

Supplementary Information

Methane adsorption in ADOR zeolites: A combined experimental and DFT/CC study

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KEYWORDS

methane, zeolites, adsorption, ab initio calculations.

Content

Figure S1. Cluster models and methane orientations (C_{2v}) used in DFT/CC parameterization.

Figure S2. Performance of the DFT/CC method for (a) 2T...CH₄ complexes and (b) 4T...CH₄ complexes (C_{2v} symmetry).

Figure S3. Wide angle XRD patterns of the zeolites: a – UTL, b – OKO, c – PCR, d – MFI.

Figure S4. (A) Adsorption isotherms of argon on investigated zeolites at the temperature of liquid argon, (B) pore size distribution calculated by NL DFT method. a – UTL, b – OKO, c – PCR, d – MFI.

Table S1. Parameters of the AI-FF potential with a functional form of $V(r) = Ar^{-7} + Br^{-9} + Cr^{-12} + D2(s_6)$, where D2 denotes the Grimme's (2006) dispersion correction.

Table S2. Isosteric heats of adsorption at zero loading (in kJ/mol) calculated using flexible and rigid-framework MD simulations.

Table S3. Self-diffusion coefficients of methane in silicalite-1 at zero loading (300 K).

Figure S1

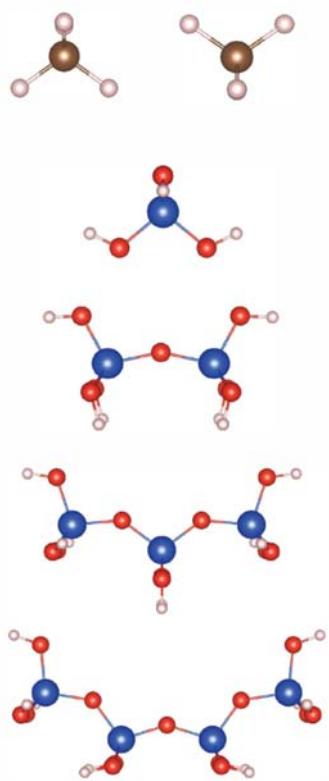


Figure S2

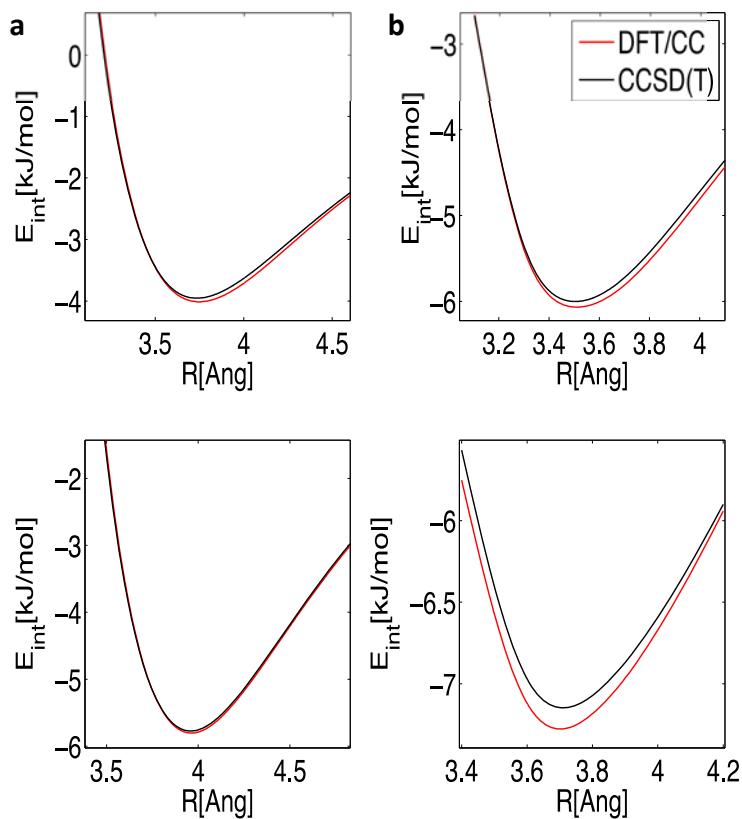


Figure S3

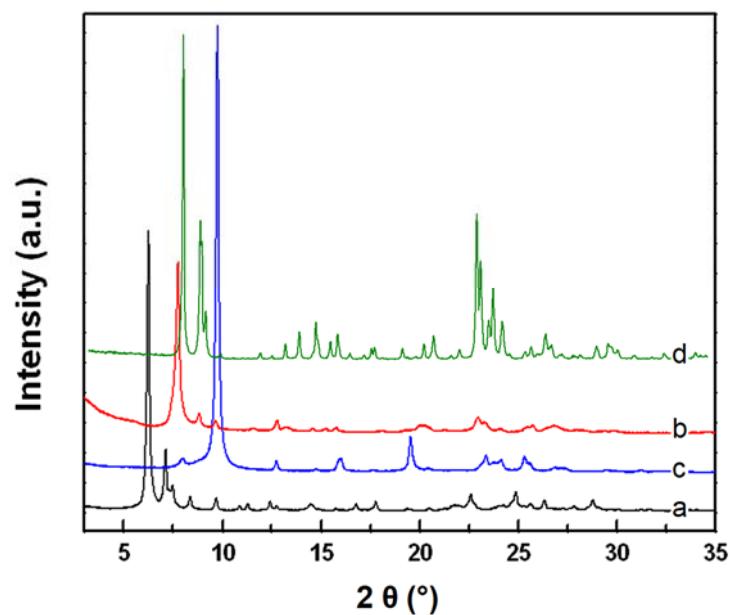


Figure S4

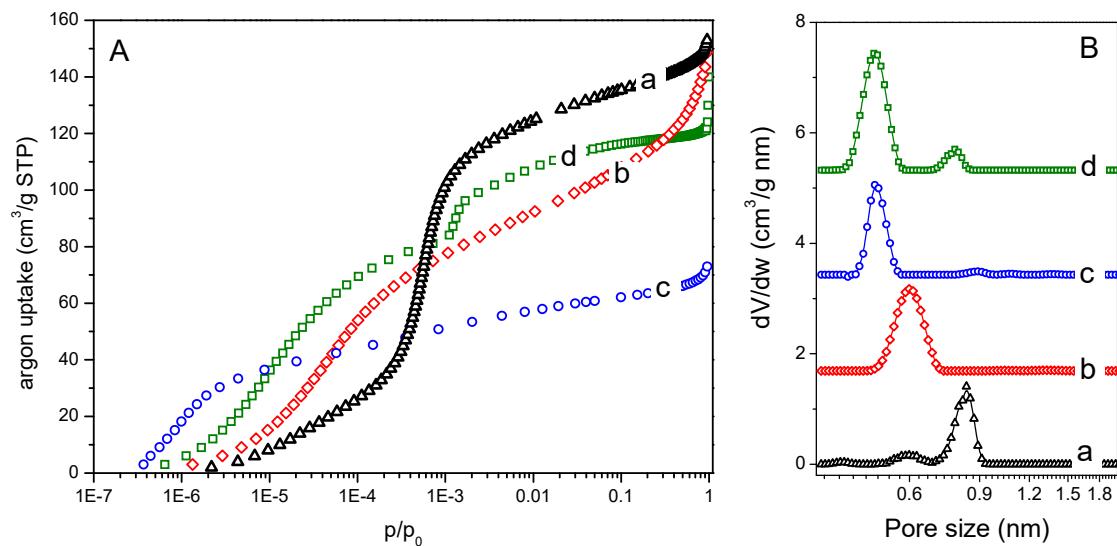


Table S1 Parameters of the AI-FF potential with a functional form of $V(r) = Ar^{-7} + Br^{-9} + Cr^{-12} + D2(s_6)$, where D2 denotes the Grimme's (2006) dispersion correction.

Interaction potential	A [kJ mol ⁻¹ nm ⁷]	B [kJ mol ⁻¹ nm ⁹]	C [kJ mol ⁻¹ nm ¹²]	s6
C – Ge	0	-1.332E-04	1.505E-09	0.9375
C – Si	0	-1.533E-04	1.505E-08	0.9375
C – O	-2.431E-03	3.788E-04	3.416E-07	0.9375
H – Ge	1.829E-04	2.189E-05	1.372E-09	0.09375
H – Si	6.576E-04	0	1.372E-09	0.9375
H – O	0	0	3.017E-08	0.9375

Table S2. Isosteric heats of adsorption at zero loading (in kJ/mol) calculated using flexible and rigid-framework MD simulations.

System	flexible framework	rigid framework
UTL	15.40	15.31
OKO	17.75	17.74
PCR	21.98	21.91
MFI	20.22	20.20

Table S3. Self-diffusion coefficients of methane in silicalite-1 at zero loading (300 K).

Description	D _s [10 ⁻⁸ m ² s ⁻¹]	Reference	Note
MFI - flexible	1.51	this work ^a	theory
MFI - rigid	1.52	this work ^a	theory
June <i>et al.</i>	1.61	[1]	theory
Beerdsen <i>et al.</i>	1.46	[2]	theory
Goodbody <i>et al.</i>	1.34	[3]	theory
Nicholas <i>et al.</i>	1.15	[4]	theory
Caro <i>et al.</i>	1.05	[5]	experiment
Jobic <i>et al.</i>	0.50 ^b	[6]	experiment
Kar and Chakravarty	0.39	[7]	theory
Catlow <i>et al.</i>	0.36	[8]	theory
Talu <i>et al.</i>	0.32	[9]	experiment
Sun <i>et al.</i>	0.08	[10]	experiment

^a Calculated using the Einstein relation.^b Measured at 250 K.

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