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

Supporting Information

for

Insight into the effect of interlayer spacings of bilayer graphene on the desolvation of H^+, Li^+, Na^+, and K^+ ions with water as solvent: A First-Principles Study

Xu Zhang,[a] Shaobin Yang,*[a] Xueying Shan,[a] Sinan Li,[b] and Shuwei Tang[a]

[a] *College of Materials Science and Engineering, Liaoning Technical University, Fuxin, Liaoning 123000, China.*

[b] *School of Metallurgy Engineering, Liaoning Institute of Science and Technology, Benxi, Liaoning 117004, China.*

**Corresponding Author: *lgdysb@163.com* (Shaobin Yang)

Table S1 Structural parameters and adsorption energies of different adsorption systems

Structure	θ/°	*R*/Å		Others/Å	E_{ad}/eV		Others
		AA	AB		AA	AB	
I	0	3.311	3.325	3.35[a]	-0.157	-0.150	-11.4 kJ/mol(-0.118)[a] -2.9 kcal/mol(-0.126)[b]
	30	3.324	3.337		-0.140	-0.181	
	60	3.321	3.336		-0.136	-0.135	
	90	3.312	3.326		-0.140	-0.130	
II	0	3.329	3.334	3.26[a]	-0.169	-0.149	-13.3 kJ/mol(-0.138)[a]
	30	3.330	3.335		-0.149	-0.128	
	60	3.330	3.335		-0.141	-0.121	
	90	3.328	3.332		-0.148	-0.127	
III	0	3.044	2.972	3.01[a]	-0.092	-0.103	-8.7 kJ/mol(-0.090)[a]
	30	3.043	2.972		-0.071	-0.083	
	60	3.039	2.974		-0.071	-0.077	
	90	3.046	3.05		-0.070	-0.063	
IV	0	3.143	3.157	3.12[a]	-0.140	-0.122	-10.0 kJ/mol(-0.104)[a]
	30	3.144	3.152		-0.129	-0.106	
	60	3.142	3.153		-0.126	-0.104	
	90	3.143	3.153		-0.133	-0.110	

[a]DFT/CC based on AVQZ[1]; [b]DFTB-D approach[2]

In the table, θ is the rotation angle, and *R* is the distance from the O atom to the carbon plane. The values in parentheses are the corresponding adsorption energy values when the unit is converted into eV.

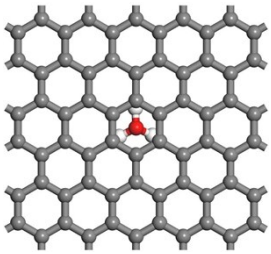
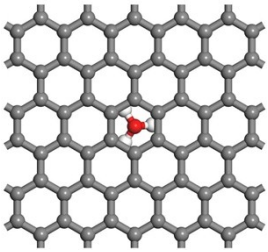
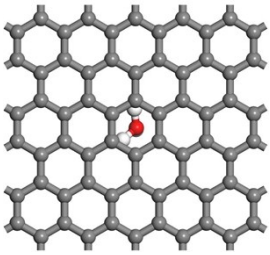
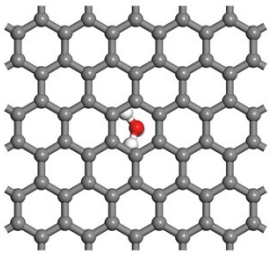
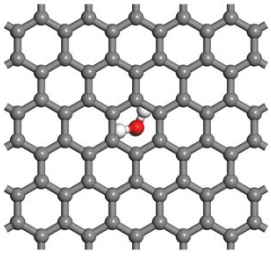
Table S2 Total energies of BG-H_2O intercalation systems

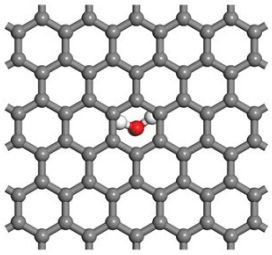
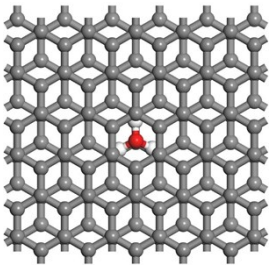
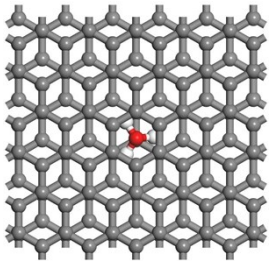
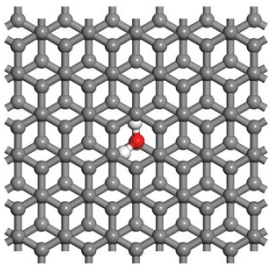
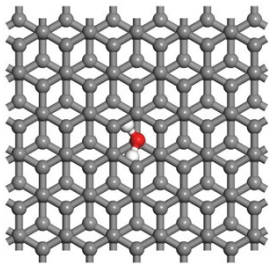
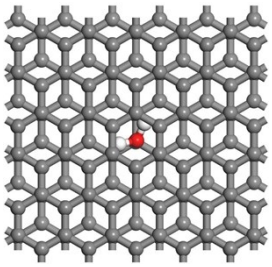
d(Å)	Types	Energies(eV)			
		0°	30°	60°	90°
d_{AA} = 4	I	-19086.596944	-19086.608445	-19086.676386	-19086.583904
	II	-19086.604511	-19086.598714	-19086.582973	-19086.582483
	IV	-19086.697450	-19086.685678	-19086.687537	-19086.682906
d_{AA} = 5	I	-19086.713836	-19086.708656	-19086.687714	-19086.695935
	II	-19086.673861	-19086.659209	-19086.642808	-19086.654551
	IV	-19086.733549	-19086.719395	-19086.720108	-19086.717490
d_{AA} = 6	I	-19086.675723	-19086.667107	-19086.657244	-19086.659636
	II	-19086.679672	-19086.659914	-19086.652610	-19086.658968
	IV	-19086.644111	-19086.640452	-19086.640778	-19086.634758
d_{AA} = 7	I	-19086.536771	-19086.528752	-19086.520774	-19086.519633
	II	-19086.538016	-19086.516788	-19086.510419	-19086.516783
	IV	-19086.512729	-19086.500955	-19086.500342	-19086.505482
d_{AA} = 8	I	-19086.381209	-19086.377293	-19086.368453	-19086.372471
	II	-19086.386141	-19086.356019	-19086.356488	-19086.364058
	IV	-19086.371021	-19085.663953	-19085.660817	-19086.363776
d_{AA} = 9	I	-19086.329571	-19086.317467	-19086.309481	-19086.313612
	II	-19086.329260	-19086.308479	-19086.301701	-19086.308355
	IV	-19086.319821	-19086.309183	-19086.307402	-19086.314030
d_{AA} = 10	I	-19086.287587	-19086.268738	-19086.264266	-19086.272312
	II	-19085.524992	-19085.504135	-19085.496258	-19085.503689
	IV	-19086.273719	-19086.265805	-19085.511970	-19086.277314
d_{AB} = 4	I	-19086.715653	-19086.721891	-19086.806519	-19086.666033
	II	-19086.784470	-19086.733999	-19086.769780	-19086.703259
	III	-19086.698284	-19086.690785	-19086.791601	-19086.667452
	IV	-19086.809648	-19086.802846	-19086.773199	-19086.785338
d_{AB} = 5	I	-19086.643035	-19086.635266	-19086.622068	-19086.622136
	II	-19086.672708	-19086.658253	-19086.642561	-19086.648800
	III	-19086.640432	-19086.628987	-19086.613288	-19086.625899
	IV	-19086.695112	-19086.677887	-19086.683073	-19086.676056
d_{AB} = 6	I	-19086.681113	-19086.672704	-19086.663586	-19086.665798
	II	-19086.662802	-19086.643303	-19086.636415	-19086.642736
	III	-19086.670678	-19086.651483	-19086.644685	-19086.651759
	IV	-19086.633727	-19086.627922	-19086.627135	-19086.627753
d_{AB} = 7	I	-19086.538022	-19086.530079	-19086.521527	-19086.521338
	II	-19086.524921	-19086.504941	-19086.498770	-19086.504891
	III	-19086.529642	-19086.509491	-19086.502489	-19086.509466
	IV	-19086.500283	-19086.487967	-19086.487541	-19086.492960
d_{AB} = 8	I	-19086.378827	-19086.357354	-19086.360531	-19086.361732
	II	-19086.369282	-19086.267482	-19086.262246	-19086.270620

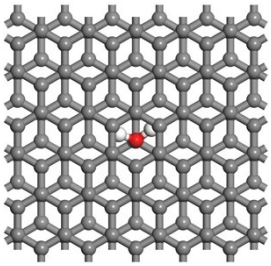
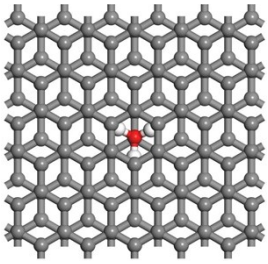
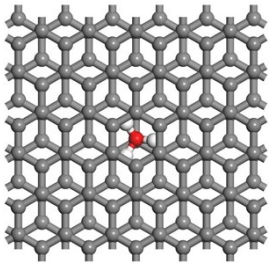
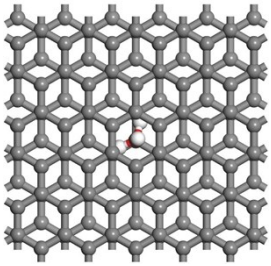
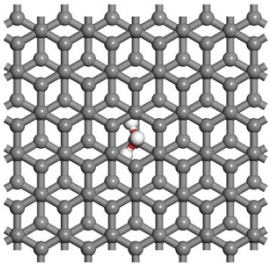
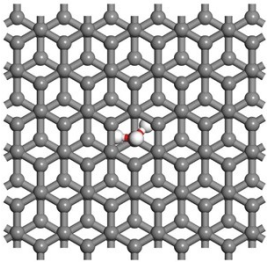
	III	-19086.369972	-19086.348121	-19086.347599	-19086.355614
	IV	-19086.358833	-19086.351839	-19086.349596	-19086.351002
$d_{AB} = 9$	I	-19086.316528	-19086.304073	-19086.295081	-19086.300100
	II	-19086.313062	-19086.293287	-19086.286063	-19086.292781
	III	-19086.313950	-19086.294690	-19086.286757	-19086.293686
	IV	-19086.303698	-19086.289743	-19086.288135	-19086.296857
$d_{AB} = 10$	I	-19086.261647	-19086.236771	-19086.247695	-19086.245384
	II	-19085.448144	-19085.427906	-19085.420522	-19085.427006
	III	-19085.446982	-19085.426674	-19085.418934	-19085.425621
	IV	-19086.256051	-19085.426516	-19085.425030	-19086.247027

In the table, d_{AA} and d_{AB} represent the original interlayer spacings of the AA- and AB-stacking BG, respectively.

Table S3 The total energies of the system of hydronium ion intercalated in BG with different interlayer spacings

models	d(Å)	Energies(eV)
	d_{AA} = 4	-19100.996408
	d_{AA} = 5	-19100.780441
	d_{AA} = 6	-19101.153689
	d_{AA} = 7	-19099.640794
	d_{AA} = 8	-19099.464329
	d_{AA} = 9	-19099.399320
	d_{AA} = 10	-19099.335089
	d_{AA} = 4	-19101.019943
	d_{AA} = 5	-19100.828248
	d_{AA} = 6	-19100.214445
	d_{AA} = 7	-19099.634752
	d_{AA} = 8	-19099.459686
	d_{AA} = 9	-19099.396181
	d_{AA} = 10	-19099.332435
	d_{AA} = 4	-19100.943544
	d_{AA} = 5	-19100.786868
	d_{AA} = 6	-19100.103174
	d_{AA} = 7	-19099.703529
	d_{AA} = 8	-19099.484368
	d_{AA} = 9	-19099.412018
	d_{AA} = 10	-19099.351182
	d_{AA} = 4	-19100.970787
	d_{AA} = 5	-19100.800329
	d_{AA} = 6	-19100.104078
	d_{AA} = 7	-19099.703369
	d_{AA} = 8	-19099.480074
	d_{AA} = 9	-19099.412913
	d_{AA} = 10	-19099.350443
	d_{AA} = 4	-19100.902589
	d_{AA} = 5	-19100.810857
	d_{AA} = 6	-19100.143511
	d_{AA} = 7	-19099.717111
	d_{AA} = 8	-19099.491653
	d_{AA} = 9	-19099.419024
	d_{AA} = 10	-19099.354440

	d_{AA} = 4	-19100.968776
	d_{AA} = 5	-19100.803413
	d_{AA} = 6	-19100.123538
	d_{AA} = 7	-19099.720362
	d_{AA} = 8	-19099.497881
	d_{AA} = 9	-19099.423116
	d_{AA} = 10	-19099.362470
	d_{AB} = 4	-19100.926303
	d_{AB} = 5	-19100.781723
	d_{AB} = 6	-19101.152514
	d_{AB} = 7	-19099.634315
	d_{AB} = 8	-19099.450606
	d_{AB} = 9	-19099.389575
	d_{AB} = 10	-19099.322569
	d_{AB} = 4	-19100.928891
	d_{AB} = 5	-19100.656067
	d_{AB} = 6	-19101.068756
	d_{AB} = 7	-19099.623563
	d_{AB} = 8	-19099.445893
	d_{AB} = 9	-19099.385779
	d_{AB} = 10	-19099.316732
	d_{AB} = 4	-19100.943984
	d_{AB} = 5	-19100.746128
	d_{AB} = 6	-19101.054696
	d_{AB} = 7	-19101.003184
	d_{AB} = 8	-19099.481753
	d_{AB} = 9	-19099.403483
	d_{AB} = 10	-19099.333364
	d_{AB} = 4	-19100.951243
	d_{AB} = 5	-19100.750141
	d_{AB} = 6	-19101.056426
	d_{AB} = 7	-19100.986124
	d_{AB} = 8	-19099.480215
	d_{AB} = 9	-19099.402588
	d_{AB} = 10	-19099.332476
	d_{AB} = 4	-19100.946666
	d_{AB} = 5	-19100.746247
	d_{AB} = 6	-19101.055422
	d_{AB} = 7	-19100.963971
	d_{AB} = 8	-19099.492024
	d_{AB} = 9	-19099.408962
	d_{AB} = 10	-19099.336565

	d_{AB} = 4	-19100.952718
	d_{AB} = 5	-19100.759975
	d_{AB} = 6	-19101.069918
	d_{AB} = 7	-19100.971196
	d_{AB} = 8	-19099.496955
	d_{AB} = 9	-19099.415962
	d_{AB} = 10	-19099.343456
	d_{AB} = 4	-19100.962525
	d_{AB} = 5	-19100.772498
	d_{AB} = 6	-19101.145415
	d_{AB} = 7	-19099.641956
	d_{AB} = 8	-19099.449962
	d_{AB} = 9	-19099.390707
	d_{AB} = 10	-19099.321818
	d_{AB} = 4	-19100.977910
	d_{AB} = 5	-19100.760223
	d_{AB} = 6	-19100.212073
	d_{AB} = 7	-19099.628480
	d_{AB} = 8	-19099.456631
	d_{AB} = 9	-19099.386171
	d_{AB} = 10	-19099.317471
	d_{AB} = 4	-19100.997803
	d_{AB} = 5	-19100.773854
	d_{AB} = 6	-19101.111840
	d_{AB} = 7	-19099.708467
	d_{AB} = 8	-19099.482680
	d_{AB} = 9	-19099.405580
	d_{AB} = 10	-19099.332363
	d_{AB} = 4	-19101.002302
	d_{AB} = 5	-19100.761704
	d_{AB} = 6	-19101.078171
	d_{AB} = 7	-19099.717433
	d_{AB} = 8	-19099.478909
	d_{AB} = 9	-19099.405355
	d_{AB} = 10	-19099.331910
	d_{AB} = 4	-19100.991541
	d_{AB} = 5	-19100.765533
	d_{AB} = 6	-19101.101391
	d_{AB} = 7	-19099.712171
	d_{AB} = 8	-19099.491874
	d_{AB} = 9	-19099.416389
	d_{AB} = 10	-19099.340043

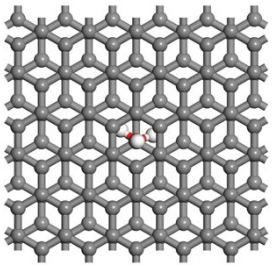
	d_{AB} = 4	-19100.961386
	d_{AB} = 5	-19100.800744
	d_{AB} = 6	-19101.114067
	d_{AB} = 7	-19099.712114
	d_{AB} = 8	-19099.494469
	d_{AB} = 9	-19099.416757
	d_{AB} = 10	-19099.342315

Table S4 Aqueous electrolyte-based supercapacitors and their performance[3]

Aqueous electrolyte/concentration	Electrode materials	Specific capacitance (F/g)	Ionic conductivity ($S/cm^2/mol$)
LiOH/1M	SWNT	25.0	38.69[4]
NaOH/1M	SWNT	19.7	50.11[4]
KOH/1M	SWNT	17.4	73.5[4]

References

1. M. Rubeš, P. Nachtigall, J. Vondrášek and O. Bludský, *J. Phys. Chem. C*, 2009, **113**, 8412-8419.
2. W.-J. Fan and R.-Q. Zhang, *J. Cluster Sci.*, 2015, **26**, 361-373.
3. X. Li, J. Rong and B. Wei, *ACS Nano*, 2010, **4**, 6039-6049.
4. J. G. Speight, *Lange's handbook of chemistry*, McGraw-Hill, New York, 16th edn., 2017.