.7358	0.6629	0.3787	-0.0064	2.8014
43.0	210	157.5602	0.7933	0.4532	-0.0070	2.8010
42.5	180	117.8405	0.9422	0.5383	-0.0084	2.7996
42.0	180	140.4297	1.1218	0.6398	-0.0104	2.7986
41.5	180	170.1545	1.3599	0.7773	-0.0126	2.7941
41.0	180	202.2267	1.6155	0.9238	-0.0144	2.7912
40.5	150	140.8714	1.9434	1.1120	-0.0176	2.7823
40.0	150	168.9731	2.3315	1.3338	-0.0210	2.7794
39.0	150	240.4698	3.3137	1.8981	-0.0302	2.7705
38.0	140	279.8213	4.7357	2.7166	-0.0428	2.7538
37.0	120	252.1862	6.7692	3.8879	-0.0607	2.7325
				T=373K		
60.0	700	117.8637	0.0177	0.0092	-0.0001	3.1011
55.0	400	110.1907	0.0887	0.0459	-0.0007	3.1005
50.0	250	135.2641	0.4457	0.2306	-0.0037	3.0971
49.0	240	165.9845	0.6182	0.3199	-0.0048	3.0957
48.0	200	131.8757	0.8483	0.4391	-0.0070	3.0919
47.0	190	156.1348	1.1715	0.6064	-0.0094	3.0900
46.0	190	217.2681	1.6292	0.8439	-0.0132	3.0842
45.0	180	254.3295	2.2423	1.1618	-0.0175	3.0789
44.5	180	298.3714	2.6308	1.3629	-0.0209	3.0759
44.0	170	296.3270	3.0996	1.6068	-0.0246	3.0702
43.5	170	347.7139	3.6374	1.8854	-0.0286	3.0664
43.0	160	339.5097	4.2577	2.2081	-0.0335	3.0599
42.0	150	388.1907	5.8964	3.0641	-0.0472	3.0400
41.0	120	275.7958	8.1717	4.2519	-0.0644	3.0190

Table S2. Summary of set up and results for GCMC simulations of ethane in gas phase.

Set up		**<Average values> on production run**				
$-\mu_x10^{-3}$ [J/mol]	**L_{box} [Å]**	**N [molecules]**	**P_x10^{-5} [Pa]**	**ρ [kg/m3]**	**U_x10^{-3} [J/mol]**	**H_x10^{-3} [J/mol]**
				T=298K		
50.0	800	140.9071	0.0113	0.0137	-0.0003	2.4772
47.0	600	198.7329	0.0378	0.0459	-0.0012	2.4759
45.0	450	188.2112	0.0849	0.1031	-0.0026	2.4733
44.0	380	170.2132	0.1275	0.1549	-0.0039	2.4720
43.0	320	153.2162	0.1921	0.2335	-0.0061	2.4685
42.0	270	136.6795	0.2851	0.3467	-0.0089	2.4636
40.0	250	243.8382	0.6392	0.7792	-0.0206	2.4461
39.0	200	187.4139	0.9578	1.1698	-0.0300	2.4321
38.0	200	283.5131	1.4442	1.7695	-0.0449	2.4089
36.0	120	140.7264	3.2809	4.0664	-0.1031	2.3230
				T=300K		
60.0	3000	155.6066	0.0002	0.0003	$-1x10^{-5}$	2.4943
55.0	1500	145.0035	0.0018	0.0021	$-4x10^{-5}$	2.4942
50.0	800	162.7269	0.0132	0.0159	-0.0004	2.4935
48.0	800	360.9576	0.0292	0.0352	-0.0009	2.4929
47.0	700	363.3899	0.0439	0.0529	-0.0014	2.4924
45.0	400	151.1103	0.0977	0.1179	-0.0029	2.4895
44.0	350	151.7404	0.1465	0.1767	-0.0044	2.4880
43.0	350	226.0409	0.2180	0.2632	-0.0068	2.4835
40.0	250	276.4793	0.7290	0.8835	-0.0231	2.4580
39.0	250	417.8892	1.1004	1.3354	-0.0341	2.4437
38.0	200	317.7693	1.6282	1.9834	-0.0501	2.4184
37.0	150	202.6490	2.4481	2.9982	-0.0779	2.3775
36.0	140	249.7444	3.6803	4.5446	-0.1161	2.3190
35.0	130	307.0634	5.5763	6.9788	-0.1790	2.2237
34.0	120	374.8447	8.4900	10.8315	-0.2761	2.0809
33.0	100	344.7104	13.0314	17.2122	-0.4378	1.8388
				T=303K		
50.0	700	134.5716	0.0164	0.0196	-0.0006	2.5185
48.0	550	143.9910	0.0362	0.0432	-0.0011	2.5177
46.0	450	175.0864	0.0804	0.0959	-0.0021	2.5164
45.0	400	182.5067	0.1192	0.1424	-0.0035	2.5139
44.0	300	114.6321	0.1775	0.2120	-0.0056	2.5116
42.0	280	206.4089	0.3923	0.4695	-0.0121	2.5002
41.0	250	220.6505	0.5886	0.7051	-0.0177	2.4925
40.0	200	168.2144	0.8739	1.0499	-0.0272	2.4757
39.0	150	105.3240	1.2964	1.5582	-0.0413	2.4605
38.0	130	103.9093	1.9545	2.3616	-0.0593	2.4293
36.0	100	145.0179	4.4442	5.4403	-0.1384	2.3180
				T=309K		
55.0	1500	286.7039	0.0036	0.0042	-0.0001	2.5691
52.0	1000	271.5227	0.0116	0.0136	-0.0004	2.5685
50.0	800	175.0864	0.0254	0.0297	-0.0012	4.4742
48.0	600	280.4209	0.0554	0.0648	-0.0016	2.5668
45.0	400	267.2047	0.1779	0.2085	-0.0050	2.5612
44.0	300	166.7204	0.2630	0.3083	-0.0074	2.5576
43.0	300	245.6885	0.3872	0.4544	-0.0112	2.5515

41.0	250	309.5956	0.8410	0.9894	-0.0252	2.5308
				T=338K		
65.0	2000	138.5996	0.0008	0.0009	-0.0015	2.8103
60.0	1000	102.8617	0.0048	0.0051	-0.0083	2.8101
55.0	550	101.4438	0.0284	0.0304	-0.0767	2.8091
53.0	550	206.6980	0.0580	0.0620	-0.2985	2.8081
51.0	400	162.6755	0.1185	0.1269	-0.4965	2.8050
50.0	400	232.2556	0.1692	0.1812	-1.0129	2.8040
48.0	300	199.8417	0.3449	0.3696	-1.8125	2.7969
47.0	300	283.9496	0.4896	0.5251	-3.7447	2.7904
46.0	250	235.0759	0.6994	0.7512	-4.2179	2.7818
45.0	200	172.1543	1.0003	1.0745	-4.3862	2.7740
44.0	190	212.0929	1.4337	1.5440	-7.8905	2.7549
43.0	180	259.4668	2.0566	2.2215	-13.6766	2.7311
42.0	150	215.9201	2.9470	3.1945	-16.6055	2.6971
41.5	150	260.1832	3.5414	3.8494	-23.7969	2.6750
41.0	130	202.8228	4.2212	4.6097	-22.5405	2.6424
40.0	110	177.8193	6.0681	6.6709	-28.0957	2.5773
				T=373K		
65.0	1000	176.2436	0.0091	0.0088	-0.0002	3.1010
60.0	700	304.1368	0.0457	0.0443	-0.0010	3.0997
57.0	450	211.8173	0.1196	0.1161	-0.0027	3.0970
55.0	350	190.9326	0.2292	0.2224	-0.0053	3.0946
54.0	300	165.3015	0.3150	0.3057	-0.0067	3.0913
53.0	300	229.7384	0.4376	0.4249	-0.0098	3.0871
52.0	250	182.5896	0.6009	0.5835	-0.0134	3.0832
51.0	250	252.0095	0.8285	0.8053	-0.0186	3.0750
50.0	250	349.2396	1.1462	1.1161	-0.0256	3.0627
48.0	200	342.9191	2.1933	2.1403	-0.0490	3.0324
47.0	150	200.2002	3.0262	2.9619	-0.0667	3.0055
46.5	140	193.2681	3.5894	3.5169	-0.0801	2.9889
46.0	130	182.2057	4.2137	4.1411	-0.0938	2.9659
45.0	130	254.2466	5.8449	5.7784	-0.1320	2.9096

Table S3. Summary of set up and results for GCMC simulations of n-propane in gas phase.

Set up			<Average values> on production run			
$-\mu_x10^{-3}$ [J/mol]	L_{box} [Å]	N [molecules]	P_x10^{-5} [Pa]	ρ [kg/m3]	U_x10^{-3} [J/mol]	H_x10^{-3} [J/mol]
			T=298 K			
60.0	3000	232.7188	0.0004	0.0006	1.2304	3.7081
55.0	1500	218.7478	0.0027	0.0047	1.2253	3.7028
52.0	1100	290.0379	0.0090	0.0160	1.2190	3.6964
50.0	600	105.4706	0.0201	0.0358	1.2295	3.7075
48.0	600	236.9382	0.0451	0.0803	1.2152	3.6911
45.0	400	236.3230	0.1515	0.2704	1.2266	3.6972
43.0	300	224.9063	0.3410	0.6099	1.2077	3.6731
40.0	200	227.2921	1.1515	2.0803	1.1478	3.5886
37.0	100	103.3689	4.0188	7.5688	0.9559	3.2972
36.0	100	165.3918	6.2102	12.1102	0.7923	3.0535
			T=300 K			
60.0	2000	81.4330	0.0004	0.0007	1.2356	3.7282
58.0	2000	182.2871	0.0009	0.0017	1.2340	3.7284
55.0	1500	255.5171	0.0031	0.0056	1.2405	3.7232
52.0	1000	252.4027	0.0105	0.0185	1.2319	3.7263
50.0	800	288.4803	0.0233	0.0414	1.2350	3.7297
49.0	600	182.5983	0.0350	0.0618	1.2293	3.7252
46.0	400	181.2697	0.1172	0.2046	1.2078	3.7229
45.0	250	269.1129	0.1738	0.3061	1.2051	3.7101
43.0	200	147.2833	0.3884	0.6969	1.2200	3.6870
41.0	150	170.5433	0.8715	1.5577	1.1869	3.6400
40.0	150	108.3523	1.3060	2.3558	1.1540	3.6037
38.0	110	99.0811	2.9700	5.5063	1.0298	3.4502
37.0	100	116.3095	4.5519	8.5167	0.9350	3.2918
36.0	100	187.0116	6.9971	13.8028	0.7587	3.0092
			T=338 K			
63.0	1500	213.7069	0.0030	0.0046	1.3917	4.2019
60.0	1000	184.5304	0.0086	0.0135	1.3849	4.1950
58.0	800	191.1396	0.0174	0.0273	1.3836	4.1934
57.0	750	224.4636	0.0248	0.0390	1.3950	4.2042
56.0	600	164.7149	0.0356	0.0558	1.3910	4.1999
55.0	500	136.7777	0.0510	0.0801	1.3822	4.1907
53.0	400	141.6002	0.1032	0.1620	1.3827	4.1911
52.0	300	86.0508	0.1485	0.2334	1.3850	4.1902
51.0	300	122.7966	0.2117	0.3330	1.3722	4.1753
50.0	280	141.6909	0.3004	0.4726	1.3699	4.1728
48.0	250	208.8714	0.6205	0.9789	1.3594	4.1548
47.0	230	230.8225	0.8798	1.3892	1.3445	4.1373
46.0	210	254.0090	1.2657	2.0084	1.3155	4.0945
45.0	180	229.5254	1.8074	2.8819	1.3039	4.0695
44.5	170	232.1655	2.1655	3.4603	1.2838	4.0435
44.0	150	192.5753	2.6048	4.1782	1.2616	4.0107
43.0	140	229.9591	3.7749	6.1366	1.2119	3.9245
42.0	130	268.0718	5.3957	8.9347	1.1276	3.7906
			T=373 K			
70.0	1200	107.9920	0.0032	0.0046	1.5603	4.6620

65.0	700	107.5424	0.0161	0.0229	1.5523	4.6601
63.0	700	204.9436	0.0308	0.0438	1.5530	4.6535
60.0	400	100.9561	0.0812	0.1153	1.5529	4.6580
59.0	400	137.5942	0.1107	0.1571	1.5569	4.6634
58.0	350	129.0678	0.1548	0.2200	1.5406	4.6450
57.0	350	177.5374	0.2129	0.3026	1.5566	4.6598
56.0	300	154.3549	0.2939	0.4177	1.5487	4.6515
55.0	300	214.7438	0.4084	0.5811	1.5471	4.6462
54.0	250	170.8245	0.5607	0.7988	1.5275	4.6228
53.0	250	235.2991	0.7711	1.1003	1.5224	4.6128
52.0	200	167.5494	1.0712	1.5303	1.5134	4.6001
51.0	180	169.9442	1.4852	2.1291	1.4969	4.5729
50.0	150	136.9382	2.0647	2.9646	1.4615	4.5326
49.0	150	191.1073	2.8602	4.1463	1.4246	4.4665
48.0	140	217.7668	3.9804	5.8112	1.3797	4.4002
47.0	110	148.7099	5.5325	8.1813	1.3022	4.2842

Table S4. Summary of set up and results for GCMC simulations of n-butane in gas phase.

Set up		<Average values> on production run				
$-\mu_x10^{-3}$	L_{box}	N	P_x10^{-5}	ρ	U_x10^{-3}	H_x10^{-3}
[J/mol]	[Å]	[molecules]	[Pa]	[kg/m3]	[J/mol]	[J/mol]
				T=300K		
65.0	8000	159.9735	1.3E-05	3.0E-05	4.1895	6.6879
63.0	6000	151.6640	2.9E-05	6.8E-05	4.1770	6.6713
60.0	3500	100.2443	9.6E-05	2.2E-04	4.2089	6.7033
58.0	3000	140.8579	2.2E-04	5.0E-04	4.2188	6.7078
57.0	3000	209.3866	3.2E-04	7.5E-04	4.2233	6.7123
56.0	2600	203.6043	4.8E-04	1.1E-03	4.2290	6.7180
55.0	2500	270.3034	7.2E-04	1.7E-03	4.2222	6.7113
53.0	2000	309.5867	1.6E-03	3.7E-03	4.2137	6.7026
50.0	1000	129.0922	5.3E-03	1.2E-02	4.1943	6.6831
48.0	800	146.3780	1.2E-02	2.8E-02	4.2248	6.7141
46.0	700	218.8095	2.6E-02	6.2E-02	4.2073	6.6943
45.0	550	158.8851	4.0E-02	9.2E-02	4.2154	6.7025
43.0	400	136.6683	8.8E-02	2.1E-01	4.2102	6.6935
40.0	300	193.4703	3.0E-01	6.9E-01	4.1808	6.6565
38.0	200	130.5237	6.7E-01	1.6E+00	4.1458	6.5970
37.0	180	145.0413	1.0E+00	2.4E+00	4.0916	6.5231
36.5	150	102.5592	1.2E+00	2.9E+00	4.0582	6.4708
36.0	180	221.6522	1.5E+00	3.7E+00	4.0156	6.4184
35.5	150	158.6552	1.9E+00	4.5E+00	3.9661	6.3463
35.0	120	102.5403	2.3E+00	5.7E+00	3.9158	6.2673
				T=338K		
70.0	5500	212.8068	6.0E-05	1.2E-04	4.9948	7.7991
67.0	4000	236.6950	1.7E-04	3.6E-04	4.9989	7.8032
63.0	2000	123.4244	7.2E-04	1.5E-03	4.9881	7.7923
60.0	1800	262.2336	2.1E-03	4.3E-03	5.0076	7.8116
58.0	1500	307.4264	4.3E-03	8.8E-03	4.9994	7.8034
55.0	900	193.2906	1.2E-02	2.6E-02	4.9832	7.7875
53.0	700	185.4873	2.5E-02	5.2E-02	4.9600	7.7640
50.0	400	101.0568	7.4E-02	1.5E-01	4.9477	7.7529
48.0	350	136.1815	1.5E-01	3.1E-01	4.9713	7.7740
47.0	300	123.4237	2.1E-01	4.4E-01	4.9990	7.8058
46.0	250	102.2263	3.0E-01	6.3E-01	4.9791	7.7784
45.0	200	75.1176	4.3E-01	9.0E-01	4.9595	7.7428
44.0	200	107.7707	6.2E-01	1.3E+00	4.9435	7.7136
43.5	180	129.5102	7.5E-01	1.6E+00	4.9186	7.6899
43.0	150	133.3290	9.0E-01	1.9E+00	4.9184	7.6869
42.0	150	165.5686	1.3E+00	2.7E+00	4.8694	7.6193
41.0	100	201.8362	1.9E+00	4.0E+00	4.8144	7.5299

40.0	100	247.4099	2.7E+00	5.8E+00	4.7301	7.4133
				T=373K		
75.0	4500	297.9745	1.7E-04	3.2E-04	8.8486	8.8486
73.0	3000	167.9641	3.2E-04	6.0E-04	8.8378	8.8378
71.0	2700	233.1518	6.1E-04	1.1E-03	8.8435	8.8435
70.0	2500	256.7499	8.5E-04	1.6E-03	5.7601	8.8615
68.0	1400	86.5065	1.6E-03	3.0E-03	5.6721	8.7691
65.0	1200	141.1246	4.2E-03	7.9E-03	5.6782	8.7813
64.0	1000	112.9043	5.8E-03	1.1E-02	5.7060	8.8023
62.0	900	157.3200	1.1E-02	2.1E-02	5.7018	8.7965
60.0	800	210.0150	2.1E-02	3.9E-02	5.6753	8.7745
58.0	600	170.6102	4.1E-02	7.6E-02	5.6942	8.7967
56.5	500	157.4826	6.5E-02	1.2E-01	5.6806	8.7776
55.0	400	132.1386	1.1E-01	2.0E-01	5.6961	8.7948
53.0	300	106.1715	2.0E-01	3.8E-01	5.6690	8.7643
50.0	200	83.4167	5.4E-01	1.0E+00	0.0000	3.0979
49.0	190	99.4377	7.4E-01	1.4E+00	5.5914	8.6564
48.0	190	139.7757	1.0E+00	2.0E+00	5.6090	8.6790
47.0	180	163.7318	1.4E+00	2.7E+00	5.5579	8.6010
46.5	170	163.1366	1.7E+00	3.2E+00	5.5720	8.6012
46.0	160	159.9960	2.0E+00	3.8E+00	5.5189	8.5376
45.5	150	158.0847	2.4E+00	4.5E+00	5.4844	8.5211
45.0	150	185.5823	2.7E+00	5.3E+00	5.4803	8.4793
44.0	120	135.9680	3.9E+00	7.6E+00	5.4236	8.3973
43.0	110	151.0320	5.5E+00	1.1E+01	5.2857	8.1894
42.5	110	184.6375	6.6E+00	1.3E+01	5.1590	8.0143
42.0	100	169.7429	7.9E+00	1.6E+01	5.0265	7.8217
41.5	100	209.9740	9.5E+00	2.0E+01	4.8519	7.5866
41.0	90	194.0684	1.2E+01	2.6E+01	4.6529	7.2812
40.7	90	227.1812	1.3E+01	3.0E+01	4.8059	6.9706

Table S5. Summary of set up and results for GCMC simulations of n-pentane in gas phase

Set up		**<Average values> on production run**				
-μx10^{-3}	**L_{box}**	**N**	**Px10^{-5}**	**ρ**	**Ux10^{-3}**	**Hx10^{-3}**
[J/mol]	**[A_0]**	**[molecules]**	**[Pa]**	**[kg/m^3]**	**[J/mol]**	**[J/mol]**
			T=300K			
63.0	6000	99.8582	0.00002	0.00006	8.1719	10.6663
61.0	5500	170.8762	0.00004	0.00012	8.1698	10.6641
60.0	5000	192.6181	0.00006	0.00018	8.1870	10.6813
59.0	4500	210.7449	0.00010	0.00028	8.1521	10.6465
58.0	4000	219.3175	0.00014	0.00041	8.1674	10.6617
57.0	3500	220.4019	0.00021	0.00062	8.1852	10.6795
55.0	2700	224.5402	0.00047	0.00137	8.1724	10.6667
53.0	2000	204.3370	0.00106	0.00306	8.1656	10.6598
50.0	1200	146.2082	0.00351	0.01014	8.1855	10.6803
48.0	1000	188.7099	0.00781	0.02261	8.1694	10.6632
47.0	800	144.6515	0.01170	0.03385	8.1654	10.6590
46.0	800	215.8013	0.01745	0.05050	8.1785	10.6719
44.0	600	203.0469	0.03889	0.11262	8.1688	10.6601
			T=303K			
63.0	6000	132.3525	2.56E-5	7.34E-5	8.2524	10.7716
61.0	5500	224.6375	5.65E-5	0.00016	8.2500	10.7692
60.0	5000	251.7758	8.43E-5	0.00024	8.2366	10.7558
59.0	4500	272.636	0.00013	0.00036	8.2419	10.7612
55.0	2700	288.304	0.00061	0.00175	8.2631	10.7823
53.0	2000	259.8782	0.00136	0.00389	8.2508	10.7700
50.0	1200	183.6915	0.00445	0.01274	8.2427	10.7615
49.0	1100	211.1018	0.00663	0.01900	8.2540	10.7729
48.0	1000	236.5272	0.00989	0.02834	8.2658	10.7843
47.0	800	179.5242	0.01466	0.04201	8.2470	10.7652
			T=323K			
65.0	3500	72.00799	7.49E-5	0.00029	8.82792	11.5135
63.0	3000	95.50574	0.00016	0.00042	8.82813	11.5137
60.0	2500	169.0155	0.00048	0.0013	8.82808	11.5136
59.0	2100	145.3085	0.0007	0.00188	8.82126	11.5067
58.0	1800	133.1318	0.00102	0.00273	8.83983	11.5254
55.0	1500	236.2611	0.00312	0.05290	8.82732	11.5124
53.0	1100	195.6026	0.01761	0.11790	8.81574	11.5004
50.0	800	229.8987	0.02002	0.05380	8.82883	11.5134
48.0	500	118.2302	0.04214	0.11332	8.81417	11.4969
45.0	450	266.653	0.13010	0.35058	8.79535	11.4728
			T=343K			
68.0	3500	126.3555	0.00014	0.00035	9.40934	12.2612
65.0	2500	131.4373	0.0004	0.00101	9.41579	12.2676

64.0	2200	127.4034	0.00057	0.00143	9.40928	12.2611
62.0	1600	98.23065	0.00114	0.00287	9.3972	12.2489
60.0	1400	133.3320	0.0023	0.00582	9.41354	12.2653
58.0	1200	169.0005	0.00463	0.01172	9.39921	12.2508
56.0	1100	261.7189	0.00931	0.02356	9.3885	12.2394
55.0	900	204.4458	0.01327	0.0336	9.40601	12.2563
52.0	600	173.3709	0.03798	0.09616	9.38760	12.2370

Table S6. Summary of set up and results for GCMC simulations of 2-methyl-pentane in gas phase

Set up		**<Average values> on production run**				
$-\mu_x 10^{-3}$	**L_{box}**	**N**	**$P_x 10^{-5}$**	**ρ**	**$U_x 10^{-3}$**	**$H_x 10^{-3}$**
[J/mol]	**[Å]**	**[molecules]**	**[Pa]**	**[kg/m3]**	**[J/mol]**	**[J/mol]**
				T=423K		
75.0	1100	15.1725	0.0003	0.0005	20.7033	24.2263
70.0	900	13.5374	0.0011	0.0022	20.6079	24.1102
67.0	800	22.3799	0.0026	0.0052	20.6676	24.1808
65.0	700	26.7378	0.0046	0.0093	20.6373	24.1523
60.0	600	68.8096	0.0186	0.0382	20.6514	24.1675
57.0	600	162.1476	0.0438	0.0899	20.5843	24.0980
55.0	500	166.2144	0.0777	0.1593	20.5884	24.1073
54.0	400	114.1535	0.1040	0.2137	20.7093	24.2205
52.0	350	134.3390	0.1830	0.3754	20.6638	24.1810
50.0	250	87.1186	0.3240	0.6680	20.6462	24.1456
47.0	200	105.8514	0.7630	1.5853	20.5209	23.9935
45.0	100	24.3190	1.3800	2.9137	20.4234	23.8407

Table S7. Summary of set up and results for GCMC simulations of n-hexane in gas phase.

Set up		<Average values> on production run				
$-\mu_x10^{-3}$	L_{box}	N	P_x10^{-5}	ρ	U_x10^{-3}	H_x10^{-3}
[J/mol]	[Å]	[molecules]	[Pa]	[kg/m3]	[J/mol]	[J/mol]
			T=300K			
73.0	20000	21.8844	1.1E-07	3.9E-7	11.4044	13.8988
65.0	10000	67.7629	2.8E-06	9.7E-6	11.3830	13.8773
64.0	10000	100.6350	4.2E-06	1.4E-05	0.0000	13.8653
62.0	9000	163.3849	9.3E-06	3.2E-05	10.9634	13.8648
60.0	8000	255.9817	2.1E-05	7.2E-05	7.2574	13.8628
57.0	4000	107.1259	6.9E-05	2.4E-04	11.3711	13.8655
53.0	3000	222.6167	3.4E-04	1.2E-03	11.3744	13.8688
50.0	2000	220.7939	1.1E-03	3.9E-03	11.4683	13.8497
48.0	1500	207.2817	2.5E-03	8.8E-03	11.3702	13.8647
47.0	1200	159.1309	3.8E-03	1.3E-02	11.3729	13.8669
45.0	1000	204.7995	8.5E-03	2.9E-02	11.3803	13.8734
42.0	600	148.4702	2.8E-02	9.8E-02	11.3555	13.8447
			T=303K			
60.0	6000	141.9201	2.7E-05	9.4E-05	11.4731	13.9923
57.0	4000	139.3243	9.1E-05	3.1E-04	11.4729	13.9922
53.0	3000	286.6590	4.4E-04	1.5E-03	11.4808	14.0000
50.0	1900	240.0986	1.5E-03	5.0E-03	11.4797	13.9990
48.0	1500	259.9204	3.2E-03	1.1E-02	11.4790	13.9977
47.0	1200	199.0786	4.8E-03	1.6E-02	11.4982	14.0170
45.0	1000	254.8045	1.1E-02	3.6E-02	11.4704	13.9882
42.0	600	182.0966	3.5E-02	1.2E-01	11.4783	13.9943
			T=343K			
75.0	4100	6.4088	4.4E-06	1.3E-05	13.1547	16.0123
73.0	3400	7.3589	8.9E-06	2.7E-05	13.0305	15.8881
70.0	3000	14.5902	2.6E-05	7.7E-05	13.0704	15.9280
68.0	2700	20.7089	5.0E-05	1.5E-04	13.0657	15.9234
65.0	2500	47.3739	1.4E-04	4.3E-04	13.0823	15.9399
63.0	2300	74.5085	2.9E-04	8.7E-04	13.0583	15.9159
60.0	2000	138.7836	8.2E-04	2.5E-03	13.1322	15.9901
-57.0	1500	118.4447	1.7E-03	5.0E-03	13.1194	15.9770
55.0	1000	73.6301	4.8E-03	1.4E-02	13.1548	16.0122
53.0	700	69.2822	9.6E-03	2.9E-02	13.0983	15.9566
50.0	700	200.2961	2.8E-02	8.3E-02	13.0944	15.9493
48.0	500	145.3230	5.5E-02	1.7E-01	13.1208	15.9750
45.0	300	91.8514	1.6E-01	4.9E-01	13.0660	15.9071
43.0	200	56.0499	3.3E-01	1.0E+00	13.0512	15.8775
40.0	100	21.0369	9.5E-01	3.0E+00	12.8746	15.6136

T=373K						
67.0	2200	147.2218	7.1E-04	2.0E-03	14.2605	17.3617
65.0	2000	211.5092	1.4E-03	3.8E-03	14.2545	17.3556
63.0	1800	292.6910	2.6E-03	7.2E-03	14.2684	17.3694
60.0	1100	176.9704	6.8E-03	1.9E-02	14.2646	17.3663
59.0	1000	184.8791	9.5E-03	2.6E-02	14.2353	17.3370
58.0	900	186.4439	1.3E-02	3.7E-02	14.2601	17.3609
57.0	800	179.1652	1.8E-02	5.0E-02	14.2651	17.3662
56.0	700	166.7742	2.5E-02	7.0E-02	14.2616	17.3615
55.0	650	144.9034	3.5E-02	9.6E-02	14.2938	17.3929
53.0	500	160.2331	6.6E-02	1.8E-01	14.2560	17.3537
52.0	450	161.0946	9.1E-02	2.5E-01	14.2464	17.3422
51.0	400	157.0130	1.3E-01	3.5E-01	14.2334	17.3248

Table S8. Summary of set up and results for GCMC simulations of carbon dioxide in gas phase

Set up		<Average values> on production run				
$-\mu_x10^{-3}$	L_{box}	N	P_x10^{-5}	ρ	U_x10^{-3}	H_x10^{-3}
[J/mol]	[Å]	[molecules]	[Pa]	[kg/m3]	[J/mol]	[J/mol]
			T=277K			
50.0	1000	94.6870	0.0036	0.0069	-0.0001	2.3029
45.0	500	103.4269	0.0316	0.0605	-0.0008	2.3021
44.0	400	81.3979	0.0486	0.0924	-0.0014	2.3007
43.0	400	126.1206	0.0753	0.1440	-0.0024	2.2992
42.0	350	129.8824	0.1158	0.2214	-0.0031	2.2992
40.0	300	196.5912	0.2780	0.5321	-0.0079	2.2914
38.0	200	138.6002	0.6600	1.2661	-0.0204	2.2738
37.0	150	91.2502	1.0264	1.9758	-0.0316	2.2545
36.0	140	115.5092	1.5942	3.0763	-0.0485	2.2322
33.0	120	281.2626	5.9957	11.8949	-0.1881	2.0302
32.0	70	89.2383	9.3628	19.0129	-0.3016	1.8656
31.0	70	152.1977	15.4145	32.4269	-0.5095	1.5826
			T=305K			
55.0	1000	111.6231	0.0047	0.0082	-0.0001	2.5357
50.0	500	100.2443	0.0338	0.0581	-0.0009	2.5346
48.0	400	112.6471	0.0741	0.1286	-0.0017	2.5349
45.0	300	155.6022	0.2424	0.4212	-0.0061	2.5267
43.0	200	102.0050	0.5358	0.9318	-0.0130	2.5176
42.0	200	151.6520	0.7954	1.3853	-0.0197	2.5070
40.0	150	141.3110	1.7506	3.0598	-0.0450	2.4729
			T=308K			
55.0	1000	140.2044	0.0060	0.0102	-0.0001	2.5606
50.0	600	211.9073	0.0417	0.0717	-0.0007	2.5595
48.0	400	137.8963	0.0916	0.1575	-0.0020	2.5583
46.0	250	73.6451	0.2003	0.3444	-0.0048	2.5549
45.0	250	108.4935	0.2951	0.5074	-0.0068	2.5529
44.0	250	161.0585	0.4377	0.7532	-0.0097	2.5476
42.0	160	92.5115	0.9555	1.6505	-0.0230	2.5247
41.0	160	137.1914	1.4169	2.4477	-0.0352	2.5124
40.0	150	167.3968	2.0919	3.6246	-0.0497	2.4903
38.0	120	190.4177	4.6052	8.0529	-0.1114	2.4053
37.0	100	166.3789	6.8625	12.1588	-0.1722	2.3117
36.0	80	131.3061	10.4562	18.7416	-0.2663	2.1890
35.0	60	201.5085	15.6367	28.7617	-0.4081	1.9845
			T=353K			

60.0	600	103.9940	0.0235	0.0352	-0.00040	2.93496
55.0	300	72.0987	0.1301	0.1951	-0.00243	2.93208
53.0	250	82.0708	0.2558	0.3838	-0.00467	2.92788
50.0	200	117.1525	0.7124	1.0702	-0.01271	2.91704
49.0	190	140.5513	0.9953	1.4975	-0.01867	2.90626
48.0	170	143.1077	1.4147	2.1287	-0.02717	2.89769
47.0	150	137.1864	1.9713	2.9705	-0.03801	2.88261
45.0	120	140.5872	3.9234	5.9456	-0.07386	2.83022
43.0	100	166.4088	7.9577	12.1609	-0.15159	2.72821
42.0	80	121.1436	11.1996	17.2910	-0.21331	2.63720
40.0	60	109.2931	23.0207	36.9769	-0.45867	2.28120
38.0	50	141.9741	47.5892	83.0023	-1.02271	1.50054

Table S9. Summary of set up and results for GCMC simulations of methanol in gas phase

Set up		<Average values> on production run				
$-\mu_x 10^{-3}$	L_{box}	N	$P_x 10^{-5}$	ρ	$U_x 10^{-3}$	$H_x 10^{-3}$
[J/mol]	[Å]	[molecules]	[Pa]	[kg/m3]	[J/mol]	[J/mol]
			T=303K			
65.0	3000	30.3829	5E-05	6E-05	1.2429	3.7635
60.0	2000	66.3230	0.0003	0.0004	1.2508	3.7713
55.0	1500	202.3240	0.0025	0.0032	1.2652	3.7855
54.0	1300	196.1296	0.0037	0.0047	1.2532	3.7731
53.0	1000	133.1017	0.0056	0.0071	1.2705	3.7896
52.0	900	143.6560	0.0082	0.0105	1.2636	3.7843
50.0	750	183.1476	0.0182	0.0231	1.2530	3.7727
48.0	600	211.0229	0.0407	0.0520	1.1210	3.6290
47.0	400	92.0837	0.0600	0.0766	1.2157	3.7265
45.0	350	141.6321	0.1437	0.1758	0.9580	3.5779

In order to show the uncertainties obtained in the gas phase simulations, the results of three independent GCMC simulations of n-butane are shown in Table 10 and also in Figure S1. It can be observed in Figure S1 that points of each independent run are essentially on top of one another. The average relative deviation on pressure and density among the three independent simulations are 0.46% and 0.37%. A small average relative deviation was also observed for the other components.

Table S10. Summary of set up and results of three independent GCMC simulations for n-butane in gas phase at 373K.

	Run1			Run2			Run3		
$-\mu_x 10^{-3}$ [J/mol]	L_{box} [Å]	N molec	$P_x 10^{-5}$ [Pa]	L_{box} [Å]	N molec	$P_x 10^{-5}$ [Pa]	L_{box} [Å]	N molec	$P_x 10^{-5}$ [Pa]
75.0	4500	297.97	0.0002	1800	19.08	0.0002	3500	139.19	0.0002
73.0	3000	167.96	0.0003	1700	30.73	0.0003	2700	97.88	0.0003
71.0	2700	233.15	0.0006	1700	58.24	0.0006	2000	94.77	0.0006
70.0	2500	256.75	0.0016	1600	67.16	0.0008	2000	131.34	0.0008
68.0	2000	249.76	0.0016	1400	86.51	0.0016	1200	53.95	0.0016
65.0	1500	276.63	0.0042	1200	141.12	0.0042	1000	82.28	0.0042
64.0	1200	195.26	0.0058	1000	112.90	0.0058	900	82.37	0.0058
62.0	1000	216.09	0.0111	900	157.32	0.0111	800	110.13	0.0111
60.0	900	300.07	0.0212	800	210.01	0.0211	600	88.80	0.0212
58.0	700	268.31	0.0403	600	170.61	0.0407	500	98.10	0.0404
56.5	600	274.35	0.0654	500	157.48	0.0648	300	34.53	0.0658
55.0	500	258.89	0.1066	400	132.14	0.1062	250	32.47	0.1068
53.0	400	252.36	0.2024	300	106.17	0.2020	200	31.69	0.2035

50.0	300	282.05	0.5348	200	83.42	0.5345	150	35.48	0.5378
49.0	250	225.56	0.7373	190	99.44	0.7380	150	49.07	0.7421
48.0	200	161.15	1.0258	190	139.78	1.0387	120	35.16	1.0351
47.0	190	192.68	1.4236	180	163.73	1.4208	110	37.65	1.4308
46.5	180	194.63	1.6823	170	163.14	1.6739	110	44.57	1.6969
46.0	170	192.54	1.9735	160	160.00	1.9617	110	52.47	1.9857
45.5	170	228.90	2.3321	150	158.08	2.3599	110	62.08	2.3276
45.0	160	228.70	2.7848	150	185.58	2.7390	110	73.82	2.7548
44.0	120	135.97	3.8854	100	77.98	3.8411	110	105.29	3.8854
43.0	110	151.03	5.4713	50	39.10	5.4736	100	114.26	5.4974
42.5	110	184.64	6.5771	50	17.70	6.8262	90	100.87	6.5281
42.0	100	169.74	7.8785	50	21.15	7.9219	80	87.23	7.9231
41.5	100	209.97	9.5349	50	26.81	9.6878	70	107.22	9.5127
41.0	90	194.07	11.618	50	33.84	11.741	100	265.16	11.600
40.7	90	227.18	13.086	50	38.84	13.143	60	68.17	13.467

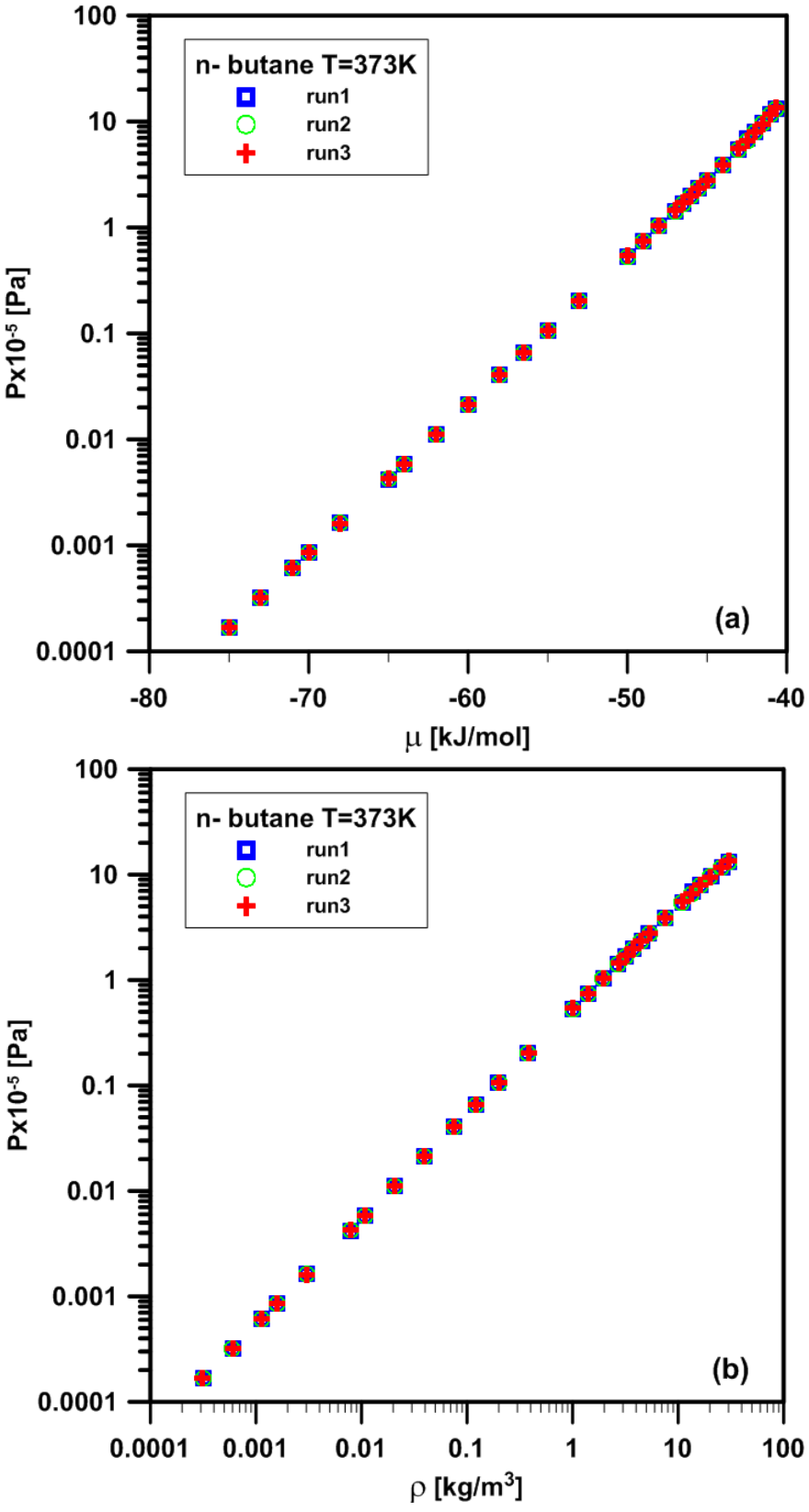

Figure S1. Results of three independent runs of GCMC simulations of n-butane in gas phase at 373K.

Figure S2 presents the results of gas phase simulation of alkanes at 300K.

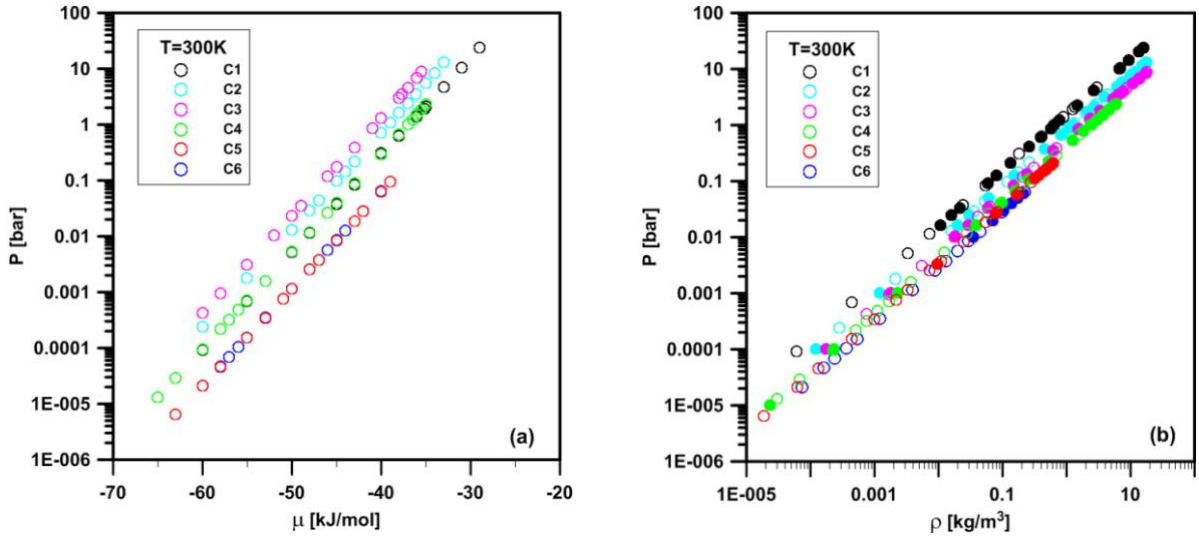

Figure S2. Results of GCMC simulation in the fluid phase at 300K. (a)- Pressure vs. chemical potential (μ) and (b)-Pressure vs. density (ρ). The color black, cyan, magenta, green, red and blue indicate the components. Open symbols are simulation and closed symbols are NIST density reference data.

1.2 – Mixtures

The values of chemical potential of each component in the gas mixture were taken considering the mixture as an ideal solution according to the following equation

$$\mu_i^{id}(T, P, y_i) = \mu_i(T, P) + RTlny_i \qquad \text{S1}$$

The results obtained for the binary mixtures of methane+ethane, methane+propane, methane+n-butane, ethane+propane, 2methylpentane+n-hexane are in Tables S11, S12, S13 and S14, respectively.

Table S11. Summary of set up and main results of GCMC simulations in the gas phase for binary mixtures of methane(1)+ethane(2) at 300K.

Set up			<Average values> on production run			
-μx10^{-3} [J/mol]		L_{box} [Å]	Number of molecules		Px10^{-5} [Pa]	y_1
C1	C2		C1	C2		
39.41	36.38	100	8.7791	78.0215	3.5203	0.1011
37.68	36.67	100	17.8331	69.5235	3.5518	0.2041
36.72	37.00	100	25.9030	60.3400	3.5178	0.3003
35.96	37.39	100	34.7957	51.6212	3.5223	0.4026
35.40	37.84	100	43.4744	43.2361	3.5424	0.5014
35.26	38.00	100	52.8940	33.2458	3.5295	0.6140
34.26	40.00	100	69.1572	17.8566	3.5778	0.7948
34.00	41.00	100	76.4204	12.0201	3.6397	0.8641
33.92	42.00	100	78.9301	8.1046	3.5842	0.9069
33.74	45.00	100	84.0007	2.3643	3.5561	0.9726

The results for the ternary mixtures of methane+ethane+propane, methane+ethane+butane and methane+propane+butane are presented in Tables S15, S16 and S17, respectively.

Table S12. Summary of set up and main results of GCMC simulations in the gas phase for binary mixtures of methane(1)+propane(2) at 300K.

Set up			<Average values> on production run			
-μx10^{-3} [J/mol]		L_{box} [Å]	Number of molecules		Px10^{-5} [Pa]	y_1
C1	C3		C1	C3		
39.41	38.00	100	8.8407	74.6726	3.3709	0.1059
36.90	38.50	100	23.8827	61.1283	3.4280	0.2809
36.00	38.75	100	34.1219	54.2465	3.5652	0.3861
35.75	39.00	100	38.1274	49.6807	3.5508	0.4342
35.20	39.50	100	47.7929	40.2271	3.5747	0.5430
34.86	40.00	100	54.0395	32.6080	3.5327	0.6237
34.39	41.00	100	65.1080	21.8213	3.5543	0.7490
34.14	42.00	100	71.8968	14.6413	3.5486	0.8308
33.97	43.00	100	77.3283	9.6648	3.5695	0.8889
33.80	45.00	100	82.1157	4.4162	3.5645	0.9490
33.69	50.00	100	87.3096	0.5602	3.6152	0.9936
33.67	52.00	100	85.9695	0.2375	3.5503	0.9972

Table S13. Summary of set up and main results of GCMC simulations in the gas phase for binary mixtures of methane(1)+n-butane(2) at 300K.

Set up			<Average values> on production run			
$-\mu x10^{-3}$ [J/mol]		L_{box} [Å]	Number of molecules		$Px10^{-5}$ [Pa]	y_1
C1	C4		C1	C4		
35.41	37.00	100	43.4806	40.3809	3.3852	0.5185
35.00	37.50	100	51.3054	32.5983	3.3848	0.6115
34.50	38.00	100	62.1060	26.4287	3.5847	0.7015
34.50	37.95	100	63.5531	26.9945	3.6778	0.7019
34.50	38.60	100	61.7777	20.3296	3.3490	0.7524
34.10	40.50	100	72.6162	9.4170	3.3690	0.8852
33.80	43.00	100	83.2920	3.4303	3.5739	0.9604
33.70	46.00	100	86.8197	1.0133	3.6160	0.9885
33.67	48.00	200	293.1139	1.5752	3.5944	0.9947

The results of GCMC simulations of a quaternary mixture of methane(1)+ethane(2)+propane(3)+n-butane(4) in gas phase at 300K are presented in Table S19.

Table S14. Summary of set up and main results of GCMC simulations in the gas phase for binary mixtures of ethane(1)+propane(2) at 300K.

Set up			<Average values> on production run			
$-\mu x10^{-3}$ [J/mol]		L_{box} [Å]	Number of molecules		$Px10^{-5}$ [Pa]	y_1
C2	C3		C2	C3		
41.84	37.931	150	30.0264	264.6944	3.4638	0.1019
40.11	38.21	150	59.8369	235.1951	3.4746	0.2028
39.1	38.54	150	89.4343	204.8533	3.4767	0.3039
38.39	38.92	150	119.1903	175.0254	3.4842	0.4051
37.83	39.38	150	148.7100	145.4789	3.4958	0.5055
37.37	39.94	150	177.6706	115.6848	3.4873	0.6057
36.87	40.96	150	207.4543	86.5457	3.5063	0.7056
36.66	41.66	150	235.7826	57.3047	3.4982	0.8045
36.36	43.39	150	267.0117	28.6500	3.5434	0.9031

Table S15. Summary of set up and main results of GCMC simulations in the gas phase for binary mixtures of 2-methyl-pentane(1)+n-hexane(2) at 423K.

Set up			<Average values> on production run			
$-\mu x10^{-3}$ [J/mol]		L_{box} [Å]	Number of molecules		$Px10^{-5}$ [Pa]	y_1
C2	C3		C2	C3		
76.29	64.48	600	24.376	219.376	0.0674	0.1000
73.79	64.90	600	48.07	191.376	0.0662	0.2008
72.33	65.38	600	71.825	169.827	0.0668	0.2972
71.30	65.94	600	95.645	146.055	0.0668	0.3957
70.50	66.60	600	119.404	120.120	0.0662	0.4985
69.84	67.40	600	143.955	96.918	0.0666	0.5976
69.28	68.43	600	166.608	73.768	0.0665	0.6931
68.80	69.89	600	186.621	48.324	0.0651	0.7943
68.38	72.39	600	214.140	24.340	0.0660	0.8979

Table S16. Summary of set up and main results of GCMC simulations in the gas phase for ternary mixtures of methane(1)+ethane(2)+propane(3) at 300K.

Set up				<Average values> on production run					
$-\mu x10^{-3}$ [J/mol]			L_{box}	Number of molecules			$Px10^{-5}$	y_1	y_2
C1	C2	C3	[Å]	C1	C2	C3	[Pa]		
39.41	37.13	41.22	160	36.519	237.378	83.775	3.52	0.1021	0.6637
37.68	37.43	41.52	160	72.010	208.781	73.356	3.51	0.2033	0.5895
36.67	37.76	41.85	160	108.548	184.493	64.809	3.55	0.3033	0.5156
35.96	38.14	42.23	160	143.754	157.312	54.997	3.53	0.4037	0.4418
35.40	38.60	42.69	160	180.234	130.225	45.879	3.55	0.5058	0.3655
34.94	39.15	43.25	160	214.532	103.704	36.626	3.55	0.6045	0.2922
34.56	39.87	43.96	160	250.135	77.608	27.342	3.55	0.7044	0.2186
34.23	40.88	44.97	160	286.256	51.904	18.105	3.57	0.8035	0.1457
33.93	42.61	46.70	160	323.504	25.895	9.003	3.59	0.9026	0.0723

Table S17. Summary of set up and main results of GCMC simulations in the gas phase for ternary mixtures of methane(1)+ethane(2)+n-butane(3) at 300K.

Set up				<Average values> on production run					
$-\mu x10^{-3}$ [J/mol]			L_{box}	Number of molecules			$Px10^{-5}$	y_1	y_2
C1	C2	C4	[Å]	C1	C2	C4	[Pa]		
36.28	37.78	39.98	160	104.359	150.609	26.554	3.39	0.3707	0.5350
35.07	38.83	41.54	160	166.522	99.981	17.695	3.44	0.5859	0.3518
34.56	39.68	41.88	160	205.752	68.700	12.383	3.48	0.7173	0.2395
34.27	40.54	42.67	160	232.405	48.869	9.145	3.53	0.8002	0.1683
34.10	41.31	43.30	160	248.592	35.957	7.102	3.55	0.8524	0.1233

Table S18. Summary of set up and main results of GCMC simulations in the gas phase for ternary mixtures of methane(1)+propane(2)+n-butane(3) at 300K.

Set up				<Average values> on production run					
-μx10^{-3} [J/mol]			L_{box}	Number of molecules			Px10^{-5}	y_1	y_2
C1	C3	C4	[Å]	C1	C3	C4	[Pa]		
36.44	40.16	37.86	100	28.925	31.379	28.952	3.56	0.3241	0.3516
35.17	41.11	38.85	100	47.848	21.230	19.016	3.57	0.5431	0.2410
34.62	41.95	39.87	100	59.411	14.984	12.545	3.53	0.6834	0.1723
34.29	42.83	40.79	100	68.106	10.616	8.539	3.56	0.7805	0.1217
34.10	43.62	41.62	100	73.729	7.611	6.051	3.58	0.8437	0.0871

Table S19. Summary of set up and main results of GCMC simulations in the gas phase for quaternary mixtures of methane(1)+ethane(2)+propane(3)+n-butane(4) at 300K.

Set up					<Average values> on production run				
-μx10^{-3} [J/mol]				L_{box}	Number of molecules				Px10^{-5}
C1	C2	C3	C4	[Å]	C1	C2	C3	C4	[Pa]
36.23	38.96	41.91	39.81	200	250.431	220.240	121.084	101.100	3.50
35.02	40.04	42.92	40.89	200	406.670	142.704	82.533	67.015	3.56
34.52	41	43.78	41.85	200	500.868	95.727	57.797	44.749	3.58
34.24	41.89	44.55	42.68	200	552.486	66.361	42.289	31.816	3.55
34.08	42.7	45.21	43.4	200	590.060	48.501	31.780	23.645	3.57

2- RESULTS OF GCMC SIMULATIONS IN ADSORBED PHASE

2.1 – Adsorption in MFI-type zeolite

2.1.1- Isotherms of pure components

The results of GCMC simulations performed using LJ parameters proposed by June et al. to describe the interaction site of MFI with the combining rule of Lorentz-Berthelot (LB) and the proposed size weighted (SW) combining rule for methane, ethane, propane, n-butane, n-pentane, 2-methyl pentane, n-hexane, carbon dioxide and methanol are presented in Tables S20, S21, S22, S23, S24, S25, S26, S27 and S28, respectively. All results simulations shown were performed using tail correction. The n-butane GCMC runs applied the intramolecular intra scaling factor (0 0 0.5 1.0) while the simulations for longer alkanes did not applied. The reason was that conditions were those for which the LB rule presented the best performance. These results were used to compute the absolute mean deviation and also the relative mean deviation. The simulation results obtained using the June et al. force field to zeolite with Lorentz-Berthelot combining rule were named LB, while the

name SW is related to results using the same force field to zeolite but with the proposed combining rule.

The simulation results for methane (C1) and ethane (C2) in silicalite are shown in Figure S3. For the sake of clarity, the results for methane adsorption are presented at just two temperatures. The results are compared to experimental data and previous simulation results. Both the LB and SW combining rules give very satisfactory agreement with the experimental data, although SW is slightly more accurate, especially at 300 K. Also, the results agree well with the simulation results using the tuned cross interaction parameters proposed by Du et al.[1] The results for adsorption of propane (C3) and n-butane (C4) in silicalite at the three studied temperatures are presented in Figure S4. The use of the SW combining rule gave slightly better agreement with experimental adsorption data for C3 than the LB combining rule. However, for adsorption of n-butane (C4) the use of the LB combining rule resulted in slightly better agreement with experimental data than SW.

The results obtained for adsorption of longer alkanes n-pentane (C5) and n-hexane (C6) are presented in Figure S5. Both combining rules (LB and SW) gave similar performance; they are less accurate than for the shorter alkanes, but are in agreement with previously reported simulation results.[5,7] In general, the simulation results overestimate the adsorption amount in the range of very low pressure.

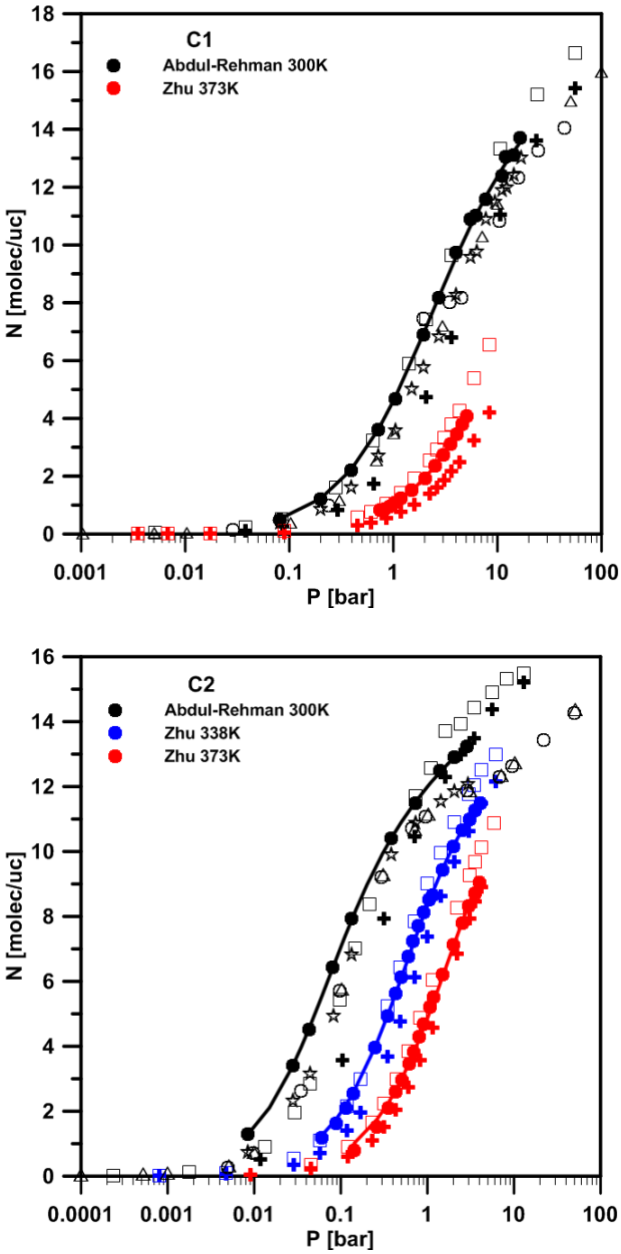

Figure S3. Adsorption isotherms for methane (top) and ethane (bottom) in silicalite. The colors black, blue and red denote temperature of 300K, 338K and 373K, respectively. The closed circles are experimental data reported by Abdul-Rehman et al.[2] and Zhu et al.[3] The open squares and crosses are GCMC simulations of this work using combining rules SW and LB, respectively. The open circles, triangles and stars are previous GCMC simulations reported by Du et al.[1], Krishna et al.[4] and Lu et al.[5], respectively. The continuous lines represent the fit of experimental data using the Dual Site Langmuir (DSL) model.

Computed adsorption isotherms for CO_2 in silicalite using the LB and SW combining rules are presented in Figure S6. The simulated CO_2 isotherms at 277, 305, 308 and 353K are in good agreement with experimental data reported by Sun et al.[6] and Choudhary and Mayadevi[7] as well as with prior GEMC simulations using the TraPPE-zeo force field[8]. At higher pressures, the TraPPE-

zeo force field slightly underestimates the CO_2 loading while SW slightly overestimates it.

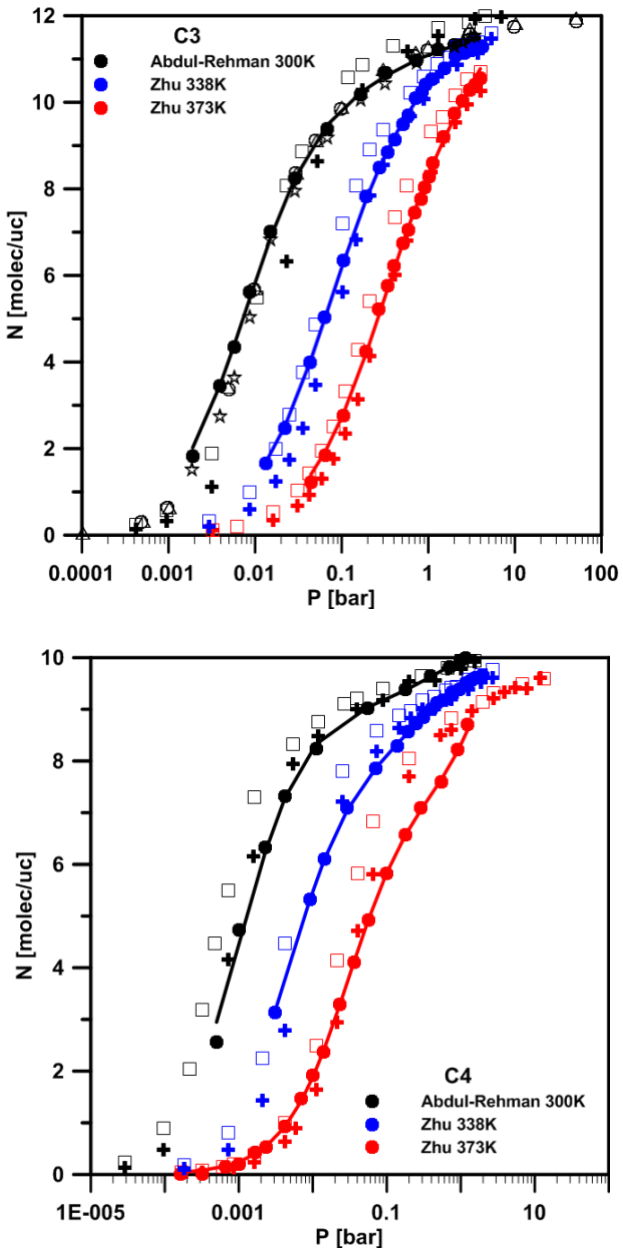

Figure S4. Adsorption isotherms for propane (top) and n-butane (bottom) in silicalite. The colors black, blue and red denote temperature of 300K, 338K and 373K, respectively. The closed circles are experimental data reported by Abdul-Rehman et al.[2] and Zhu et al.[3]. The open squares and crosses denote GCMC simulations of this work using combining rules SW and LB respectively. The open circles, triangles and stars are previous GCMC simulations reported by Du et al.[1], Krishna et al.[4] and Lu et al.[5], respectively. The continuous line represents the fit of experimental data using the Dual Site Langmuir (DSL) model.

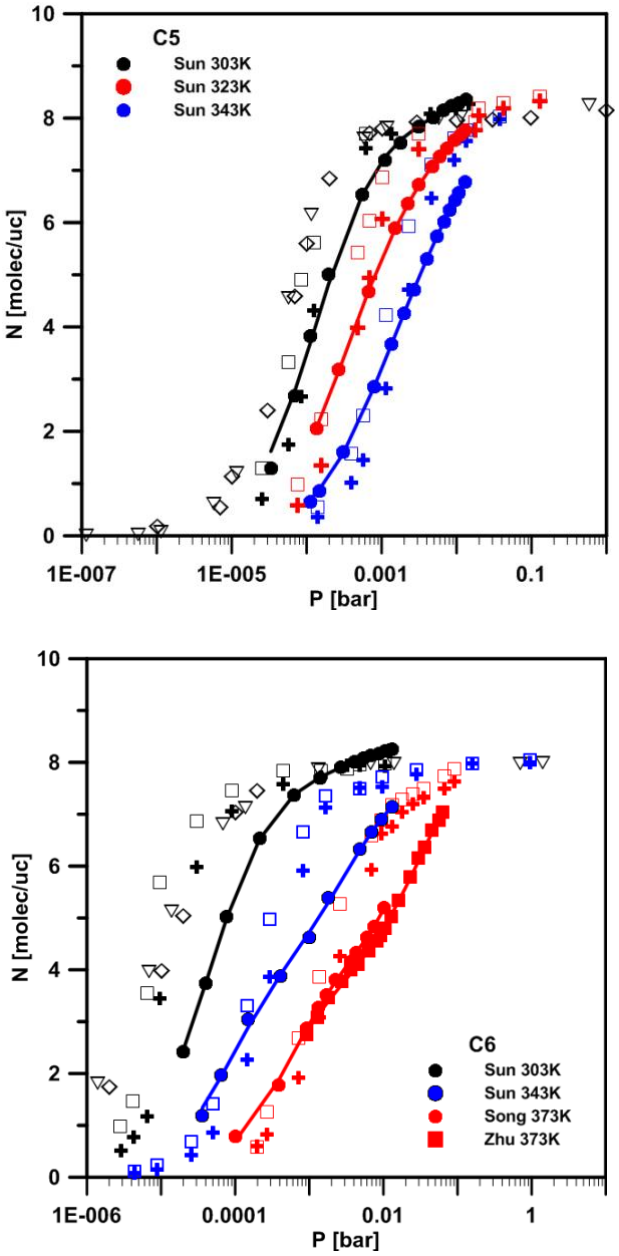

Figure S5. Adsorption isotherms for n-pentane (top) and n-hexane (bottom) in silicalite. The colors black, blue and red denote temperature of 300K, 343K and 373K, respectively. The closed circles and squares are experimental data reported by Zhu et al.[3], Sun et al.[9] and Song and Rees.[10] The open squares and crosses are GCMC simulations of this work using SW and LB respectively. The down triangles and diamonds are previous GCMC simulations reported by Vlugt et al.[11] and Pascual et al.[12], respectively. The continuous lines represent the fit of experimental data using the Dual Site Langmuir (DSL) model.

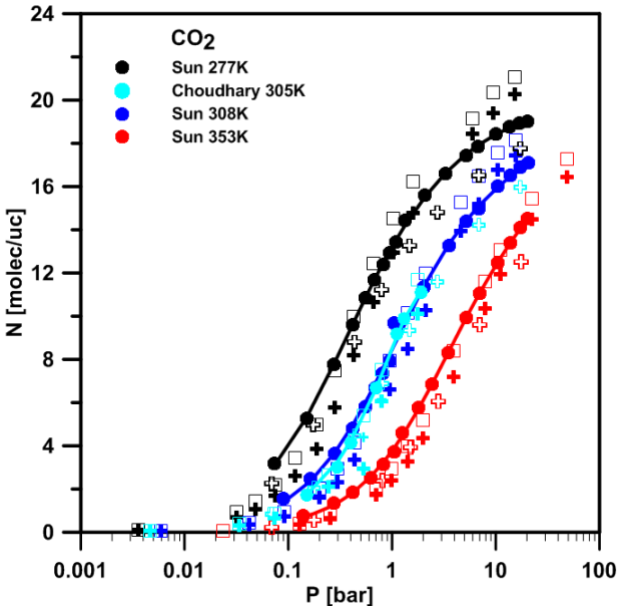

Figure S6. Adsorption isotherms for CO_2 in silicalite. The colors black, cyan, blue and red denote temperature of 277K, 305K, 308K and 353K, respectively. The closed circles are experimental data reported by Sun et al.[6] and Choudhary and Mayadevi[7]. The open squares and filled crosses are GCMC simulations of this work using combing rules SW and LB, respectively. The open crosses are previous GEMC simulations reported by Bai et al.[8] The continuous line represents the fit of experimental data using the Dual Site Langmuir (DSL) model.

Table S20- Summary of set up and results of GCMC simulations for methane adsorbed in MFI-type zeolite.

Set-up $-\mu_x 10^{-3}$ [J/mol]	**P [bar]**	**Adsorbed amount X [molecules/unit cell]**			
		LB		**SW**	
		<X>	**σ**	**<X>**	**σ**
			T=300K		
55.0	0.0007	0.0016	0.0001	0.0040	0.0003
50.0	0.0051	0.014	0.002	0.0319	0.0006
45.0	0.0378	0.107	0.006	0.233	0.001
43.0	0.0849	0.216	0.001	0.512	0.001
40.0	0.2821	0.75	0.05	1.614	0.004
38.0	0.6302	1.521	0.003	3.25	0.01
36.0	14.092	3.09	0.01	5.890	0.005
35.0	20.937	4.25	0.04	7.41	0.01
33.7	36.005	6.23	0.03	9.634	0.001
31.0	105.753	10.55	0.03	13.30	0.01
29.0	241.283	13.33	0.15	15.17	0.04
27.0	565.803	15.14	0.01	16.64	0.02
			T=338K		
55.0	0.0111	0.0140	0.0006	0.0280	0.0003
50.0	0.0658	0.0781	0.0009	0.166	0.002
47.0	0.1907	0.226	0.001	0.480	0.002
45.0	0.3921	0.457	0.003	0.950	0.001
44.0	0.5580	0.646	0.001	1.324	0.003
43.5	0.6629	0.769	0.002	1.564	0.009
43.0	0.7898	0.912	0.005	1.83	0.01

42.5	0.9422	1.075	0.007	2.168	0.007
42.0	1.1218	1.274	0.003	2.505	0.003
41.5	1.3599	1.506	0.004	2.923	0.007
41.0	1.6143	1.766	0.005	3.40	0.01
40.5	1.9434	2.073	0.005	3.90	0.01
40.0	2.3315	2.41	0.02	4.435	0.006
39.0	3.2867	3.29	0.02	5.69	0.02
38.0	4.7417	4.318	0.001	7.08	0.01
37.0	6.6955	5.558	0.006	8.48	0.03
			T=373K		
65.0	0.0035	0.0025	0.0002	0.0046	0.0001
63.0	0.0067	0.0042	0.0002	0.0087	0.0005
60.0	0.0176	0.0118	0.0005	0.0234	0.0008
55.0	0.0895	0.0603	0.0007	0.1189	0.0006
50.0	0.4496	0.291	0.008	0.570	0.004
49.0	0.6158	0.407	0.003	0.784	0.005
48.0	0.8503	0.558	0.006	1.065	0.007
47.0	1.1753	0.768	0.004	1.4333	0.0007
46.0	1.6101	1.032	0.005	1.95	0.02
45.0	2.2204	1.402	0.009	2.545	0.006
44.5	2.6223	1.626	0.007	2.919	0.004
44.0	3.0902	1.864	0.007	3.34	0.02
43.5	3.6440	2.15	0.02	3.784	0.007
43.0	4.2918	2.495	0.002	4.28	0.01
42.0	5.9319	3.24	0.01	5.371	0.002
41.0	8.3033	4.21	0.01	6.548	0.009

Table S21. Summary of set up and results of GCMC simulations performed for ethane adsorbed in MFI-type zeolite.

Set-up	**P**	**Adsorbed amount X[molecules/unit cell]**			
$-\mu_x 10^{-3}$	**[bar]**	**LB**		SW	
[J/mol]		**<X>**	**σ**	**<X>**	**σ**
			T=300K		
60	0.0002	0.0086	0.0001	0.0169	0.0003
55	0.0018	0.074	0.002	0.1210	0.0002
50	0.0132	0.523	0.003	0.903	0.007
48	0.0293	1.153	0.008	1.962	0.001
47	0.0437	1.75	0.02	2.86	0.03
45	0.0977	3.54	0.02	5.43	0.02
44	0.1459	5.02	0.02	6.992	0.009
43	0.2171	6.45	0.05	8.38	0.01
42	0.32311	7.97	0.02	9.75	0.05
40	0.7251	10.462	0.003	11.68	0.03
39	1.0941	11.447	0.006	12.54	0.01
38	1.6314	12.32	0.03	13.26	0.05
37	2.4408	13.05	0.07	13.88	0.05
36.1	3.4969	13.6	0.1	14.40	0.02
35	5.5924	14.31	0.09	14.91	0.02
34	8.3368	14.82	0.02	15.28	0.05
33	13.0348	15.20	0.01	15.50	0.06
			T=338K		

65.0	0.0008	0.0100	0.0002	0.0154	0.0003
60.0	0.0048	0.0589	0.0009	0.0943	0.0008
55.0	0.0284	0.349	0.002	0.550	0.005
53.0	0.0580	0.710	0.005	1.113	0.008
51.0	0.1185	1.41	0.01	2.170	0.008
50.0	0.1692	1.944	0.008	3.01	0.02
48.0	0.3449	3.62	0.04	5.233	0.007
47.0	0.4896	4.81	0.03	6.51	0.06
46.0	0.6994	6.10	0.01	7.86	0.02
45.0	1.0003	7.35	0.03	8.91	0.07
44.0	1.4337	8.61	0.03	9.97	0.02
43.0	2.0566	9.678	0.003	10.91	0.04
42.0	2.9470	10.631	0.007	11.69	0.06
41.5	3.5414	11.14	0.05	12.10	0.05
41.0	4.2212	11.46	0.05	12.29	0.01
40.0	6.0681	12.14	0.02	12.98	0.03
			T=373K		
65.0	0.0091	0.0464	0.0002	0.069	0.001
60.0	0.0457	0.229	0.002	0.349	0.003
57.0	0.1196	0.602	0.005	0.904	0.004
55.0	0.2292	1.111	0.008	1.663	0.008
54.0	0.3150	1.517	0.005	2.24	0.01
53.0	0.4376	2.04	0.01	2.984	0.008
52.0	0.6009	2.742	0.009	3.849	0.009
51.0	0.8285	3.57	0.01	4.88	0.01
50.0	1.1462	4.58	0.01	6.01	0.03
48.0	2.1933	6.90	0.04	8.28	0.03
47.0	3.0262	7.95	0.03	9.25	0.04
46.5	3.5894	8.39	0.07	9.58	0.03
46.0	4.2137	8.90	0.02	10.12	0.01
45.0	5.8702	9.88	0.03	10.90	0.03

Table S22- Summary of set up and results of GCMC simulations for propane adsorbed in MFI-type zeolite.

$-\mu_x 10^{-3}$ [J/mol]	**P [bar]**	**Adsorbed amount [molecules/unit cell]**			
		LB		**SW**	
		<X>	**σ**	**<X>**	**σ**
			T=300K		
60.0	0.0004	0.146	0.002	0.245	0.001
58.0	0.0009	0.327	0.003	0.56	0.01
55.0	0.0031	1.13	0.02	1.92	0.03
52.0	0.0105	3.80	0.06	5.46	0.02
50.0	0.0233	6.43	0.08	8.06	0.02
49.0	0.0350	7.62	0.07	8.86	0.02
48.0	0.0521	8.63	0.02	9.46	0.03
46.0	0.1172	9.81	0.06	10.57	0.02
45.0	0.1738	10.33	0.04	10.86	0.01
43.0	0.3884	10.95	0.01	11.305	0.008
42.0	0.5779	11.10	0.06	11.47	0.04
40.0	1.3060	11.47	0.05	11.73	0.03
38.0	2.9700	11.781	0.007	11.836	0.002

37.7	3.4392	11.85	0.05	11.97	0.02
37.0	4.5330	11.858	0.003	11.95	0.04
36.0	6.9971	11.97	0.04	11.854	0.008
			T=338K		
60	0.0086	0.600	0.001	0.983	0.007
58	0.0173	1.22	0.01	1.98	0.01
57	0.0247	1.73	0.02	2.71	0.05
56	0.0354	2.44	0.02	3.74	0.01
55	0.0505	3.453	0.005	4.95	0.06
53	0.1031	5.63	0.02	7.12	0.06
52	0.1470	6.86	0.02	7.96	0.09
51	0.2111	7.84	0.03	8.84	0.04
50	0.2999	8.60	0.04	9.34	0.03
48	0.6129	9.63	0.04	10.25	0.03
47	0.8827	10.06	0.04	10.52	0.06
46	1.2646	10.53	0.04	10.895	0.009
45	1.7970	10.73	0.02	11.14	0.07
44.5	2.1674	10.93	0.05	11.24	0.02
44	2.5820	11.03	0.02	11.27	0.04
43	3.7291	11.18	0.04	11.45	0.04
42	5.3532	11.40	0.05	11.57	0.03
			T=373K		
70.0	0.0032	0.073	0.002	0.106	0.002
65.0	0.0161	0.350	0.003	0.544	0.007
63.0	0.0308	0.69	0.01	1.027	0.005
62.0	0.0428	0.94	0.01	1.428	0.008
61.0	0.0583	1.29	0.01	1.97	0.02
60.0	0.0812	1.756	0.003	2.51	0.08
59.0	0.1107	2.36	0.01	3.33	0.07
58.0	0.1548	3.11	0.02	4.31	0.03
57.0	0.2129	4.04	0.07	5.26	0.05
56.0	0.2939	5.02	0.02	6.32	0.08
55.0	0.4084	6.10	0.07	7.28	0.05
54.0	0.5607	6.91	0.08	8.07	0.02
53.0	0.7711	7.85	0.07	8.69	0.05
52.0	1.0712	8.38	0.09	9.30	0.02
51.0	1.4852	9.11	0.03	9.59	0.05
50.0	2.0647	9.57	0.03	10.13	0.02
49.0	2.8602	9.92	0.03	10.48	0.04
48.0	3.9804	10.30	0.04	10.72	0.02

Table S23. Summary of set up and results of GCMC simulations for n-butane adsorbed in MFI-type zeolite.

-μx10^{-3} [J/mol]	**P [bar]**	**Adsorbed amount X[molecules/unit cell]**			
		LB		**SW**	
		<X>	**σ**	**<X>**	**σ**
			T=300K		
65.0	1E-5	0.064	0.002	0.112	0.003
63.0	3E-5	0.143	0.007	0.25	0.01
60.0	0.0001	0.48	0.02	0.85	0.03
58.0	0.0002	1.09	0.02	2.01	0.02
57.0	0.0003	1.19	0.60	3.05	0.02
56.0	0.0005	2.65	0.03	4.40	0.07
55.0	0.0007	3.98	0.02	5.40	0.08
53.0	0.0016	6.20	0.03	7.24	0.06
50.0	0.0053	7.95	0.01	8.24	0.07
48.0	0.0118	8.56	0.07	8.79	0.03
46.0	0.0264	8.713	0.005	9.08	0.02
45.0	0.0395	9.01	0.01	9.12	0.06
43.0	0.0882	9.19	0.02	9.40	0.02
41.0	0.1979	9.54	0.02	9.557	0.008
40.0	0.2952	9.53	0.01	9.641	0.007
39.0	0.4466	9.556	0.005	9.74	0.07
38.0	0.6655	9.60	0.08	9.79	0.02
37.0	1.0063	9.80	0.02	9.85	0.07
36.0	1.5197	9.85	0.05	10.09	0.07
			T=338K		
70.0	0.0001	0.0412	0.0007	0.0606	0.0004
67.0	0.0002	0.113	0.002	0.190	0.006
63.0	0.0007	0.474	0.007	0.79	0.01
60.0	0.0021	1.421	0.001	2.248	0.007
58.0	0.0042	2.793	0.007	4.29	0.01
55.0	0.0123	5.89	0.07	6.81	0.06
53.0	0.0249	7.13	0.06	7.86	0.04
50.0	0.0736	8.25	0.04	8.63	0.04
48.0	0.1481	8.64	0.01	8.90	0.02
47.0	0.2126	8.85	0.02	8.964	0.008
46.0	0.3038	8.89	0.08	9.13	0.03
45.0	0.4340	9.05	0.04	9.23	0.02
44.0	0.6214	9.39	0.04	9.33	0.03
43.5	0.7497	9.24	0.04	9.38	0.02
43.0	0.8893	9.23	0.08	9.434	0.004
42.0	1.2864	9.36	0.02	9.47	0.07
41.0	1.8550	9.51	0.03	9.62	0.03
40.0	2.6809	9.60	0.02	9.69	0.06
			T=373K		
73.0	0.0003	0.0497	0.0736	0.074	0.002
71.0	0.0006		0.1448	0.145	0.004
70.0	0.0008	0.1273	0.1961	0.196	0.001
68.0	0.0016	0.2386	0.3710	0.371	0.008
65.0	0.0042	0.6331	0.9732	0.95	0.02
64.0	0.0058	0.8845	1.3721	1.360	0.009
62.0	0.0111	1.6334	2.5051	2.50	0.02

60.0	0.0211	2.9419	4.1228	4.04	0.05
58.0	0.0407	4.7057	5.7538	5.79	0.08
56.5	0.0648	5.7976	6.7681	6.76	0.05
55.0	0.1062		7.5149	7.41	0.04
53.0	0.2020	7.6987	8.0502	8.09	0.06
50.0	0.5345	8.4899	8.6495	8.658	0.006
49.0	0.7380	8.6076	8.8291	8.84	0.02
47.0	1.4208	8.9628	9.1574	9.06	0.07
46.0	1.9617		9.1767	9.27	0.04
45.0	2.7390	9.2093	9.3736	9.35	0.06
44.0	3.8411	9.3289	9.3948	9.36	0.03
43.0	5.4736	9.4134	9.5232	9.53	0.03
42.5	6.8262		9.5263	9.52	0.02
42.0	7.9219	9.3918	9.4891	9.59	0.07
41.5	9.6878		9.6265	9.58	0.04
41.0	11.7408	9.6023	9.6423	9.68	0.03

Table S24. Summary of set up and results of GCMC simulations performed for n-pentane adsorbed in MFI-type zeolite. Simulation were performed with tail correction and without intramolecular scaling factor (0 0 0 1)

Set-up	**P**	**Adsorbed amount X [molecules/unit cell]**			
$-\mu_x 10^{-3}$	**[bar]**	**LB**		**LB**	
[J/mol]		**<X>**	**σ**	**<X>**	**σ**
			T=303K		
63.0	3E-05	0.70	0.02	1.29	0.05
61.0	6E-05	1.74	0.04	3.32	0.04
60.0	8E-05	2.66	0.06	4.90	0.07
59.0	0.0001	4.31	0.05	5.62	0.07
55.0	0.0006	7.43	0.04	7.69	0.02
53.0	0.0014	7.70	0.03	7.97	0.02
50.0	0.0044	8.08	0.02	8.08	0.02
48.0	0.0099	8.18	0.05	8.20	0.02
47.0	0.0147	8.279	0.003	8.29	0.02
			T=323K		
65.0	8E-05	0.59	0.01	0.98	0.01
63.0	0.0002	1.34	0.02	2.23	0.07
60.0	0.0005	3.98	0.07	5.43	0.06
59.0	0.0007	4.94	0.09	6.03	0.07
58.0	0.0010	6.06	0.03	6.87	0.02
55.0	0.0031	7.40	0.02	7.69	0.02
53.0	0.0176	7.76	0.02	7.93	0.05
50.0	0.0200	8.04	0.02	8.19	0.01
48.0	0.0421	8.18	0.05	8.29	0.02
45.0	0.1301	8.32	0.02	8.41	0.03
			T=343K		
68.0	0.0001	0.35	0.01	0.543	0.002
65.0	0.0004	1.02	0.02	1.58	0.09
64.0	0.0006	1.44	0.04	2.31	0.01
62.0	0.0011	2.81	0.06	4.22	0.08

60.0	0.0023	4.71	0.03	5.93	0.04
58.0	0.0046	6.47	0.06	7.11	0.00
56.0	0.0093	7.20	0.05	7.60	0.06
55.0	0.0133	7.56	0.05	7.77	0.04
52.0	0.0380	7.97	0.03	8.03	0.02

Table S25. Summary of set up and results of GCMC simulations performed for 2-methyl-pentane adsorbed in MFI-type zeolite. Simulation were performed with tail correction and with intramolecular scaling factor (0 0 0.5 1)

-μx10^{-3} [J/mol]	**Px10^{-5} [Pa]**	**Adsorbed amount X[molecules/unit cell] LB**		**SW**	
		<X>	**σ**	**<X>**	**σ**
		T=423K			
82.0	0.0007	0.092	0.001	0.207	0.005
80.0	0.0013	0.169	0.003	0.345	0.004
78.0	0.0023	0.285	0.007	0.62	0.007
75.0	0.0053	0.630	0.006	1.26	0.03
73.0	0.0094	1.023	0.006	1.86	0.01
70.0	0.0218	1.89	0.05	2.81	0.02
67.0	0.0513	2.83	0.01	3.57	0.03
65.0	0.0907	3.32	0.04	3.94	0.05

Table S26. Summary of set up and results of GCMC simulation performed for n-hexane adsorbed in MFI-type zeolite. Simulation were performed with tail correction and without intramolecular scaling factor (0 0 0 1)

-μx10^{-3} [J/mol]	**P [bar]**	**Adsorbed amount X[molecules/unit cell] LB**		**SW**	
		<X>	**σ**	**<X>**	**σ**
		T=303K			
60.0	3E-05	6.07	0.08	6.82	0.03
57.0	9E-05	7.15	0.06	7.43	0.02
53.0	0.0004	7.60	0.02	7.80	0.02
50.0	0.0015	7.88	0.05	7.88	0.03
48.0	0.0032	7.99	0.04	7.95	0.02
47	0.0048	7.99	0.04	7.96	0.03
45.0	0.0107	7.99	0.07	7.98	0.02
		T=343K			
68	5E-05	0.86	0.02	1.41	0.02
65	0.0001	2.268	0.007	3.307	0.009
63	0.0003	3.87	0.09	4.97	0.08
60	0.0008	5.90	0.05	6.652	0.009
57	0.0017	7.13	0.01	7.346	0.008
55	0.0048	7.51	0.03	7.48	0.08
53	0.0096	7.53	0.05	7.72	0.02
50	0.0276	7.76	0.03	7.85	0.06
45	0.1602	7.97	0.02	7.982	0.009
		T=373K			

67.0	0.0007	1.93	0.03	2.68	0.02
65.0	0.0014	3.09	0.02	3.86	0.01
63.0	0.0026	4.26	0.04	5.27	0.02
60.0	0.0068	5.92	0.08	6.59	0.09
59.0	0.0095	6.62	0.04	6.89	0.04
58.0	0.0132	6.76	0.03	7.18	0.03
57.0	0.0180	7.04	0.04	7.28	0.02
56.0	0.0250	7.20	0.04	7.38	0.06
55.0	0.0345	7.31	0.04	7.49	0.02
53.0	0.0659	7.50	0.01	7.73	0.04
52.0	0.0909	7.63	0.03	7.87	0.02

Table S27. Summary of set up and results of GCMC simulations for carbon dioxide adsorbed in MFI-type zeolite.

$-\mu x10^{-3}$ [J/mol]	**$Px10^{-5}$ [Pa]**	**Adsorbed amount X[molecules/unit cell] LB**		**SW**	
		<X>	**σ**	**<X>**	**σ**
			T=277K		
50.0	0.0036	0.082	0.002	0.1045	0.0007
45.0	0.0316	0.70	0.01	0.92	0.02
44.0	0.0486	1.08	0.01	1.429	0.001
43.0	0.0753	1.69	0.02	2.20	0.02
42.0	0.1158	2.607	0.007	3.42	0.02
41.0	0.1899	3.86	0.05	4.97	0.04
40.0	0.2780	5.76	0.02	7.47	0.04
39.0	0.4269	8.20	0.07	9.97	0.02
38.0	0.6600	10.63	0.09	12.45	0.07
37.0	1.0264	12.95	0.08	14.54	0.04
36.0	1.5942	14.78	0.04	16.24	0.03
33.0	5.9957	18.42	0.05	19.14	0.07
32.0	9.3628	19.38	0.02	20.34	0.07
31.0	15.4145	20.28	0.05	21.05	0.03
			T=305K		
55.0	0.0047	0.041	0.002	0.053	0.001
50.0	0.0338	0.297	0.003	0.379	0.002
48.0	0.0741	0.657	0.004	0.832	0.006
45.0	0.2424	2.11	0.01	2.66	0.02
43.0	0.5358	4.36	0.01	5.41	0.03
42.0	0.7954	6.06	0.05	7.51	0.01
40.0	1.7506	10.11	0.04	11.70	0.02
			T=308K		
55.0	0.0059	0.050	0.003	0.059	0.002
50.0	0.0417	0.342	0.002	0.428	0.003
48.0	0.0916	0.73	0.01	0.93	0.01
46.0	0.2003	1.600	0.005	2.03	0.01
45.0	0.2951	2.29	0.02	2.937	0.004
44.0	0.4377	3.35	0.02	4.16	0.06
42.0	0.9555	6.60	0.06	7.92	0.06
41.0	1.4169	8.50	0.02	10.2	0.1
40.0	2.0919	10.2	0.1	11.97	0.07

38.0	4.6052	13.93	0.06	15.25	0.01
37.0	6.8625	15.20	0.03	16.5	0.1
36.0	10.4562	16.8	0.1	17.6	0.1
35.0	15.6367	17.45	0.09	18.1	0.1
			T=353K		
50.0	0.0036	0.114	0.002	0.074	0.002
45.0	0.0316	0.331	0.002	0.407	0.002
44.0	0.0486	0.660	0.003	0.797	0.005
43.0	0.0753	1.73	0.01	2.13	0.01
42.0	0.1158	2.39	0.02	2.934	0.002
40.0	0.2780	3.29	0.03	3.96	0.03
38.0	0.6600	4.37	0.02	5.19	0.04
37.0	1.0264	7.17	0.02	8.38	0.01
36.0	1.5942	10.365	0.007	11.6	0.1
33.0	5.9957	11.91	0.02	13.06	0.04
32.0	9.3628	14.5	0.1	15.45	0.01
31.0	15.4145	16.4	0.1	17.2	0.1

Table S28 Summary of set up and results of GCMC simulation for methanol adsorbed in MFI-type zeolite.

$-\mu x10^{-3}$ [J/mol]	**$Px10^{-5}$ [Pa]**	**Adsorbed amount X[molecules/unit cell] LB**		**SW**	
		<X>	**σ**	**<X>**	**σ**
			T=303K		
65.0	5E-05	0.016	0.001	0.022	0.003
60.0	0.0003	0.132	0.002	0.173	0.006
58.0	0.0008	0.28	0.01	0.41	0.02
55.0	0.0025	1.10	0.04	1.62	0.02
54.0	0.0037	1.97	0.06	3.04	0.04
53.0	0.0056	3.32	0.05	4.96	0.04
52.0	0.0082	5.77	0.04	8.16	0.03
50.0	0.0182	11.89	0.09	14.16	0.04
48.0	0.0407	15.73	0.07	16.70	0.05
47.0	0.0600	16.76	0.02	18.149	0.006
45.0	0.1437	18.38	0.07	19.11	0.03

2.1.2- Adsorption of mixtures

Binary

The uncertainties were evaluated by the standard deviation of three independent set of GCMC simulations. The results of the 3 simulations for adsorption of methane(1)+n-butane(2) using the proposed combining rule (June+SW) are presented in Table S34. The load of butane on equilibration runs for the adsorption of the mixture with 70.15% of methane is also presented in Figure S7. The average deviation on the number of molecules adsorbed per unit cell was around 5% to methane and

0.4% for butane. The larger deviation on methane is due to the low number of adsorbed molecules. Also, in the Figure S7 it can be observed that even using different seeds and start types all three simulations equilibrate at around the same value.

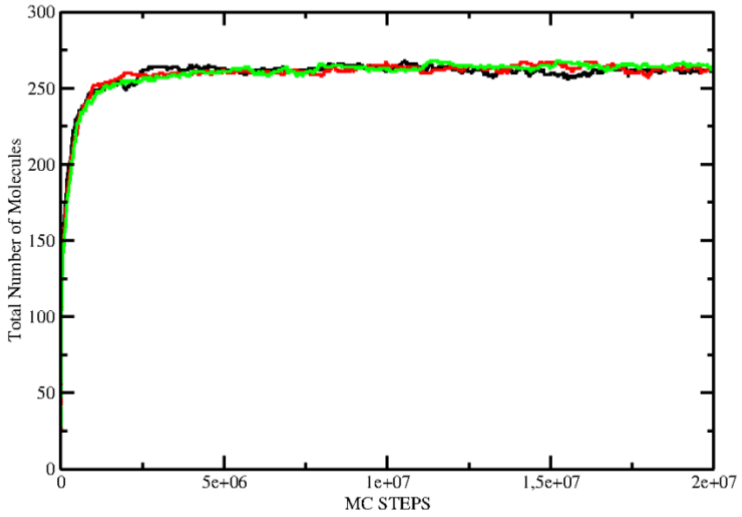

Figure S7. Total number of butane molecules adsorbed in MFI (27uc) at 300K,. Result for adsorption. The colors black, red and green indicate independent simulations (equilibration step) performed for μ_{CH4}=-34.50kJ/mol; μ_{C4H10}=-38.0kJ/mol using June+SW.

Table S29. Results of three independent runs of GCMC simulations performed for adsorption of methane (1)+n-butane(2) in MFI at 300K. The simulations used force field of June and combining rule SW.

-μx10^{-3} [J/mol]		**P_x10^{-5}**	**y_1**	**Molecules/uc**							
				RUN1		**RUN2**		**RUN3**		**Average**	
C1	**C4**	**[Pa]**		**C1**	**C4**	**C1**	**C4**	**C1**	**C4**	**C1**	**C4**
35.41	37.00	3.3852	0.5185	0.1198	9.9230	0.0995	9.8883	0.1128	9.8303	0.1107	9.8805
35.00	37.50	3.3848	0.6115	0.1393	9.7448	0.1350	9.8479	0.1714	9.7314	0.1486	9.7747
34.50	38.00	3.5847	0.7015	0.1902	9.7644	0.1875	9.7945	0.1841	9.7540	0.1873	9.7710
34.50	38.60	3.3490	0.7524	0.2353	9.6549	0.2539	9.6358	0.2196	9.6701	0.2363	9.6536
34.10	40.50	3.3690	0.8852	0.4026	9.4837	0.3957	9.3986	0.4455	9.4598	0.4146	9.4474
33.80	43.00	3.5739	0.9604	0.7581	8.9674	0.7269	9.0086	0.7290	9.0224	0.7380	8.9995

The Tables S30, S31, S32, S33 and S34 present the average values and standard deviation (σ) obtained by the GCMC simulations performed for the adsorption of following binary mixtures in silicalite: methane(1)+ethane(2), methane(1)+propane(2), methane(1)+butane(2) and

ethane(1)+propane(2) at 300K and 2-methyl-pentane(1)+n-hexane(2) at 473K, respectively. Each Table presents the results obtained by different set of force field/combining rule. The simulation results obtained by the use of June et al. force field to zeolite with Lorentz-Berthelot combining rule were named LB. while the name SW is related to results using the same force field to zeolite but with the proposed combining rule. The name TraZeo was used to describe the use of the force field proposed by Bai et al. named TraPPE-Zeo what use the LB combining rule. Finally the name Du. reports the results obtained using the tuned LJ parameters for the cross interaction proposed by Du et al. The uncertainties were evaluated by the standard deviation of three independent set of GCMC simulations. The results of the 3 simulations for adsorption of methane(1)+n-butane(2) using the proposed combining rule (June+SW) are presented in Table S34. The load of butane on equilibration runs for the adsorption of the mixture with 70.15% of methane is also presented in Figure S7. The average deviation on the number of molecules adsorbed per unit cell was around 5% to methane and 0.4% for butane. The larger deviation on methane is due to the low number of adsorbed molecules. Also, in the Figure S7 it can be observed that even using different seeds and start types all three simulations equilibrate at around the same value.

Table S30. Summary of set up and results of GCMC simulation for adsorption of binary mixtures of methane(1)+ethane(2) in MFI-type zeolite at 300K.

Set up				**<Average values> adsorbed amount**			
$-\mu x10^{-3}$ [J/mol]		**$Px10^{-5}$ [Pa]**	**y_1**	**molecules/uc**			
C1	**C2**			**C1**	**σ**	**C2**	**σ**
June+LB							
36.72	37.00	3.518	0.3003	0.358	0.002	12.784	0.008
35.96	37.39	3.523	0.4026	0.538	0.003	12.42	0.01
35.40	37.84	3.542	0.5014	0.758	0.007	11.87	0.02
35.26	38.00	3.530	0.6140	1.13	0.02	11.07	0.01
34.56	39.11	3.556	0.6997	1.49	0.05	10.28	0.03
34.26	40.00	3.578	0.7948	2.08	0.01	9.09	0.05
34.00	41.00	3.640	0.8641	2.824	0.007	7.597	0.003
33.92	42.00	3.584	0.9069	3.56	0.02	6.04	0.01
June+SW							
39.41	36.38	3.531	0.1011	0.168	0.005	13.95	0.01
37.68	36.67	3.535	0.2041	0.384	0.006	13.70	0.01
36.72	37.00	3.518	0.3003	0.58	0.03	13.52	0.02
35.96	37.39	3.522	0.4026	0.87	0.01	12.97	0.04

35.40	37.84	3.542	0.5014	1.13	0.02	12.42	0.04
34.90	38.45	3.530	0.6140	1.72	0.007	11.64	0.04
34.56	39.11	3.556	0.7948	2.33	0.007	10.76	0.02
34.26	40.00	3.578	0.8640	3.21	0.05	9.46	0.09
34.00	41.00	3.640	0.9069	4.32	0.02	7.87	0.02
33.92	42.00	3.584	0.9726	5.308	0.005	6.39	0.01
33.68	50.00	3.571	0.9961	9.373	0.003	0.414	0.002
TraZeo+LB							
36.72	37.00	3.5178	0.3003	0.341	0.004	12.26	0.05
35.96	37.39	3.5223	0.4026	0.52	0.02	11.96	0.01
35.40	37.84	3.5424	0.5014	0.75	0.01	11.62	0.06
35.26	38.00	3.5295	0.614	1.02	0.02	11.08	0.03
34.56	39.11	3.5559	0.6997	1.41	0.02	10.59	0.04
34.26	40.00	3.5778	0.7948	2.55	0.07	8.63	0.05
34.00	41.00	3.6397	0.8641	2.840	0.004	8.42	0.02
33.92	42.00	3.5842	0.9069	3.60	0.03	7.25	0.05
Du							
39.41	36.38	3.5314	0.1011	0.085	0.002	11.95	0.01
37.68	36.67	3.5351	0.2041	0.169	0.002	11.879	0.006
36.72	37.00	3.5178	0.3003	0.275	0.001	11.70	0.03
35.96	37.39	3.5223	0.4026	0.43	0.01	11.44	0.03
35.40	37.84	3.5424	0.5014	0.61	0.01	11.19	0.02
34.90	38.45	3.5295	0.6140	0.94	0.02	10.76	0.04
34.56	39.11	3.5559	0.7948	1.784	0.004	9.596	0.001
34.26	40.00	3.5778	0.864	2.681	0.006	8.387	0.006
34.00	41.00	3.6397	0.9069	4.47	0.02	6.52	0.01
33.92	42.00	3.5842	0.9726	6.00	0.03	3.526	0.009
33.68	50.00	3.5712	0.9961	7.89	0.01	0.577	0.005

Table S31. Summary of set up and results of GCMC simulation for adsorption of binary mixtures of methane(1)+propane(2) in MFI-type zeolite at 300K.

Set up				**<Average values> adsorbed amount**			
-μx10^{-3} [J/mol]		**Px10^{-5} [Pa]**	**y_1**	**molecules/uc**			
C1	**C3**			**C1**	**σ**	**C3**	**σ**
June+LB							
39.41	38.00	3.3709	0.1059	0.02	11.80	0.001	0.01
36.90	38.50	3.4280	0.2809	0.06	11.74	0.002	0.02
36.00	38.75	3.5652	0.3861	0.09	11.68	0.002	0.01
35.75	39.00	3.5508	0.4342				
35.20	39.50	3.5747	0.5430	0.17	11.54	0.01	0.02
34.86	40.00	3.5327	0.6237	0.20	11.39	0.01	0.02
34.39	41.00	3.5543	0.7490	0.32	11.18	0.01	0.01
34.14	42.00	3.5486	0.8308	0.47	10.86	0.01	0.02
33.97	43.00	3.5695	0.8889	0.70	10.43	0.006	0.04
33.80	45.00	3.5645	0.9490	1.37	9.33	0.008	0.01
33.69	50.00	3.6152	0.9936	4.17	4.30	0.039	0.01
June+SW							

39.41	38.00	3.3709	0.1059	0.03	0.01	11.96	0.04
36.90	38.50	3.4280	0.2809	0.09	0.003	11.89	0.01
36.00	38.75	3.5652	0.3861	0.14	0.01	11.84	0.05
35.75	39.00	3.5508	0.4342	0.14	0.002	11.87	0.02
35.20	39.50	3.5747	0.5430	0.24	0.01	11.72	0.02
34.86	40.00	3.5327	0.6237	0.30	0.01	11.64	0.02
34.39	41.00	3.5543	0.7490	0.45	0.02	11.45	0.04
34.14	42.00	3.5486	0.8308	0.64	0.01	11.18	0.003
33.97	43.00	3.5695	0.8889				
33.80	45.00	3.5645	0.9490	1.66	0.07	9.90	0.06
TraZeo+LB							
39.41	38.00	3.3709	0.1059	0.01	0.02	11.76	0.06
36.90	38.50	3.4280	0.2809	0.082	0.007	11.50	0.02
36.00	38.75	3.5652	0.3861	0.07	0.01	11.75	0.03
35.75	39.00	3.5508	0.4342	0.08	0.02	11.64	0.01
35.20	39.50	3.5747	0.5430	0.098	0.008	11.52	0.01
34.86	40.00	3.5327	0.6237	0.195	0.007	11.36	0.02
34.39	41.00	3.5543	0.7490	0.30	0.02	11.26	0.01
34.14	42.00	3.5486	0.8308	0.42	0.04	10.97	0.02
33.97	43.00	3.5695	0.8889	0.611	0.005	10.63	0.01
33.80	45.00	3.5645	0.9490	1.26	0.07	9.72	0.03
33.69	50.00	3.6152	0.9936	4.31	0.01	5.55	0.02
Du							
39.41	36.38	3.5314	0.1011	0.022	0.002	11.48	0.02
37.68	36.67	3.5351	0.2041	0.078	0.001	11.33	0.02
36.72	37.00	3.5178	0.3003	0.14	0.02	11.19	0.07
35.96	37.39	3.5223	0.4026	0.15	0.04	11.2	0.1
35.40	37.84	3.5424	0.5014	0.20	0.05	11.1	0.1
34.90	38.45	3.5295	0.6140	0.23	0.02	11.09	0.06
34.56	39.11	3.5559	0.7948	0.37	0.03	10.84	0.05
34.26	40.00	3.5778	0.864	0.59	0.02	10.501	0.008
34.00	41.00	3.6397	0.9069	0.76	0.08	10.28	0.09
33.92	42.00	3.5842	0.9726	1.49	0.01	9.32	0.04
33.68	50.00	3.5712	0.9961	4.53	0.02	5.28	0.01

Table S32. Summary of set up and results of GCMC simulation for adsorption of binary mixtures of methane(1)+butane(2) in MFI-type zeolite at 300K.

Set up				**<Average values> adsorbed amount**			
-μx10^{-3} [J/mol]		**Px10^{-5} [Pa]**	**y_1**	**molecules/uc**			
C1	**C4**			**C1**	**σ**	**C4**	**σ**
June+LB							
36.00	36.50	3.3677	0.4108	0.04	0.01	10.01	0.01
35.41	37.00	3.3852	0.5185	0.06	0.02	9.83	0.02
35.00	37.50	3.3848	0.6115	0.103	0.005	9.73	0.01
34.50	38.00	3.5847	0.7015	0.12	0.02	9.68	0.07
34.50	37.95	3.6778	0.7018	0.10	0.04	9.76	0.04
34.50	38.60	3.3490	0.7524	0.11	0.01	9.66	0.05
34.10	40.50	3.3690	0.8852	0.20	0.05	9.42	0.03

33.80	43.00	3.5739	0.9604	0.37	0.03	9.08	0.02
33.70	46.00	3.6160	0.9885	0.73	0.02	8.504	0.005
33.67	48.00	3.5944	0.9947	1.29	0.01	7.73	0.01
June+SW							
36.00	36.50	3.3677	0.4108	0.101	0.008	9.86	0.03
35.41	37.00	3.3852	0.5185	0.111	0.02	9.88	0.04
35.00	37.50	3.3848	0.6115	0.15	0.002	9.77	0.05
34.50	38.00	3.5847	0.7015	0.187	0.01	9.77	0.02
34.50	38.60	3.3490	0.7524	0.24	0.02	9.65	0.01
34.10	40.50	3.3690	0.8852	0.41	0.01	9.45	0.04
33.80	43.00	3.5739	0.9604	0.74	0.008	9.00	0.02
33.70	46.00	3.6160	0.9885	1.49	0.02	8.199	0.04
33.67	48.00	3.5944	0.9947	2.320	0.002	7.426	0.007
TraZeo+LB							
36.00	36.50	3.3677	0.4108	0.044	0.003	9.58	0.01
35.41	37.00	3.3852	0.5185	0.07	0.01	9.35	0.01
35.00	37.50	3.3848	0.6115	0.11	0.01	9.45	0.02
34.50	38.00	3.5847	0.7015	0.10	0.02	9.51	0.01
34.50	37.95	3.6778	0.7018	0.15	0.05	9.45	0.03
34.50	38.60	3.3490	0.7524	0.11	0.02	9.39	0.02
34.10	40.50	3.3690	0.8852	0.23	0.03	9.20	0.04
33.80	43.00	3.5739	0.9604	0.37	0.04	9.03	0.02
33.70	46.00	3.6160	0.9885	0.70	0.01	8.48	0.01
33.67	48.00	3.5944	0.9947	1.11	0.06	8.12	0.02
Du							
36.00	36.50	3.3677	0.4108	0.050	0.004	9.380	0.005
35.41	37.00	3.3852	0.5185	0.072	0.006	9.407	0.001
35.00	37.50	3.3848	0.6115	0.07	0.02	9.426	0.005
34.50	38.00	3.5847	0.7015	0.110	0.004	9.36	0.07
34.50	37.95	3.6778	0.7018				
34.50	38.60	3.3490	0.7524	0.169	0.02	9.15	0.06
34.10	40.50	3.3690	0.8852	0.193	0.02	9.12	0.06
33.80	43.00	3.5739	0.9604	0.384	0.04	8.87	0.07
33.70	46.00	3.6160	0.9885	0.664	0.02	8.55	0.01
33.67	48.00	3.5944	0.9947	1.259	0.05	7.86	0.08

Table S33. Summary of set up and results of GCMC simulation for adsorption of binary mixtures of ethane(1)+propane(2) in MFI-type zeolite at 300K.

Set up				**<Average values> adsorbed amount molecules/uc**			
-μx10^{-3} [J/mol]		**Px10^{-5} [Pa]**	**y_1**				
C2	**C3**			**C2**	**σ**	**C3**	**σ**
June+LB							
41.84	37.931	3.4638	0.1019	0.19	0.03	11.67	0.01
40.11	38.21	3.4746	0.2028				
39.1	38.54	3.4767	0.3039	0.88	0.05	11.06	0.02
38.39	38.92	3.4842	0.4051	1.39	0.02	10.59	0.02
37.83	39.38	3.4958	0.5055	1.89	0.01	10.1	0.1
37.37	39.94	3.4873	0.6057	2.6	0.1	9.42	0.06
36.87	40.96	3.5063	0.7056	4.02	0.08	8.21	0.09

36.66	41.66	3.4982	0.8045	5.08	0.05	7.34	0.03
June+SW							
41.84	37.931	3.4638	0.1019	0.237	0.002	11.76	0.02
40.11	38.21	3.4746	0.2028				
39.1	38.54	3.4767	0.3039	0.95	0.04	11.11	0.07
38.39	38.92	3.4842	0.4051	1.46	0.09	10.71	0.07
37.83	39.38	3.4958	0.5055	1.91	0.07	10.35	0.05
37.37	39.94	3.4873	0.6057	2.991	0.003	9.418	0.004
36.87	40.96	3.5063	0.7056	4.19	0.03	8.30	0.05
36.66	41.66	3.4982	0.8045	5.77	0.04	7.123	0.001
TraZeo+LB							
41.84	37.931	3.4638	0.1019	0.32	0.01	11.43	0.01
40.11	38.21	3.4746	0.2028				
39.1	38.54	3.4767	0.3039	0.8	0.03	10.86	0.09
38.39	38.92	3.4842	0.4051	1.2	0.1	10.64	0.09
37.83	39.38	3.4958	0.5055	1.68	0.04	10.12	0.02
37.37	39.94	3.4873	0.6057	2.42	0.08	9.4	0.1
36.87	40.96	3.5063	0.7056	3.45	0.08	8.4	0.1
36.66	41.66	3.4982	0.8045	4.53	0.01	7.39	0.01
Du							
41.84	37.931	3.4638	0.1019	0.60	0.09	10.8	0.1
40.11	38.21	3.4746	0.2028	0.9	0.10	10.64	0.08
39.1	38.54	3.4767	0.3039	0.78	0.09	10.84	0.05
38.39	38.92	3.4842	0.4051	1.39	0.05	10.21	0.06
37.83	39.38	3.4958	0.5055	1.72	0.02	9.864	0.007
37.37	39.94	3.4873	0.6057	2.03	0.09	9.70	0.08
36.87	40.96	3.5063	0.7056	3.4	0.10	8.4	0.1
36.66	41.66	3.4982	0.8045	3.8	0.20	7.79	0.06
36.36	43.39	3.5434	0.9031	4.9	0.10	7.0	0.2

Table S34. Summary of set up and results of GCMC simulation for adsorption of binary mixtures of 2-methyl-pentane(1)+n-hexane(2) in MFI-type zeolite at 473K.

Set up				**<Average values> adsorbed amount**			
$-\mu x10^{-3}$ [J/mol]		**$Px10^{-5}$ [Pa]**	**y_1**	**molecules/uc**			
2mC5	**nC6**			**2mC5**	**σ**	**nC6**	**σ**
June+LB							
76.29	64.48	0.0674	0.1000	0.11	0.01	4.23	0.05
72.33	65.38	0.0668	0.2972	0.44	0.01	3.53	0.02
70.50	66.60	0.0662	0.4985	0.90	0.01	2.68	0.04
69.28	68.43	0.0665	0.6931	1.43	0.05	1.83	0.06
68.38	72.39	0.0660	0.8979	2.26	0.02	0.61	0.01
June+SW							
76.29	64.48	0.0674	0.1000	0.19	0.01	4.80	0.07
72.33	65.38	0.0668	0.2972	0.688	0.004	3.94	0.09
70.50	66.60	0.0662	0.4985	1.28	0.04	3.07	0.08
69.28	68.43	0.0665	0.6931	2.08	0.07	1.82	0.04
68.38	72.39	0.0660	0.8979	2.98	0.01	0.547	0.003

Ternary

The results for ternary mixtures of methane(1)+ethane(2)+propane(3), methane(1)+ethane(2) + n-butane(3), methane(1)+ethane(2)+n-butane(3) are presented in Tables S35, S36 and S37, respectively. The uncertainties were estimated by dividing the production runs into three blocks.

Table S35. Summary of set up and results of GCMC simulation for adsorption of ternary mixtures of methane(1)+ethane(2)+propane(3) in MFI-type zeolite at 300K.

Set-up						**Adsorbed amount [molecule/uc]**		
-μx10^{-3} [J/mol]			**Px10^{-5}**					
C1	**C2**	**C3**	**[Pa]**	**y_1**	**y_2**	**C1**	**C2**	**C3**
June+LB								
36.67	37.76	41.85	3.55	0.3033	0.5156	0.15$_7$	3.76$_7$	8.$_1$
35.96	38.14	42.23	3.53	0.4037	0.4418	0.24$_1$	3.79$_2$	7.9$_9$
35.40	38.60	42.69	3.55	0.5058	0.3655	0.328$_9$	3.53$_5$	8.0$_5$
34.94	39.15	43.25	3.55	0.6045	0.2922	0.452$_7$	3.34$_6$	7.93$_4$
34.56	39.87	43.96	3.55	0.7044	0.2186	0.63$_8$	3.1$_9$	7.82$_3$
34.23	40.88	44.97	3.57	0.8035	0.1457	0.944$_5$	2.8$_6$	7.49$_7$
June+SW								
39.41	37.13	41.22	3.52	0.1021	0.6637	0.08$_3$	4.3$_1$	8.2$_5$
37.68	37.43	41.52	3.51	0.2033	0.5895	0.17$_4$	4.32$_5$	8.0$_6$
36.67	37.76	41.85	3.55	0.3033	0.5156	0.28$_7$	4.24$_2$	7.9$_9$
35.96	38.14	42.23	3.53	0.4037	0.4418	0.41$_3$	4.05$_6$	8.0$_1$
35.40	38.60	42.69	3.55	0.5058	0.3655	0.547$_9$	3.83$_2$	7.99$_3$
34.94	39.15	43.25	3.55	0.6045	0.2922	0.78$_5$	3.701$_2$	7.79$_6$
34.56	39.87	43.96	3.55	0.7044	0.2186	1.08$_3$	3.34$_3$	7.74$_3$
34.23	40.88	44.97	3.57	0.8035	0.1457	1.59$_2$	3.09$_3$	7.3$_2$
33.93	42.61	46.70	3.59	0.9026	0.0723	2.69$_3$	2.48$_2$	6.53$_4$
TraZeo+LB								
39.41	37.13	41.22	3.52	0.1021	0.6637	0.03$_2$	3.27$_5$	8.5$_1$
37.68	37.43	41.52	3.51	0.2033	0.5895	0.07$_2$	3.4$_3$	8.3$_2$
36.67	37.76	41.85	3.55	0.3033	0.5156	0.11$_1$	3.37$_2$	8.29$_5$
35.96	38.14	42.23	3.53	0.4037	0.4418	0.168$_5$	3.28$_1$	8.30$_4$
35.40	38.60	42.69	3.55	0.5058	0.3655	0.246$_4$	3.31$_5$	8.16$_1$
34.94	39.15	43.25	3.55	0.6045	0.2922	0.37$_3$	3.25$_3$	8.03$_3$
34.56	39.87	43.96	3.55	0.7044	0.2186	0.53$_1$	3.10$_1$	7.91$_2$
34.23	40.88	44.97	3.57	0.8035	0.1457	0.82$_5$	2.86$_2$	7.7$_2$
33.93	42.61	46.70	3.59	0.9026	0.0723	1.52$_4$	2.40$_3$	7.15$_1$
Du								
39.41	37.13	41.22	3.52	0.1021	0.6637	0.15$_3$	4.51$_2$	8.3$_4$
37.68	37.43	41.52	3.51	0.2033	0.5895	0.16$_2$	4.17$_4$	7.89$_2$
36.67	37.76	41.85	3.55	0.3033	0.5156	0.104$_5$	3.95$_7$	7.63$_1$
35.96	38.14	42.23	3.53	0.4037	0.4418	0.053$_2$	3.82$_3$	7.49$_6$
35.40	38.60	42.69	3.55	0.5058	0.3655	0.08$_4$	3.70$_3$	7.44$_3$
34.94	39.15	43.25	3.55	0.6045	0.2922	0.27$_1$	3.56$_2$	7.4$_1$
34.56	39.87	43.96	3.55	0.7044	0.2186	0.56$_2$	3.3$_4$	7.33$_5$
34.23	40.88	44.97	3.57	0.8035	0.1457	0.89$_1$	2.97$_4$	7.18$_3$
33.93	42.61	46.70	3.59	0.9026	0.0723	4.46$_3$	2.42$_6$	6.9$_1$

Table S36. Summary of set up and results of GCMC simulation for adsorption of ternary mixtures of methane(1)+ethane(2)+butane(3) in MFI-type zeolite at 300K.

Set-up						**Adsorbed amount [molecule/uc]**		
-μx10^{-3} [J/mol]			**Px10^{-5}**					
C1	**C2**	**C4**	**[Pa]**	**y_1**	**y_2**	**C1**	**C2**	**C4**
June+LB								
36.28	37.78	39.98	3.39	0.3707	0.5350	0.074$_2$	1.46$_3$	8.63$_2$
35.07	38.83	41.54	3.44	0.5859	0.3518	0.151$_5$	1.33$_2$	8.6$_1$
34.56	39.68	41.88	3.48	0.7173	0.2395	0.217$_5$	1.05$_1$	8.64$_3$
34.27	40.54	42.67	3.53	0.8002	0.1683	0.31$_2$	1.02$_3$	8.49$_1$
34.10	41.31	43.30	3.55	0.8524	0.1233	0.39$_5$	0.86$_5$	8.48$_4$
June+SW								
36.28	37.78	39.98	3.39	0.3707	0.5350	0.14$_1$	1.77$_5$	8.51$_9$
35.07	38.83	41.54	3.44	0.5859	0.3518	0.28$_2$	1.49$_4$	8.52$_1$
34.56	39.68	41.88	3.48	0.7173	0.2395	0.406$_5$	1.38$_1$	8.5$_1$
34.27	40.54	42.67	3.53	0.8002	0.1683	0.58$_3$	1.17$_2$	8.4$_3$
34.10	41.31	43.30	3.55	0.8524	0.1233	0.69$_1$	0.95$_7$	8.40$_2$
TraZeo+LB								
36.28	37.78	39.98	3.39	0.3707	0.5350	0.06$_3$	1.22$_5$	8.56$_5$
35.07	38.83	41.54	3.44	0.5859	0.3518	0.13$_2$	1.17$_9$	8.5$_3$
34.56	39.68	41.88	3.48	0.7173	0.2395	0.19$_5$	0.89$_6$	8.58$_1$
34.27	40.54	42.67	3.53	0.8002	0.1683	0.273$_9$	0.90$_8$	8.47$_2$
34.10	41.31	43.30	3.55	0.8524	0.1233	0.304$_4$	0.67$_5$	8.59$_6$
Du								
36.28	37.78	39.98	3.39	0.3707	0.5350	0.094$_8$	1.64$_2$	8.2$_3$
35.07	38.83	41.54	3.44	0.5859	0.3518	0.12$_1$	1.1$_1$	8.45$_2$
34.56	39.68	41.88	3.48	0.7173	0.2395	0.21$_3$	1.09$_3$	8.40$_3$
34.27	40.54	42.67	3.53	0.8002	0.1683	0.35$_5$	1.06$_2$	8.19$_6$
34.10	41.31	43.30	3.55	0.8524	0.1233	0.296$_1$	0.76$_5$	8.49$_3$

Table S37. Summary of set up and results of GCMC simulation for adsorption of ternary mixtures of methane(1)+propane(2)+butane(3) in MFI-type zeolite at 300K.

Set-up						Adsorbed amount [molecule/uc]		
-μx10^{-3} [J/mol]			Px10^{-5}					
C1	C3	C4	[Pa]	y_1	y_2	C1	C3	C4
June+LB								
36.44	40.16	37.86	3.56	0.3241	0.3516	0.044$_2$	3.19$_2$	7.26$_5$
35.17	41.11	38.85	3.57	0.5431	0.2410	0.09$_5$	2.44$_3$	7.72$_4$
34.62	41.95	39.87	3.53	0.6834	0.1723	0.126$_7$	2.51$_6$	7.62$_3$
34.29	42.83	40.79	3.56	0.7805	0.1217	0.19$_2$	2.3$_1$	7.6$_3$
34.10	43.62	41.62	3.58	0.8437	0.0871	0.25$_4$	2.39$_5$	7.42$_2$
June+SW								
36.44	40.16	37.86	3.56	0.3241	0.3516	0.07$_2$	3.04$_2$	7.53$_2$
35.17	41.11	38.85	3.57	0.5431	0.2410	0.15$_5$	2.72$_5$	7.66$_3$
34.62	41.95	39.87	3.53	0.6834	0.1723	0.23$_4$	2.57$_3$	7.66$_1$
34.29	42.83	40.79	3.56	0.7805	0.1217	0.30$_1$	2.6$_1$	7.55$_5$
34.10	43.62	41.62	3.58	0.8437	0.0871	0.42$_2$	2.5$_1$	7.44$_1$
TraZeo+LB								
36.44	40.16	37.86	3.56	0.3241	0.3516	0.042$_1$	2.88$_4$	7.2$_1$
35.17	41.11	38.85	3.57	0.5431	0.2410	0.071$_4$	2.34$_1$	7.58$_3$
34.62	41.95	39.87	3.53	0.6834	0.1723	0.10$_1$	2.23$_2$	7.61$_4$
34.29	42.83	40.79	3.56	0.7805	0.1217	0.17$_3$	2.02$_5$	7.7$_1$
34.10	43.62	41.62	3.58	0.8437	0.0871	0.19$_4$	1.86$_1$	7.8$_1$
Du								
36.44	40.16	37.86	3.56	0.3241	0.3516	0.10$_1$	3.49$_1$	6.60$_2$
35.17	41.11	38.85	3.57	0.5431	0.2410	0.10$_2$	3.61$_3$	6.5$_1$
34.62	41.95	39.87	3.53	0.6834	0.1723	0.15$_5$	3.43$_5$	6.42$_3$
34.29	42.83	40.79	3.56	0.7805	0.1217	0.16$_3$	2.95$_1$	6.76$_5$
34.10	43.62	41.62	3.58	0.8437	0.0871	0.343$_9$	3.01$_3$	6.5$_1$

Quaternary

The results of GCMC simulations of the adsorption of quaternary mixture of methane(1)+ethane(2)+propane(3).+n-butane(4) are shown in Table S38.

Table S38. Summary of set up and main results of GCMC simulations in the gas phase for quaternary mixtures of methane(1)+ethane(2)+propane(3)+n-butane(4) at 300K.

						Adsorbed amount molecules/uc			
-μx10^{-3} [J/mol]				Px10^{-5}					
C1		C3	C4	[Pa]	y_1	C1	C2	C3	C4
June+LB									
36.23	3.50	41.91	39.81	3.50	0.3614	0.065$_5$	0.71$_9$	2.5$_1$	7.17$_5$
35.02	3.56	42.92	40.89	3.56	0.5818	0.142$_8$	0.69$_5$	2.2$_2$	7.3$_1$
34.52	3.58	43.78	41.85	3.58	0.7164	0.21$_6$	0.6$_2$	2.24$_5$	7.1$_4$
34.24	3.55	44.55	42.68	3.55	0.7973	0.28$_3$	0.52$_6$	1.94$_3$	7.3$_5$
34.08	3.57	45.21	43.40	3.57	0.8502	0.35$_1$	0.446$_7$	1.8$_2$	7.4$_3$

June+SW									
36.23	3.50	41.91	39.81	3.50	0.3614	0.1389	1.11	2.72	6.93
35.02	3.56	42.92	40.89	3.56	0.5818	0.252	0.813	2.381	7.22
34.52	3.58	43.78	41.85	3.58	0.7164	0.3928	0.733	2.321	7.11
34.24	3.55	44.55	42.68	3.55	0.7973	0.4997	0.5755	2.1645	7.181
34.08	3.57	45.21	43.40	3.57	0.8502	0.618	0.541	2.104	7.125
TaPPEZeo+LB									
36.23	3.50	41.91	39.81	3.50	0.3614	0.065	0.734	2.285	7.15
35.02	3.56	42.92	40.89	3.56	0.5818	0.127	0.623	2.083	7.25
34.52	3.58	43.78	41.85	3.58	0.7164	0.192	0.546	2.041	7.22
34.24	3.55	44.55	42.68	3.55	0.7973	0.2275	0.42	1.862	7.43
34.08	3.57	45.21	43.40	3.57	0.8502	0.292	0.522	2.065	7.09
Du									
36.23	3.50	41.91	39.81	3.50	0.3614	0.081	0.9849	3.12	6.15
35.02	3.56	42.92	40.89	3.56	0.5818	0.156	0.8348	2.73	6.43
34.52	3.58	43.78	41.85	3.58	0.7164	0.202	0.6275	1.91	7.14
34.24	3.55	44.55	42.68	3.55	0.7973	0.273	0.4397	2.364	6.71
34.08	3.57	45.21	43.40	3.57	0.8502	0.331	0.4454	1.81	7.11

2.2 – Adsorption in other zeolites

TON

The results of GCMC simulations of methane, ethane and propane in TON-type zeolite is presented in Tables S39, S40 and S41 respectively. The simulations used framework of 64 unit cells of TON (4x2x6).

Table S39. Summary of set up and results of GCMC simulations for methane (C1) adsorbed in TON-type zeolite

$-\mu_x 10^{-3}$ [J/mol]	$P_x 10^{-5}$ [Pa]	Adsorbed amount [molecules/unit cell]			
		LB	σ	SW	σ
T=298K					
35.0	1.8967	0.639	0.003	1.02	0.01
36.0	1.2657	0.464	0.005	0.767	0.002
37.0	0.8414	0.322	0.003	0.631	0.003
38.0	0.5641	0.23	0.01	0.455	0.002
40.0	0.2483	0.11	0.01	0.217	0.001
42.0	0.1122	0.048	0.002	0.087	0.003
45.0	0.0331	0.015	0.001	0.03	0.01
50.0	0.0044	0.002	0.002	0.004	0.001
55.0	0.0006	0.0002	0.0001	0.0006	0.0001
T=309K					
36.0	2.2999	0.617	0.001	1.088	0.003
38.0	1.0524	0.32	0.01	0.614	0.002

40.0	0.4834	0.157	0.004	0.312	0.001
42.0	0.2233	0.074	0.005	0.15	0.01
45.0	0.0689	0.023	0.002	0.049	0.001
50.0	0.0098	0.004	0.001	0.007	0.002

Table S40. Summary of set up and results of GCMC simulations for ethane (C2) adsorbed in TON-type zeolite.

$-\mu_x 10^{-3}$ [J/mol]	$P_x 10^{-5}$ [Pa]	Adsorbed amount [molecules/unit cell] LB	σ	[molecules/unit cell] SW	σ
T=298K					
50.0	0.0113	0.066	0.001	0.099	0.003
48.0	0.0254	0.144	0.005	0.215	0.001
45.0	0.0855	0.43	0.01	0.617	0.002
43.0	0.1895	0.80	0.03	1.07	0.02
42.0	0.2835	1.02	0.01	1.31	0.03
40.0	0.6414	1.435	0.005	1.718	0.001
38.0	1.4470	1.794	0.003	2.012	0.004
36.0	3.2798	2.02	0.03	2.11	0.01
T=309K					
55.0	0.0036	0.015	0.003	0.022	0.006
52.0	0.0117	0.05	0.01	0.069	0.002
50.0	0.0253	0.11	0.01	0.152	0.001
48.0	0.0556	0.212	0.002	0.310	0.002
45.0	0.1768	0.548	0.005	0.79	0.02
43.0	0.3870	0.847	0.002	1.22	0.01
41.0	0.8375	1.15	0.02	1.66	0.01

Table S41. Summary of set up and results of GCMC simulations for propane (C3) adsorbed in TON-type zeolite.

$-\mu_x 10^{-3}$ [J/mol]	$P_x 10^{-5}$ [Pa]	Adsorbed amount [molecules/unit cell] LB	σ	[molecules/unit cell] SW	σ
T=298K					
60.0	0.0004			0.023	0.001
58.0	0.0008	0.032	0.005		
55.0	0.0027	0.105	0.004	0.173	0.002
52.0	0.0090	0.31	0.02	0.53	0.02
50.0	0.0201	0.61	0.01	0.85	0.01
48.0	0.0451	1.008	0.008	1.236	0.003
45.0	0.1519	1.463	0.007	1.598	0.001
40.0	1.1515	1.86	0.04	1.91	0.01
36.0	6.2759	2.022	0.002	2.066	0.004
T=373K					
60.0	0.0816	0.191	0.003	0.285	0.002
58.0	0.1549	0.32	0.01	0.47	0.01
57.0	0.2140	0.434	0.002	0.585	0.001
55.0	0.4053	0.685	0.001	0.858	0.005

54.0	0.5636	0.80	0.01	1.00	0.04
53.0	0.7758	0.944	0.005	1.13	0.01
52.0	1.0631	1.08	0.01	1.257	0.003
50.0	2.0592	1.294	0.003	1.47	0.01

LTA

The results of GCMC simulations of adsorption of methane, ethane, propane and carbon dioxide in LTA-type zeolite is presented at Tables S42, S43, S44 and S45, respectively.

Table S42. Summary of set up and results of GCMC simulations for methane (C1) adsorbed in LTA-type zeolite.

$-\mu x10^{-3}$ [J/mol]	$Px10^{-5}$ [Pa]	Adsorbed amount [molecules/unit cell]		[molecules/unit cell]	
		LB	σ	SW	σ
T=300K					
31.0	10.5903	3.86	0.01	4.725	0.002
32.0	7.0001	3.062	0.004	3.89	0.01
33.7	3.5881	1.979	0.002	2.63	0.06
35.0	2.0861	1.34	0.01	1.86	0.01
36.0	1.4010	0.995	0.003	1.41	0.03
40.0	0.2811	0.25	0.01	0.422	0.001
45.0	0.0378	0.037	0.006	0.071	0.002
50.0	0.0051	0.005	0.002	0.009	0.003

Table S43. Summary of set up and results of GCMC simulations for ethane (C2) adsorbed in LTA-type zeolite.

$-\mu_x10^{-3}$ [J/mol]	P_x10^{-5} [Pa]	Adsorbed amount [molecules/unit cell]		[molecules/unit cell]	
		LB	σ	SW	σ
T=303K					
50.0	0.0164	0.31	0.02	0.335	0.005
46.0	0.0804	0.875	0.006	0.913	0.002
44.0	0.1775	1.334	0.005	1.38	0.01
42.0	0.3924	2.102	0.003	2.170	0.005
41.0	0.5875	2.69	0.02	2.79	0.01
40.0	0.8739	3.33	0.01	3.36	0.01
38.0	1.9545	4.611	0.004	4.703	0.003
36.0	4.4442	5.55	0.01	5.634	0.002
34.0	10.1136	6.24	0.01	6.28	0.02

Table S44. Summary of set up and results of GCMC simulations for propane (C3) adsorbed in LTA-type zeolite.

$-\mu_x 10^{-3}$ [J/mol]	$P_x 10^{-5}$ [Pa]	Adsorbed amount [molecules/unit cell]		[molecules/unit cell]	
		LB	σ	SW	σ
T=300K					
52.0	0.0105	0.563	0.02	0.73	0.01
49.0	0.0350	1.158	0.003	1.488	0.002
47.0	0.0783			2.651	0.003
46.0	0.1172	2.71	0.01	3.35	0.01
45.0	0.1738			3.908	0.005
43.0	0.3883	4.239	0.005	4.41	0.01
41.0	0.8728	4.57	0.02	4.74	0.01
39.0	1.9718	5.031	0.002	5.204	0.004
38.0	2.9747	5.21	0.03	5.42	0.02
37.0	4.5307	5.43	0.01	5.52	0.01

Table S45. Summary of set up and results of GCMC simulations for carbon dioxide (CO_2) adsorbed in LTA-type zeolite.

$-\mu_x 10^{-3}$ [J/mol]	$P_x 10^{-5}$ [Pa]	Adsorbed amount [molecules/unit cell]		[molecules/unit cell]	
		LB	σ	SW	σ
T=303K					
37.0	5.2512	5.46	0.01	7.012	0.005
38.0	3.4817	4.139	0.002	5.30	0.01
40.0	1.5523	1.858	0.005	2.227	0.005
42.0	0.7045	0.84	0.03	1.013	0.002
43.0	0.4747	0.57	0.01	0.67	0.02
45.0	0.2155	0.263	0.003	0.305	0.005

CHA

The results of GCMC simulations of methane, ethane and CO_2 adsorbed in CHA-type zeolite are presented in Table S46.

Table S46. Summary of set up and results of GCMC simulations of methane, ethane and CO_2 adsorbed in CHA-type zeolite at 303K.

$-\mu_x10^{-3}$ [J/mol]	P_x10^{-5} [Pa]	Adsorbed amount [molecules/unit cell] LB	σ	[molecules/unit cell] SW	σ
		CH_4			
45.0	0.0466	0.032	0.003	0.051	0.002
42.0	0.1527	0.11	0.01	0.210	0.009
40.0	0.3391	0.228	0.006	0.373	0.007
39.0	0.5020	0.34	0.01	0.55	0.02
38.0	0.7522	0.518	0.002	0.807	0.002
37.0	1.1158	0.714	0.007	1.16	0.01
		C_2H_6			
50.0	0.0113	0.087	0.005	0.13	0.01
47.0	0.0378	0.29	0.02	0.445	0.005
45.0	0.0855	0.626	0.003	0.955	0.001
43.0	0.1908	1.32	0.02	1.95	0.02
40.0	0.6414	3.26	0.01	4.05	0.01
38.0	1.4470	4.54	0.01	5.1	0.1
		CO_2			
50	0.0299	0.07	0.01	0.09	0.01
45	0.2155	0.530	0.003	0.698	0.005
43	0.4747	1.16	0.05	1.617	0.002
42	0.7045	1.703	0.004	2.459	0.001
41	1.0458	2.46	0.02	3.604	0.003
40	1.5523	3.53	0.01	5.30	0.02
38	3.4817	6.23	0.02	8.65	0.01
35	11.7925	10.26	0.01	12.29	0.02

3- FITTING THE EXPERIMENTAL DATA

3.1.1- Isotherms of pure components adsorption

The experimental data of adsorption of pure components were fit by the Dual-Site Langmuir (DSL) model/isotherm aiming to get numerical results to compare the performance of different set of zeolite force field/combining rules. The Dual-Site Langmuir isotherm is a model with 4 adjustable parameters which provides a flexible mathematical expression to correlate pure-gas adsorption isotherms. The model proposed by Myers[13] for adsorption on heterogeneous surfaces derives from

the use of bimodal two-site discrete distribution with the Langmuir isotherm. Mathias et al[14] presents a clear description of the model which is expressed by:

$$q = \frac{{q_{max}}^{(I)} K^{(I)} P}{1 + K^{(I)} P} + \frac{{q_{max}}^{(II)} K^{(II)} P}{1 + K^{(II)} P} \qquad \text{S1}$$

in eq. S1 q_{max} and K denote the adsorption capacity and affinity parameter, respectively. The superscript I and II denote the two discrete sites. Despite of the parameters present a thermodynamic meaning in the present work the fitting was made in order to maximize the agreement with experimental data only inside the data range. Therefore, no bounds were applied in the correlation what lead sometimes the best fit was achieved by a set of parameters without thermodynamic meaning such as negative values for some parameters.

The fit of experimental isotherm data were performed using software STATISTICA (7). The nonlinear estimation applied the minimization of least squares function by the Levenberg-Marquard method.

Table S.47 presents the values of Dual-Site Langmuir (DSL) parameters obtained for each pure-gas adsorption isotherm on zeolite type MFI. Table S48 presents the DSL parameters for adsorption of methane. Ethane, propane and CO2 on zeolites type CHA, LTA and TON. The absolute mean deviation (AMD) and R_2 are also presented in the tables.

Table S47. Summary of Dual Site Langmuir (DSL) fitted parameters for pure components in MFI. The fit correspond to the experimental data

Adsorbate			Dual Site Langmuir parameters					
		$T_{[Ref]}$	$q_{max(I)}$	$K_{(I)}$	$q_{max(II)}$	$K_{(II)}$	R^2	AMD
Formula	**Acronym**	**[K]**	**molec/uc**	**[bar^{-1}]**	**molec/uc**	**[bar^{-1}]**	**-**	**molec/uc**
CH_4	C1	300[2]	14.6040	0.4539	25.4764	0.0016	0.99964	0.092
CH_4	C1	338[3]	2.7617	0.01956	2.8494	0.1336	0.99997	0.010
CH_4	C1	373[3]	15.2003	0.04198	3.2657	0.1521	0.99992	0.012
C_2H_6	C2	300[2]	3.0449	0.5675	11.6432	14.4982	0.99997	0.027
C_2H_6	C2	338[3]	6.5441	1.7545	6.5336	1.7544	0.9997	0.052
C_2H_6	C2	373[3]	5.9494	0.5868	7.1779	0.5868	0.9984	0.111
C_3H_8	C3	300[2]	1.1565	1.6927	10.4521	125.7293	0.9997	0.062
C_3H_8	C3	338[3]	1.8960	1.2389	9.9818	14.8942	0.9999	0.056
C_3H_8	C3	373[3]	4.3159	0.1202	10.0243	3.6163	0.9995	0.082
C_4H_{10}	C4	300[2]	1.3079	2.0160	9.0756	960.7012	0.9980	0.085
C_4H_{10}	C4	338[3]	8.0205	208.7907	1.9212	3.1204	0.9999	0.016
C_4H_{10}	C4	373[3]	8.8343	27.0013	120.8748	0.0021	0.9998	0.032
C_5H_{12}	C5	303[9]	17.818	1.6000	8.0940	7490.770	0.9987	0.074
C_5H_{12}	C5	323[9]	2.160	326.252	6.126	3532.04	0.9999	0.015
C_5H_{12}	C5	343[9]	3.5410	152.949	4.646	1424.131	0.9999	0.012
C_6H_{14}	nC6	303[9]	0.6800	147.5400	7.84	22777.82	0.9999	0.008
C_6H_{14}	nC6	303[27]	7.5800	34129.15	0.77	39.10	0.9998	0.010
C_6H_{14}	nC6	343[9]	3.770	277.30	4.22	12363.70	0.9992	0.060
C_6H_{14}	nC6	373[9]	3.6148	103.1040	4.2776	2.8096	0.9999	0.002
C_6H_{14}	nC6	373[27]	239.753	0.410	4.402	1849.141	0.9995	0.027
C_6H_{14}	nC6	373[10]	5.092	27.934	3.840	2516.878	0.9994	0.042
C_6H_{14}	2mC5	423[15]	4.1830	332.3215	-0.0309	5.3236	0.9997	0.021
CO_2	CO_2	277[6]	3.0088	0.2757	16.7432	2.9999	0.9998	0.052
CO_2	CO_2	305[7]	260.7788	0.12868	-2505.25	0.0086	0.9995	0.104
CO_2	CO_2	308[6]	16.8822	0.9906	9.0421	0.0065	0.9984	0.172
CO_2	CO_2	353[6]	15.5438	0.3126	7.8355	0.0080	0.9998	0.055
CH_4O	C1OH	303[16]	7.4023	53.1747	13.0688	53.1759	0.9888	0.739

1) Adsorption of binary, ternary and quaternary mixtures

The correlation of adsorbed amount of each component present in the mixture was done most of cases by general polynomial expressions:

$$q_i = ao + a1y_1 + a2_i{y_1}^2 + a3_i{y_1}^3 + a4_i{y_1}^4 + a5_i{y_1}^5 + a6_i{y_1}^6 \quad \text{S2}$$

where q_i is the adsorbed amount of component *i*. *ao*, *a1*, *a2*, *a3*, *a4*, *a5* and *a6* are the polynomial coefficients and y_1 is the molar fraction of component 1 in gas phase.

In most of cases of methane adsorbed in mixtures, the data need to be split on up to 3 data sets to get a high correlation. Also in a few cases, different expressions, as following, were used:

$$q_{cH4} = \frac{ao y_{CH4}}{1+a1 y_{CH4}} exp(a2 y_{CH4}) \quad \text{S3}$$

Tables S49, S50, S51 and S52 present the coefficients values for methane (C1), ethane (C2), propane (C3) and n-butane (C4), respectively. The parameters were fitted from experimental data of adsorption of these components in mixtures at 300K and 3.45 bar in silicalite (all silica MFI).

The absolute mean deviation (AMD) expressed in terms of molecules adsorbed by unit cell and the relative mean deviation (RMD) expressed in percentage were evaluated by the following expressions:

$$AMD = \frac{\sum_{j=1}^{NP} \left| q_j^{GCMC} - q_j^{fitexp} \right|}{NP} \quad \text{S4}$$

$$RMD = \frac{1}{NP} \sum_{j=1}^{NP} \frac{\left| q_j^{GCMC} - q_j^{fitexp} \right|}{qj_j^{fitexp}} x\, 100\% \quad \text{S5}$$

where q_{jGCMC} is the average of adsorbed amount obtained from 3 independent GCMC simulations. $q_{jfit\ exp}$ is the adsorbed amount calculated by the expression used to fit the experimental data and *NP* is the number of experimental data.

It is important to notice that the AMD and RMD were evaluated only inside the data range of experimental data.

Table S48. Summary of Dual Site Langmuir (DSL) fitted parameters for pure components in LTA. CHA and TON-type zeolite. The fits correspond to experimental data

			Dual Site Langmuir parameters					
Adsorbate	**Zeolite**	**$T_{[Ref]}$**	**q_{maxI}**	**K_I**	**q_{maxII}**	**K_{II}**	**R^2**	**AMD**
		[K]	**molec/uc**	**[bar^{-1}]**	**molec/uc**	**[bar^{-1}]**	**-**	**molec/uc**
CH_4	LTA	300[17]	5.9994	0.1813	0.0552	-235899	0.9988	0.024
CH_4	CHA	303[22]	2.9715	0.1956	3.0563	0.1954	>0.9999	0.001
CH_4	TON	298[23]	0.6693	0.7421	0.5373	0.7420	0.9993	0.004
CH_4	TON	309[24]	0.0715	4.4278	1.7012	0.4179	>0.9999	0.001
C_2H_6	LTA	303[18]	-2.2315	-3.053	8.1761	5.0756	>0.9999	0.010
C_2H_6	LTA	303[19]	330.7163	0.0001	5.8430	3.9076	0.9990	0.046
C_2H_6	CHA	303[22]	6.5356	4.3215	-0.2990	0.5497	>0.9999	0.008
C_2H_6	CHA	303[17]	4.3305	5.3726	4.9809	0.3096	0.9994	0.027
C_2H_6	TON	298[25]	1.5547	11.6322	0.4557	1.2238	0.9989	0.020
C_2H_6	TON	309[24]	0.49364	27.6683	1.24553	4.4556	0.9999	0.003
C_3H_8	LTA	300[17]	3.1189	61.9345	1.0284	0.2628	0.9956	0.060
C_3H_8	LTA	323[20]	3.2509	88.3104	3.6543	0.5337	0.9998	0.011
C_3H_8	TON	298[25]	0.3929	2.1255	1.3374	123.0813	0.9996	0.012
C_3H_8	TON	373[13]	1.2857	9.1697	-0.6866	0.02333	0.9994	0.006
CO2	LTA	303[21]	8.61864	0.1050	9.1606	0.1050	0.9986	0.104
CO2	CHA	303[26]	3.3931	0.3850	11.4306	0.3851	0.9999	0.015

Table S49. Summary of coefficient values fitted from experimental data of methane adsorption in n-alkane mixture in silicalite at 300K and 345kPa. Experimental data of Abdul- Rehman et al.[3]

Mixture	Data Range	$q_{CH4}=ao+a1y_{Ch4}+a2*y_{Ch42}+a3y_{Ch43}+a4y_{Ch44}+a5y_{Ch45}+a6y_{Ch46}$ [molecule/uc]							AMD
		Ao	a1	a2	a3	a4	a5	a6	
C1C2	$0.30<y_1<0.84$	0	-139.26	1223.56	-4049.78	6444.45	-4960.34	1490.0	0.06
C1C3	$0.09<y_1<0.58$	0	2.8789	-12.3615	15.8997	0	0	0	0.00
	$0.58<y_1<0.94$	0	138.75	-745.64	1512.63	-1367.58	466.52	0	0.05
C1C4	$0.5<y_1<0.85$	0	2.0718	-3.250	1.6719	0	0	0	0.0
	$0.85<y_1<0.94$	$q_{CH4}=28600.6y_{Ch4}/(1+10737.6y_{Ch4})\exp(-13.2y_{Ch4})$							0.02
	$0.94<y_1<0.97$	$q_{CH4}=0.0586y_{Ch4}/(1+1.0025y_{Ch4})$							0.04
C1C2C3	$0.35<y_1<0.68$	0	2.49	-10.87	12.45	0	0	0	0.06
	$0.68<y_1<0.80$	25.79	-71.12	50.03	0	0	0	0	0.00
C1C2C4	$0.35<y_1<0.70$	0	5.07	-17.59	16.12	0	0	0	0.00
	$0.70<y_1<0.84$	0	4.23	-10.51	7.72	0	0	0	0.00
C1C3C4	$0.32<y_1<0.55$	0	0	0	0	0	0	0	0.00
	$0.55<y_1<0.84$	1.58	-4.52	3.31	0	0	0	0	0.01
C1C2C3C4	$0.35<y_1<0.85$	-3.82	20.65	-34.30	19.02	0	0	0	0.03

Table S50. Summary of polynomial coefficients fitted from experimental data of ethane adsorption in n-alkane mixture in silicalite at 300K and 345kPa. Experimental data of Abdul- Rehman et al.[3]

Mixture	Data Range	$q_{C2H6}=ao+a1y_1+a2*y_{12}+a3y_{13}+a4y_{14}+a5y_{15}+a6y_{16}$ [molecule/uc]							AMD
		ao	a1	a2	a3	a4	a5	a6	
C1C2	$0.30<y_1=y_{C1}<0.84$	15.71	-45.92	263.07	754.99	-4960.34	-321.97	0	0.09
C2C3	$0.28<y_1=y_{C2}<0.74$	0	-59.75	569.23	-2010.98	3474.0	-2916.34	954.48	0.06
C1C2C3	$0.35<y_1=y_{C1}<0.80$	5.03	-6.31	11.74	-8.83	0	0	0	0.00
C1C2C4	$0.35<y_1=y_{C1}<0.84$	2.17	-2.83	4.25	-3.19	0	0	0	0.00
C1C2C3C4	$0.35<y_1=y_{C1}<0.85$	1.204	-0.593	1.087	-1.218	0	0	0	0.00

Table S51. Summary of polynomial coefficients fitted from experimental data of propane adsorption in n-alkane mixture in silicalite at 300K and 345kPa. Experimental data of Abdul- Rehman et al.[3]

Mixture	Data Range	$q_{C3H8}=ao+a1y_1+a2*y_{12}+a3y_{13}+a4y_{14}+a5y_{15}+a6y_{16}$ [molecule/uc]							AMD
		ao	a1	a2	a3	a4	a5	a6	
C1C3	$0.09<y_1=y_{C1}<0.94$	11.56	19.54	126.88	399.19	663.00	554.59	185.68	0.01
C2C3	$0.28<y_1=y_{C2}<0.74$	10.82	25.30	-229.30	793.24	-1371.12	1155.74	-384.68	0.00
C1C2C3	$0.35<y_1=y_{C1}<0.80$	9.02	-8.20	14.55	-8.95	0	0	0	0.01
C1C3C4	$0.32<y_1=y_{C1}<0.84$	3.14	-5.19	10.69	-7.06	0	0	0	0.08
C1C2C3C4	$0.35<y_1=y_{C1}<0.85$	2.57	-2.05	3.67	-2.56	0	0	0	0.00

Table S52. Summary of polynomial coefficients fitted from experimental data of butane adsorption in n-alkane mixture in silicalite at 300K and 345kPa. Experimental data of Abdul-Rehman et al.[3]

Mixture	Data Range	$q_{C4H10}=ao+a1y_{ch4}+a2*y_{ch42}+a3y_{ch43}+a4y_{ch44}+a5y_{ch45}\ a6y_{ch46}$ [molecule/uc]							AMD
		Ao	a1	a2	a3	a4	a5	a6	
C1C4	$0.50<y_{ch4}<0.97$	10.09	1095.33	-7274.75	18997.20	-24464.43	15576.7	-3931.48	0.00
C1C2C4	$0.35<y_{ch4}<0.84$	8.378	-2.427	4.185	-2.6257	0	0	0	0.06
C1C3C4	$0.32<y_{ch4}<0.84$	8.811	-4.076	8.251	-5.3559	0	0	0	0.00
C1C2C3C4	$0.35<y_{ch4}<0.85$	8.671	-0.445	1.006	-0.9335	0	0	0	0.00

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