Supporting Information: Reversible Hydrogen Storage in Metal-Decorated Honeycomb Borophene Oxide

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Free energy plots as a function of time during Born-Oppenheimer Molecular dynamics simulations of H_2 saturated, Li-decorated 2D honeycomb B_2O (Figure S1); different initial and final configurations for a single H_2 adsorption on 2D B_2O (Figure S2); different configurations of a single Li, Na and K atom on 2D B_2O and their respective energy differences (Figure S3); Bader charge analysis of single Li, Na and K decorated 2D B_2O (Table S1); different configurations for a single H_2 adsorption on Li, Na and K-decorated 2D B_2O and their respective adsorption energies (Figure S4).

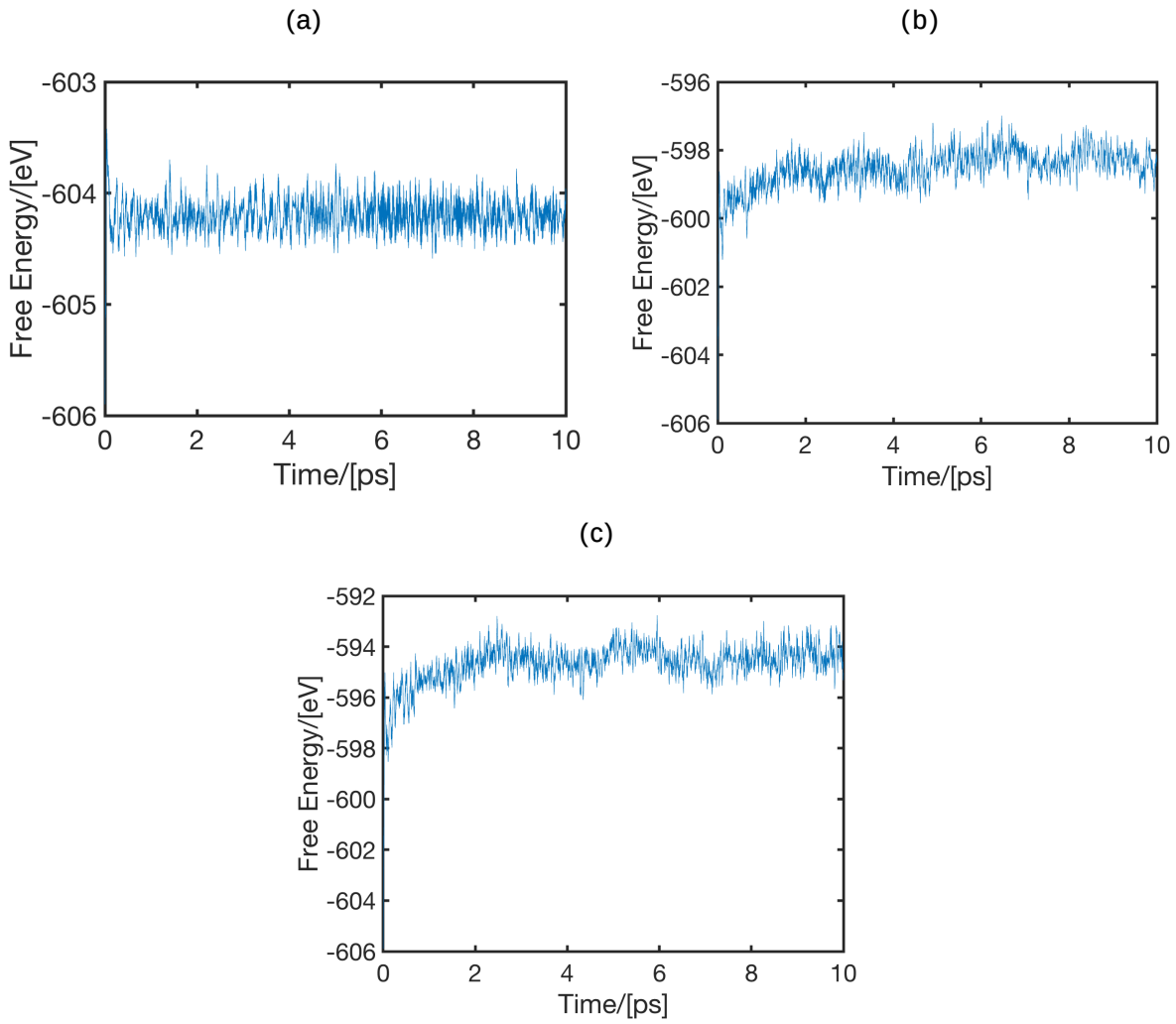

Figure S1: Plot of free energy versus time during Born-Oppenheimer Molecular Dynamics simulations at three different temperatures of (a) 100 K, (b) 300 K, and (c) 500 K. A H_2 saturated, Li decorated 2D B_2O structure is simulated with a time step of 1 fs.

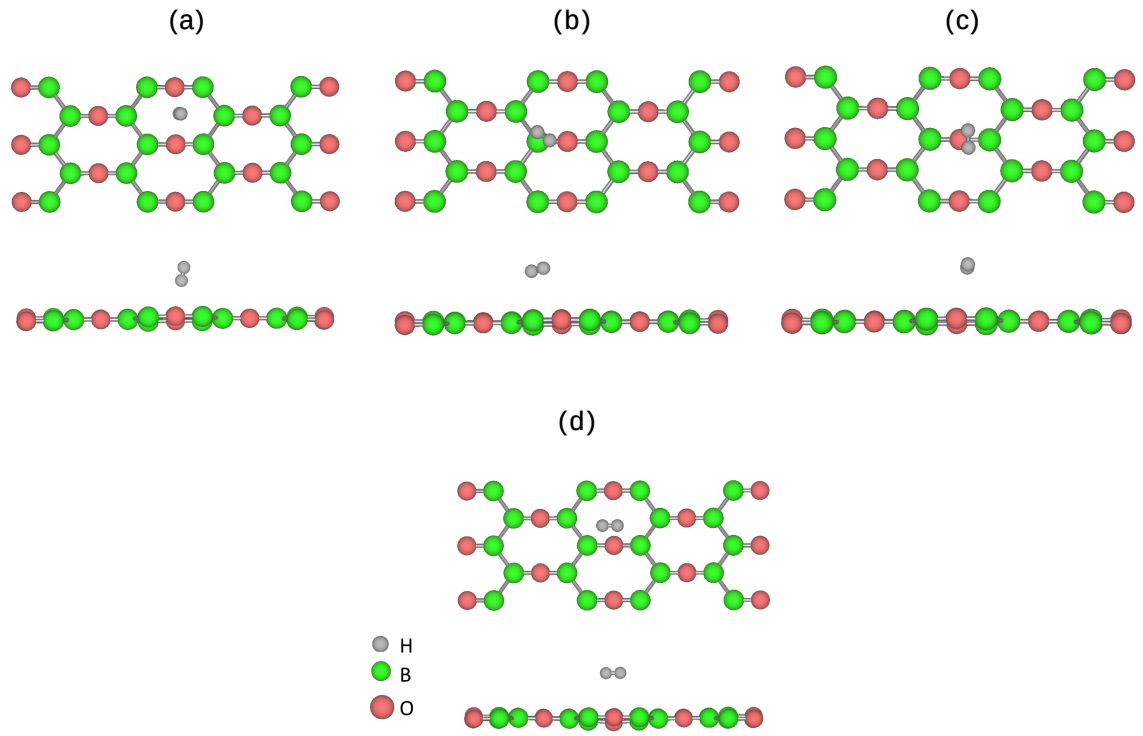

Figure S2: Different initial configurations tested for H_2 adsorption on 2D B2O are shown in (a)-(c). In (a) H_2 is placed in the hollow site of B_2O while in (b) and (c) it is placed on the B and O atom, respectively. After relaxing the atomic positions and lattice parameters with a force convergence criterion of 10 meV/Å, configuration (d) is found for all the initial configurations. The same structure is found after further relaxation with a force convergence criterion of 1 meV/Å. Based on the energy of the final relaxed structure (d), the H_2 binding energy is found as -0.10 eV / H_2.

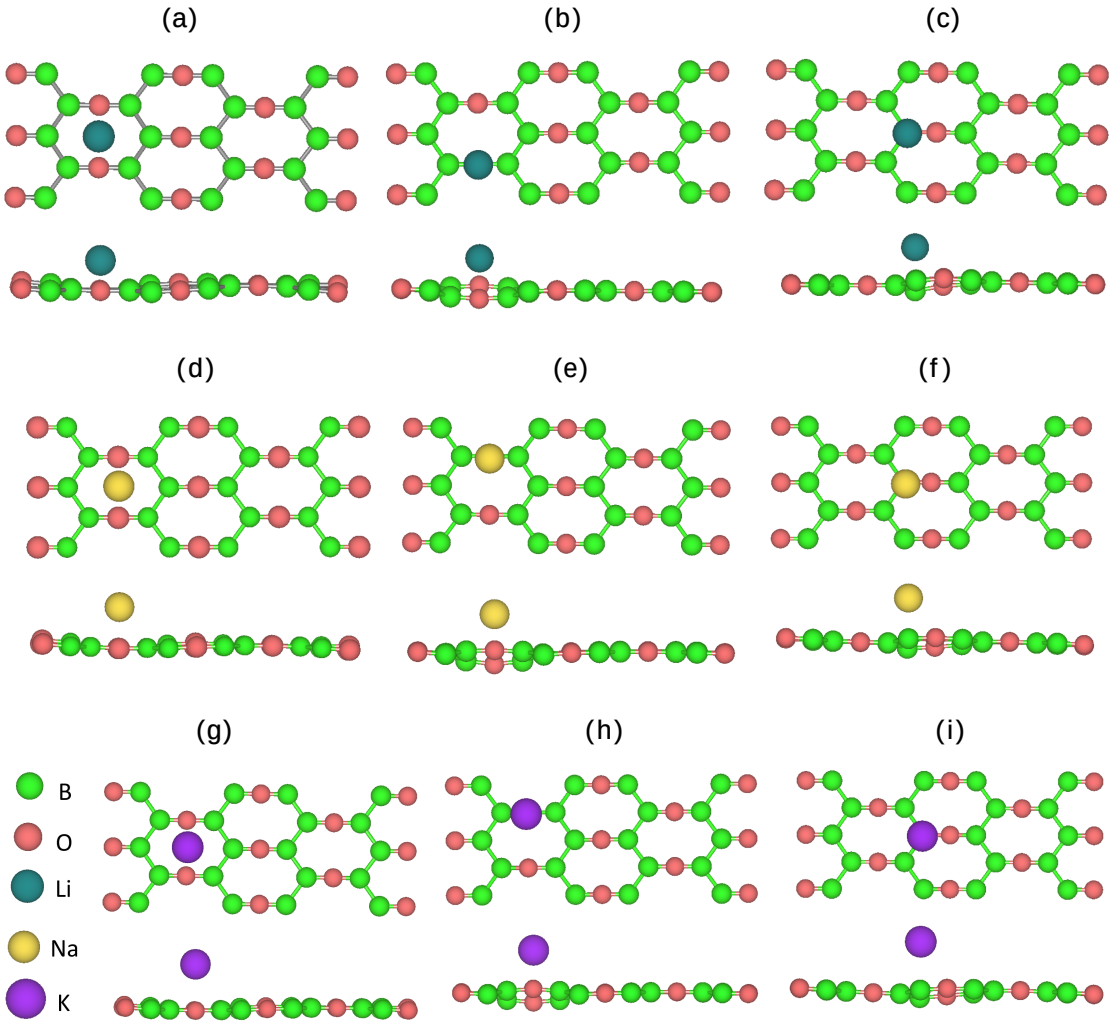

Figure S3: Top and side views of (a-c) Li (d-f) Na, and (g-i) K decorated honeycomb B_2O structures. Different configurations are investigated for each metal atom on the 2D substrate. In structures (a), (d), and (g) the metal atoms are placed on the hollow site, in structures (b), (e), and (h) the metal atoms are placed on top of an O atom, and in structures (c), (f), and (i) the metal atoms are placed on top of a B atom. The hollow site proved to be the most energetically stable. For the Li atom, the energy of structures (b) and (c) differed by 0.66 and 0.95 eV from structure (a), respectively. For the Na atom, the energy differences between structures (e) and (f) from structure (d) are 0.29 and 0.49 eV, respectively. For the K atom, the energy differences between structures (h) and (i) from structure (g) are 0.13 and 0.39 eV, respectively.

Table S1: Average Bader net atomic charges of different atoms in metal decorated (Li, Na and K) 2D honeycomb B_2O. The plus sign indicates a gain of electronic charge and a minus sign indicates a loss of electronic charge with respect to the atomic charge of respective elements (net charge 0). The Bader charges are obtained using the code of Henkelman group (J. Phys.: Condens. Matter 21, 084204 (2009)).

Average Bader charge gain/loss of atoms					
	B [e]	O [e]	Li [e]	Na [e]	K [e]
$LiB_{16}O_8$	-0.726	+1.562	-0.887	-	-
$NaB_{16}O_8$	-0.725	+1.553	-	-0.832	-
$KB_{16}O_8$	-0.725	+1.551	-	-	-0.811