

碱金属修饰的多孔石墨烯的储氢性能*

元丽华^{1)†} 巩纪军¹⁾ 王道斌¹⁾ 张材荣¹⁾ 张梅玲¹⁾³⁾ 苏俊燕¹⁾ 康龙²⁾

1) (兰州理工大学理学院, 兰州 730050)

2) (兰州理工大学材料科学与工程学院, 兰州 730050)

3) (兰州大学核科学与技术学院, 兰州 730000)

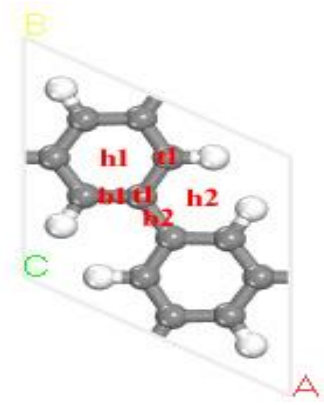


图 S1 PG 单胞的 6 个不同的吸附位置(完整 C 环中心 h1、CH 环中心 h2、C1-C2 桥位 b1、C2-C2 桥位 b2、C1 原子顶位 t1 以及 C2 原子顶位 t2)

Fig. S1. Different initial adsorption sites of a alkaline metal atom on PG layer: Hollow center of the C hexagon (h1), hollow center of the C-H hexagon near the porosity (h2), the bridge sites of C—C bond (b1, b2), and the top sites of C atom (t1, t2).

表 S1 碱金属原子吸附在 PG 的初始位置、弛豫后的位置及结合能 ΔE

Table S1. Initial adsorption sites and optimized sites of a alkaline metal atom on PG layer, and the binding energy ΔE .

	Initial sites					
	h1	h2	b1	b2	t1	t2
Optimized sites of Li	h1	h2	h1	h2	h1	h2
$\Delta E_{\text{Li}}/\text{eV}$	-1.654	-1.489	-1.651	-1.489	-1.651	-1.489
Optimized sites of Na	h1	h2	h1	h2	h1	h1
$\Delta E_{\text{Na}}/\text{eV}$	-0.877	-0.879	-0.883	-0.879	-0.881	-0.878
Optimized sites of K	h1	b2	h1	b2	h1	h1
$\Delta E_{\text{K}}/\text{eV}$	-1.037	-0.955	-1.039	-0.956	-1.048	-1.043

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† 通信作者. E-mail: yuanlh@lut.edu.cn

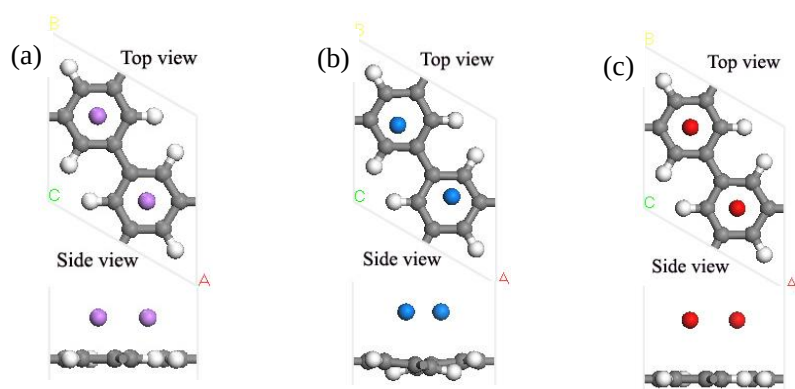


图 S2 PG 单侧吸附 2 个碱金属原子弛豫后的几何结构(紫色球代表 Li 原子, 蓝色球代表 Na 原子, 红色球代表 K 原子, 灰色球代表 C 原子及白色球代表 H 原子) (a) 2Li-PG; (b) 2Na-PG; (c) 2K-PG

Fig. S2. Optimized geometry structure of two alkaline metal atoms decorated PG: (a) 2Li-PG; (b) 2 Na-PG; (c) 2K-PG. The purple, blue, red, gray and white balls in this and following figures denote Li, Na, K, C and H atoms, respectively.

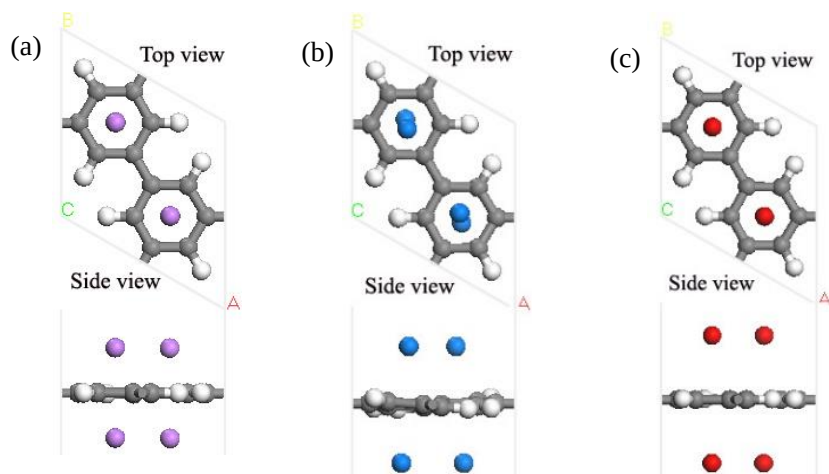


图 S3 PG 两侧吸附 4 个碱金属原子弛豫后的几何结构 (a) 4Li-PG; (b) 4Na-PG; (c) 4K-PG

Fig. S3. Optimized geometry structure of four alkaline metal atoms adsorbed on double sides of PG unit cell: (a) 4Li-PG; (b) 4Na-PG; (c) 4K-PG.

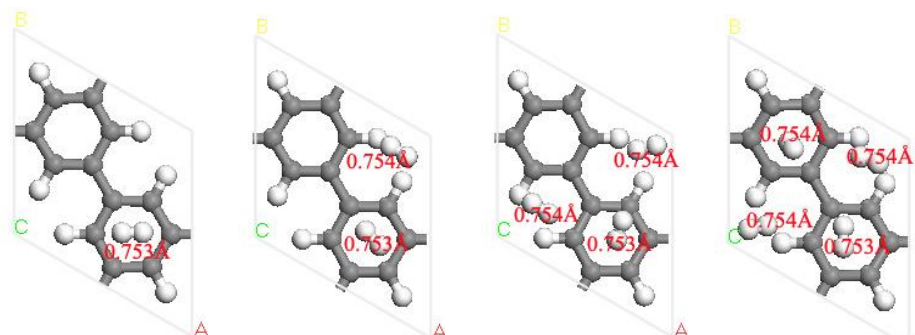


图 S4 PG 单侧吸附氢气分子弛豫后的几何结构(红色字体表示 H—H 键长)
Fig. S4. Optimized geometry structure of the PG with H_2 molecules adsorption. Red digits represent the corresponding bond length of H—H.

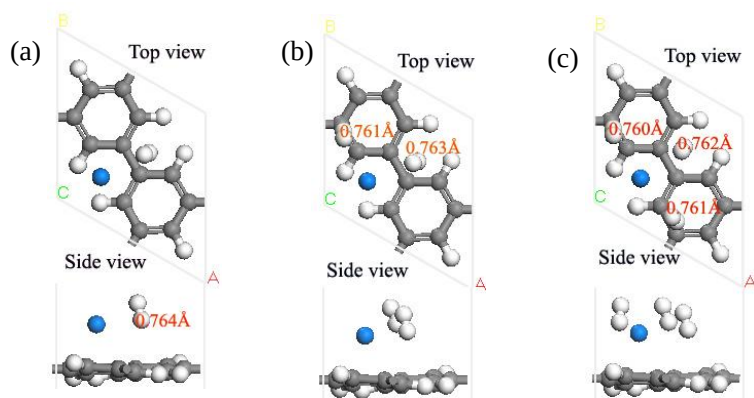


图 S5 Na-PG 吸附 1—3 个 H_2 分子优化后的几何结构(Na 原子位于 PG 表面 h2 位置;
红色数字表示 H—H 键长)

Fig. S5. Optimized geometry structure of the Na-decorated PG with 1–3 H_2 molecules adsorption, the Na atom adsorbs on the h2 site of the PG. Red digits represent the corresponding bond length of H—H.

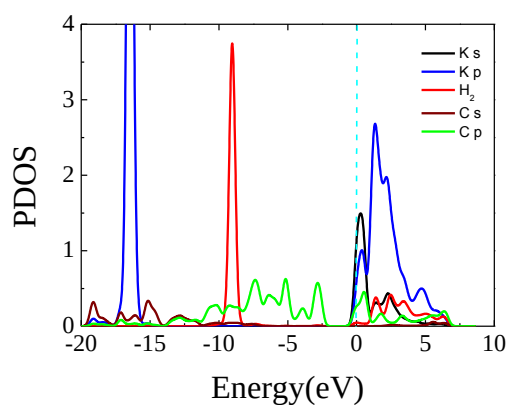


图 S6 K-PG 吸附 1 个 H_2 分子的分波态密度图

Fig. S6. Partial density of states of a H_2 molecule on K-PG system.

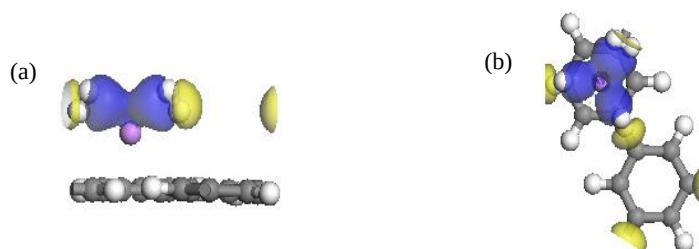


图 S7 Li-PG 单侧吸附 2 个(a)和 3 个(b) H_2 分子的差分电荷密度图(蓝色和黄色区域分别代表电荷聚集和减少的区域; 电荷密度等值面是 $0.01 \text{ e}/\text{\AA}^3$)

Fig. S7. Charge density differences of n H_2 adsorbed on Li-PG system for (a) $n=2$ and (b) $n=3$. The blue and yellow colors represent electron accumulation and depletion regions. The isovalue is taken to be $0.01 \text{ e}/\text{\AA}^3$.

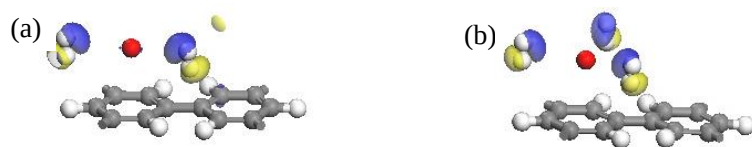


图 S8 K-PG 单侧吸附 2 个 (a) 和 3 个 (b) H_2 分子的差分电荷密度图(蓝色和黄色区域分别代表电荷聚集和减少的区域; 电荷密度等值面是 $0.01 \text{ e}/\text{\AA}^3$)

Fig. S8. Charge density differences of $n \text{ H}_2$ adsorbed on K-PG system for (a) $n=2$ and (b) $n=3$. The blue and yellow colors represent electron accumulation and depletion regions. The isovalue is taken to be $0.01 \text{ e}/\text{\AA}^3$.

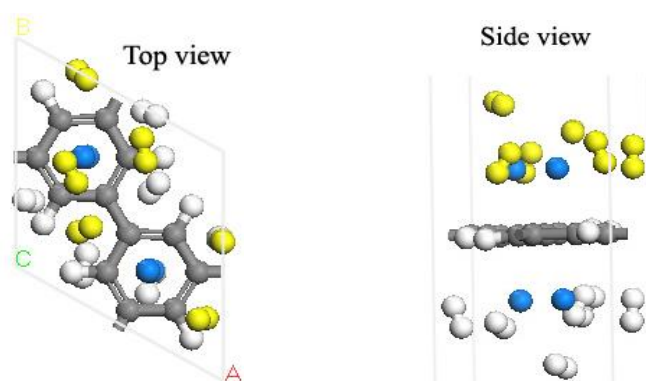


图 S9 Na-PG 结构两侧吸附 12 个 H_2 分子弛豫后的几何结构图(黄色的球代表上层吸附的 H_2 分子, 白色的球代表下层吸附的 H_2 分子)

Fig. S9. Optimized geometry structure of the Na-decorated PG with 12 H_2 adsorption. The yellow and white balls denote the H_2 molecules adsorbed on the upper and under PG layer, respectively.

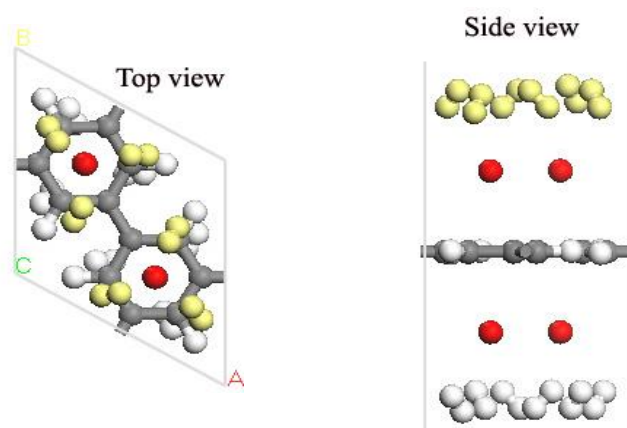


图 S10 12 个 H_2 分子吸附在 K 原子修饰的 PG 两侧优化后的几何结构图

Fig. S10. Optimized geometry structure of the K-decorated PG with 12 H_2 adsorption.

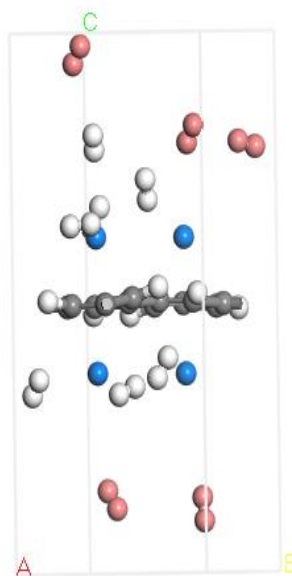


图 S11 在 300 K 的条件下， $12\text{H}_2\text{-Na-PG}$ 结构经过 AIMD 模拟后的平衡结构
Fig. S11. The equilibrated structure of the $12\text{H}_2\text{-Li-PG}$ in AIMD simulations at 300 K. The

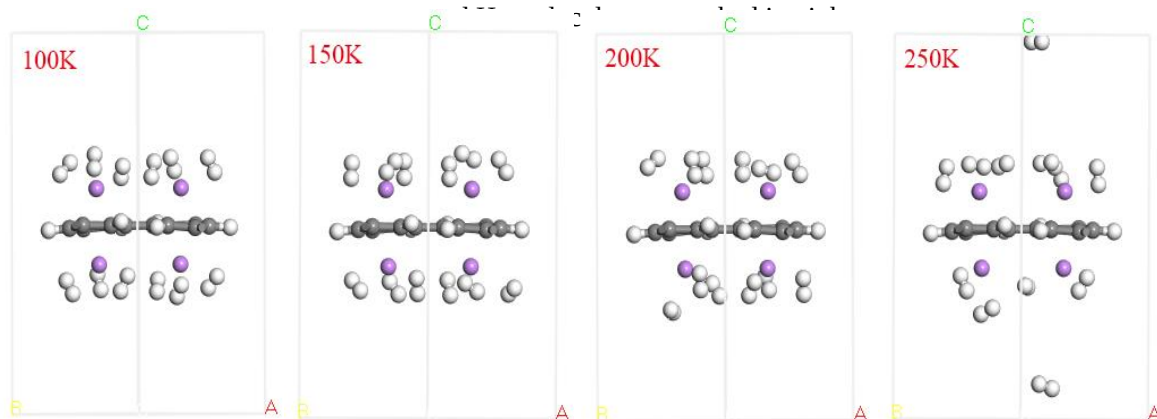


图 S12 $12\text{H}_2\text{/Li-PG}$ 在不同温度下经过 AIMD 模拟后的平衡结构
Fig. S12. Equilibrated structure of the $12\text{H}_2\text{/Li-PG}$ after ab initio MD simulations at different temperatures.