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

Supporting Information

Theoretical prediction on T-graphene as promising Alkali-ion batteries anode offering ultrahigh capacity

Junping Hu,[a*] Yu Liu,[a] Ning Liu,[a] Jianwen Li,[a] Chuying Ouyang[b*]

a. School of Science, Nanchang Institute of Technology, Nanchang 330099, China

b. Department of Physics, Laboratory of Computational Materials Physics, Jiangxi Normal University, Nanchang 330022, China.

*Corresponding author's E-mail: jphu@nit.edu.cn (Junping Hu)

*Corresponding author's E-mail: cyouyang@jxnu.edu.cn (Chuying Ouyang)

1. Dispersion energies of Li (Na or K) adsorption on T-graphene tested with different vdW methods.

The tests are performed with PBE functional, because some of the vdW methods are not supported by LDA functional. Seven types of vdW methods are considered. Before Li (Na, or K) adsorption, the dispersion energies range from -2.58 eV (D3bj) to -1.13 eV (D3). The dispersion energies are originated from the vdW interaction between C atoms in the T-graphene layer.

For the Li/Na/K adsorption, when one Li (Na or K) is put on the T-graphene surface and the dispersion energies become lower for all types of vdW methods tested, showing that Li-ion (Na-ion or K-ion) contributes to the dispersion energy for all tested cases. As Li-ion (Na-ion or K-ion) is fully ionized, the vdW interaction should be very close to zero. Therefore, inclusion of vdW interaction will overestimate the adsorption energy.

Furthermore, as listed in Tab. S1, for different type of vdW methods, the contribution from the Li-ion ranges from -0.29 eV (D2) to -0.03 eV (D3bj), the contribution from the Na-ion ranges from -0.27 eV (D2) to -0.07 eV (D3bj), and the contribution from the K-ion ranges from -0.49 eV (TS+SCS) to -0.09 eV (D3bj), which makes it very difficult to decide which vdW method should be used in the calculations. Therefore, the vdW interaction is not included in this study.

Tab. S1. The dispersion energies (E_{dis}) of Li (Na or K) adsorption on T-graphene obtained from different vdW methods.

vdW type		no-vdW	D2[a]	D3[b]	D3bj[c]	dDsC[d]	TS[e]	TSHI[f]	TS+SCS[g]
T-graphene	Total energy (eV)	**-313.49**	**-314.67**	**-314.62**	**-316.07**	**-315.60**	**-315.27**	**-315.19**	**-315.50**
	E_{dis} (eV)	**0.00**	**-1.17**	**-1.13**	**-2.58**	**-2.11**	**-1.78**	**-1.70**	**-2.00**
1 Li on T-graphene	Total energy (eV)	**-316.37**	**-317.83**	**-317.53**	**-319.00**	**-319.00**	**-318.38**	**-318.14**	**-318.71**
	E_{dis} (eV)	**0.00**	**-1.46**	**-1.16**	**-2.63**	**-2.63**	**-2.01**	**-1.77**	**-2.34**
E_{dis} contributed by Li		**0.00**	**-0.29**	**-0.03**	**-0.06**	**-0.53**	**-0.23**	**-0.07**	**-0.34**
Li adsorption height (Å)		**1.427**	**1.446**	**1.422**	**1.416**	**1.436**	**1.389**	**1.435**	**1.358**
1 Na on T-graphene	Total energy (eV)	**-315.78**	**-317.22**	**-316.98**	**-318.43**	**-317.97**	**-317.81**	**-317.65**	**-318.12**
	E_{dis} (eV)	**0.00**	**-1.44**	**-1.20**	**-2.66**	**-2.20**	**-2.03**	**-1.87**	**-2.34**
E_{dis} contributed by Na		**0.00**	**-0.27**	**-0.07**	**-0.08**	**-0.09**	**-0.25**	**-0.17**	**-0.33**
Na adsorption height (Å)		**1.916**	**1.924**	**1.91**	**1.903**	**1.909**	**1.906**	**1.907**	**1.912**
1 K on T-graphene	Total energy (eV)	**-316.09**	**-317.46**	**-317.31**	**-318.75**	**-318.28**	**-318.33**	**-318.11**	**-318.58**
	E_{dis} (eV)	**0.00**	**-1.37**	**-1.23**	**-2.66**	**-2.19**	**-2.24**	**-2.02**	**-2.49**
E_{dis} contributed by K		**0.00**	**-0.20**	**-0.10**	**-0.09**	**-0.09**	**-0.46**	**-0.33**	**-0.49**
K adsorption height (Å)		**2.374**	**2.364**	**2.376**	**2.366**	**2.368**	**2.45**	**2.324**	**2.342**

a) DFT-D2 method of Grimme [1]

b) DFT-D3 method of Grimme with zero damping [2]

c) DFT-D3 method with Becke-Jonson damping [3]

d) DFT-dDsC dispersion correction [4]

e) Tkatchenko-Scheffler method [5]

f) Tkatchenko-Scheffler method with iterative Hirshfeld partitioning [6].

g) Tkatchenko-Scheffler method with self-consistent screening [7]

References

[1] S. Grimme, Semiempirical GGA-type density functional constructed with a long-range dispersion correction. J. Comp. Chem. 27 (2006) 1787-1799.

[2] S. Grimme, J. Antony, S. Ehrlich, S. Krieg, A consistent and accurate ab initio parameterization of density functional dispersion correction for the 94 elements H-Pu. J. Chem. Phys. 132 (2010) 154104.

[3] S. Grimme, S. Ehrlich, L. Goerigk, Effect of the damping function in dispersion corrected density functional theory. J. Comp. Chem. 32 (2011) 1456-1465.

[4] S. N. Steinmann, C. Corminboeuf, A generalized-gradient approximation exchange hole model for dispersion coefficients. J. Chem. Phys. 134 (2011) 044117.

[5] A. Tkatchenko, M. Scheffler, Accurate molecular van der Waals interactions from ground-state electron density and free-atom reference data. Phys. Rev. Lett. 102 (2009) 073005.

[6] P. Bultinck, C. V. Alsenoy, P. W. Ayers, R. Carbó Dorca, Critical analysis and extension of the Hirshfeld atoms in molecules. J. Chem. Phys. 126 (2007) 144111.

[7] A. Tkatchenko, R. A. Di Stasio, R. Car, M. Scheffler, Accurate and efficient method for many-body van der Waals interactions. Phys. Rev. Lett. 108 (2012) 236402.