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

Supporting Information

Exploring novel two-dimensional metallic Y_4C_3 sheet applied in anode material for sodium-ion battery

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Structure Prediction Details

The particle swarm optimization (PSO) method within the evolutionary algorithm as implemented in the CALYPSO code[1, 2] was employed to find the lowest energy structures of 2D Y-C system. The unit cells containing 1, 2, 3, and 4 formula units (f.u.) were considered. In the first step, random structures with certain symmetry are constructed in which atomic coordinates are generated by the 2D crystallographic symmetry operations. Local optimizations using the VASP code[3] were done with the conjugate gradients method and stopped when Gibbs free energy changes became smaller than 1×10^{-5} eV per cell. After processing the first generation structures, 60% of them with lower Gibbs free energies are selected to construct the next generation structures by PSO. 40% of the structures in the new generation are randomly generated. A structure fingerprinting technique of bond characterization matrix is applied to the generated structures, so that identical structures are strictly forbidden. These procedures significantly enhance the diversity of the structures, which is crucial for structural global search efficiency. In most cases, structural searching simulations for each calculation were stopped after generating 1000 ~ 1200 structures (e.g., about 20 ~ 35 generations).

Supporting Figures:

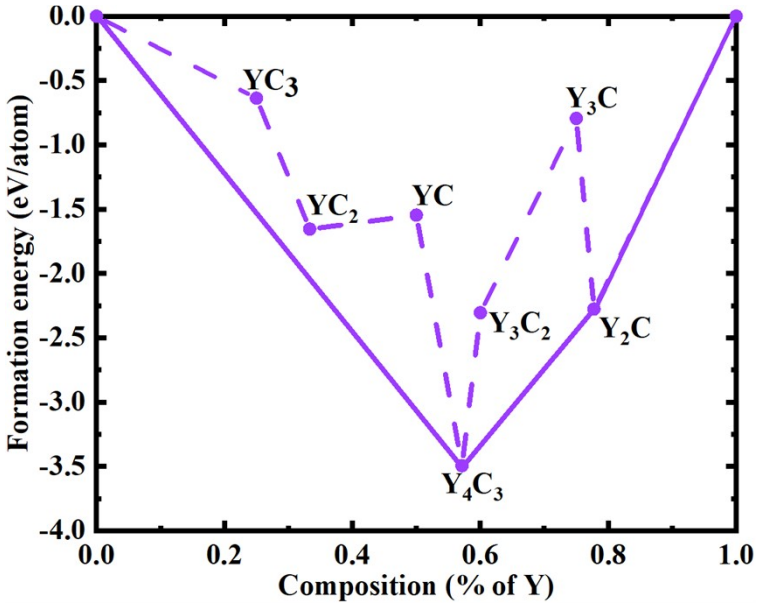

Figure S1. The relative stability of various 2D Y_mC_n structures with respect to element Y and graphene at 0K. The thermodynamically stable compositions are corresponding to the data points located on the convex hull.

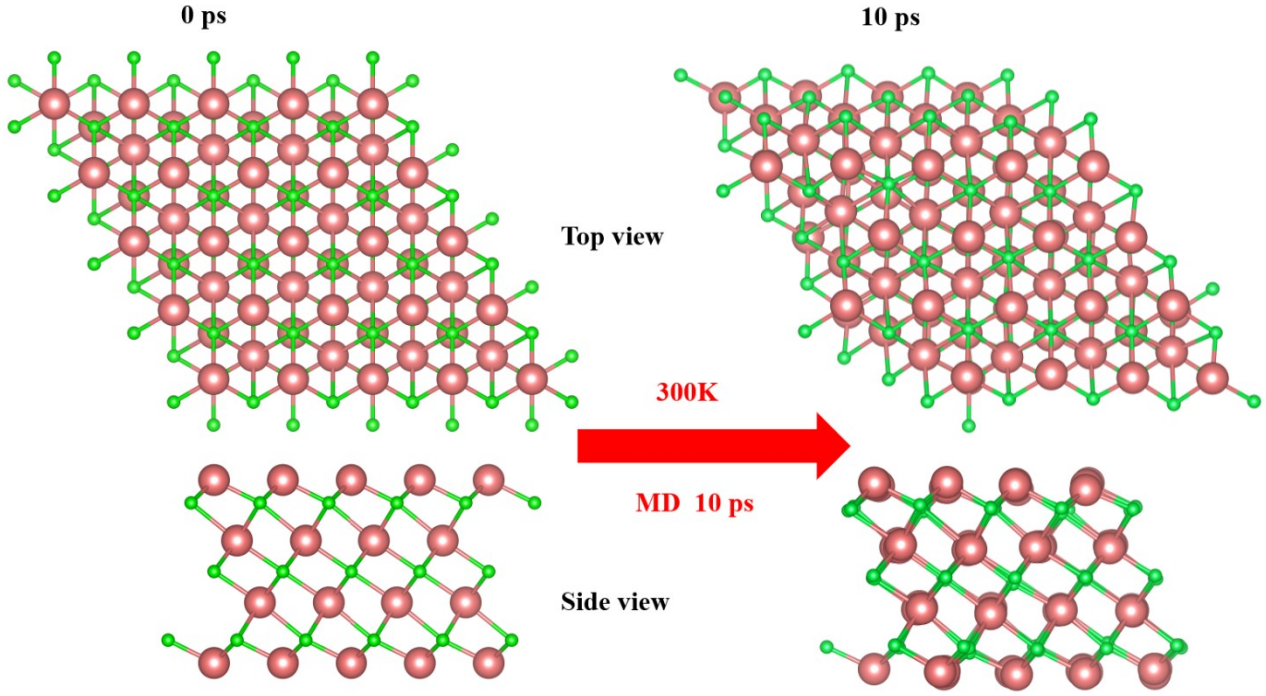

Figure S2. Snapshots of the Y_4C_3 *P*-3*m*1 equilibrium structure at the end of 10 ps by AIMD simulation at the temperature of 300 K.

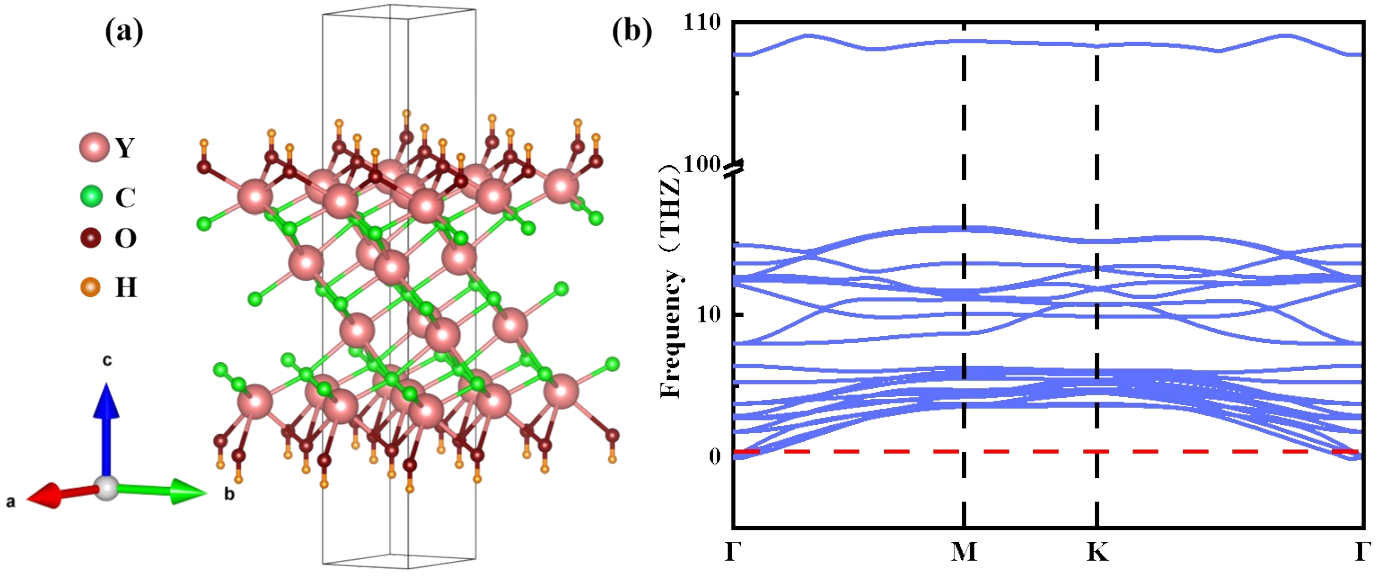

Figure S3. (a) The energy stable configuration of functionalized $Y_4C_3(OH)_2$, the pink balls are Y atoms, the green balls are C atoms, the brown balls are O atoms and the orange balls are the H atom. (b) Phonon dispersion curves of the $Y_4C_3(OH)_2$ sheet.

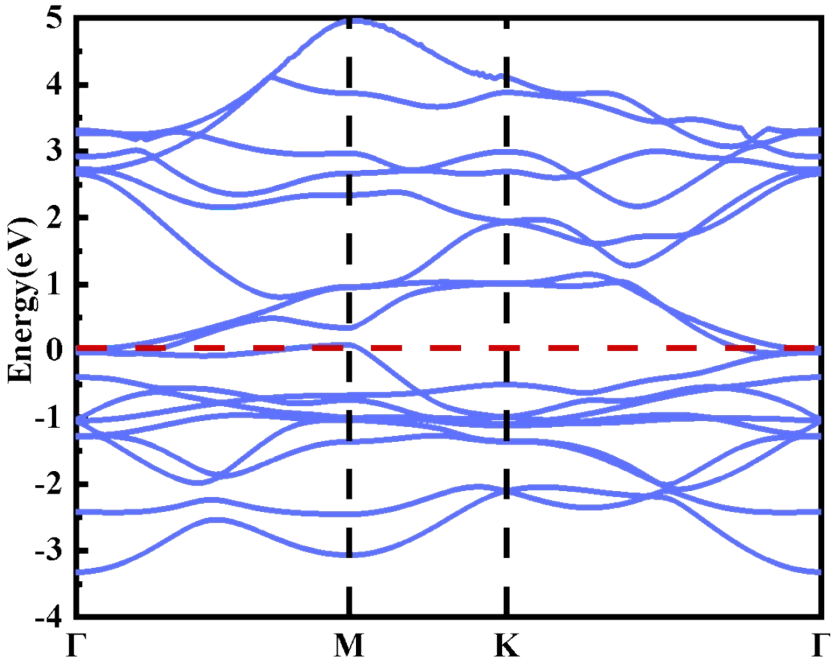

Figure S4. Energy band diagram of Y_4C_3 *P*-3*m*1 structure computed using PBE functional.

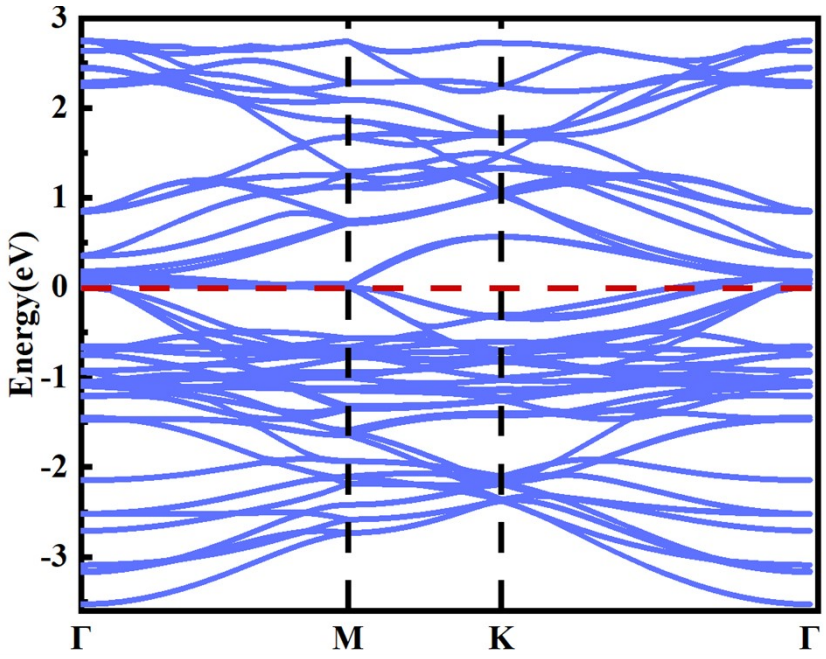

Figure S5. Energy band diagram of the sodiated-Y_4C_3 configurations (taken $Y_4C_3Na_1$ as example) computed using PBE functional.

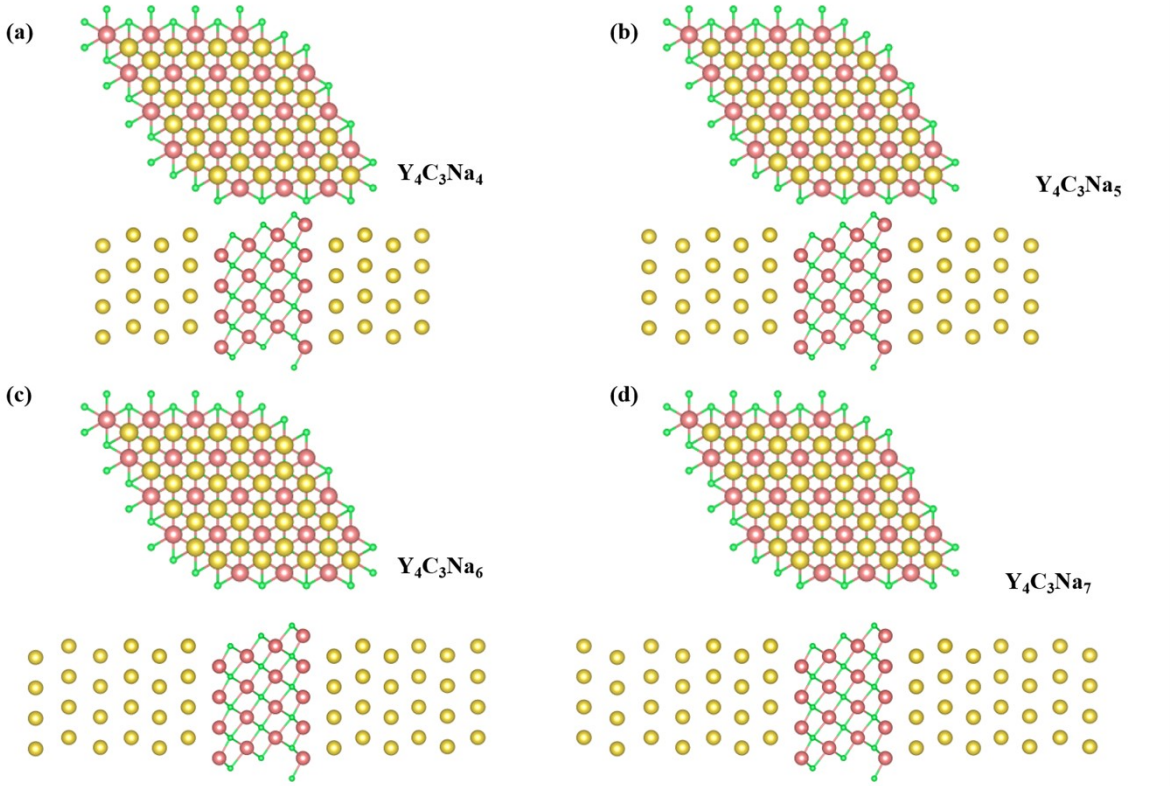

Figure S6. (a)-(d) Selected and shown some typical adsorption configurations with different Na concentrations in $Y_4C_3Na_n$.

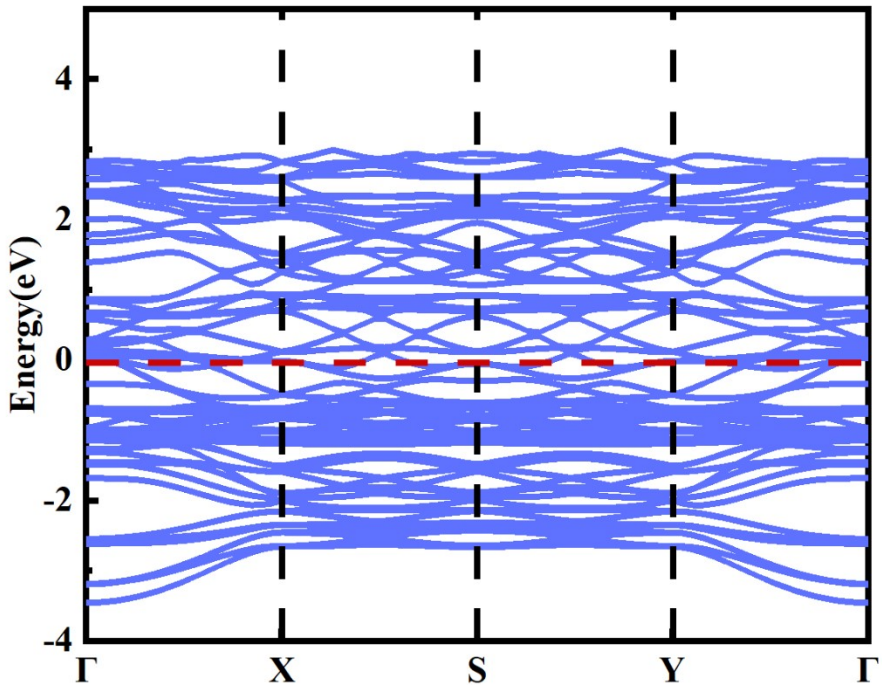

Figure S7. Electronic band structure of Y_4C_3K sheet.

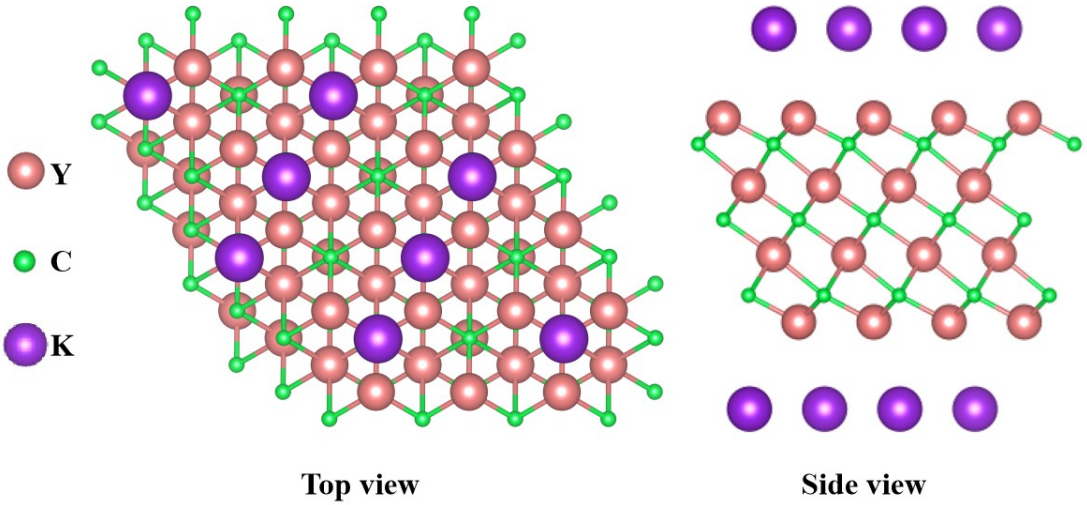

Figure S8. The structure of Y_4C_3 adsorbed a layer of K ions.

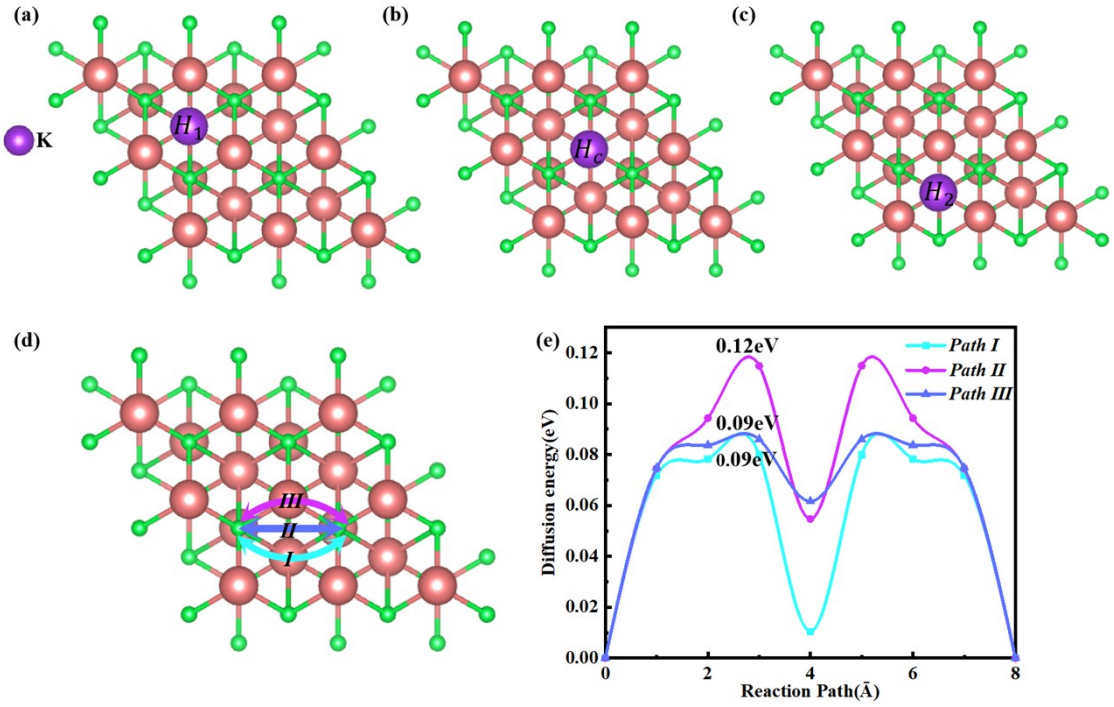

Figure S9. Top views of the geometric structures for K atom adsorption on the (a) H_1, (b) H_C, and (c) H_2 sites of the Y_4C_3 sheet. The purple ball represents the metal K atom. (d) Migration paths and (e) diffusion barrier profiles for K on the surface of the Y_4C_3 sheet. I, II and III labels $H_C \rightarrow H_2 \rightarrow H_C$, $H_C \rightarrow H_C$ and $H_C \rightarrow H_1 \rightarrow H_C$ paths, respectively.

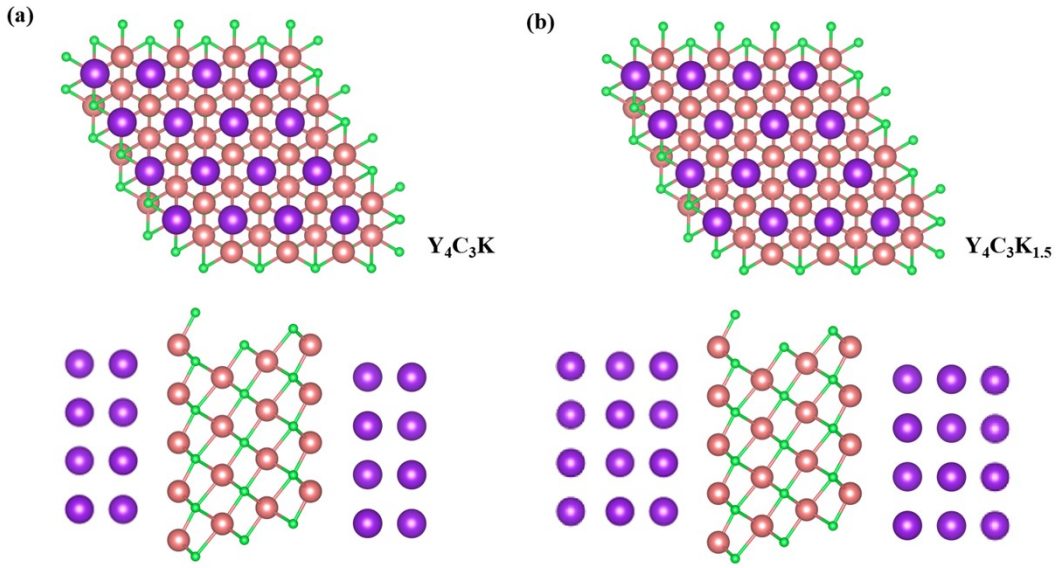

Figure S10**.** Adsorbed (a) two layers of K on the Y_4C_3 sheet and (b) three layers of K on the Y_4C_3 sheet.

References

1. Y. Wang, J. Lv, L. Zhu and Y. Ma, *Comput. Phys. Commun.*, 2012, **183**, 2063-2070.
2. Y. Wang, J. Lv, L. Zhu and Y. Ma, *Phys. Rev. B.*, 2010, **82**.
3. G. Kresse and J. Furthmüller, *Phys. Rev. B.*, 1996, **54**, 11169-11186.