Supporting Information

A First Principle Framework for Total Charging Energies in Electrocatalytic Materials and Charge-Responsive Molecular Binding at Gas-Surface Interfaces

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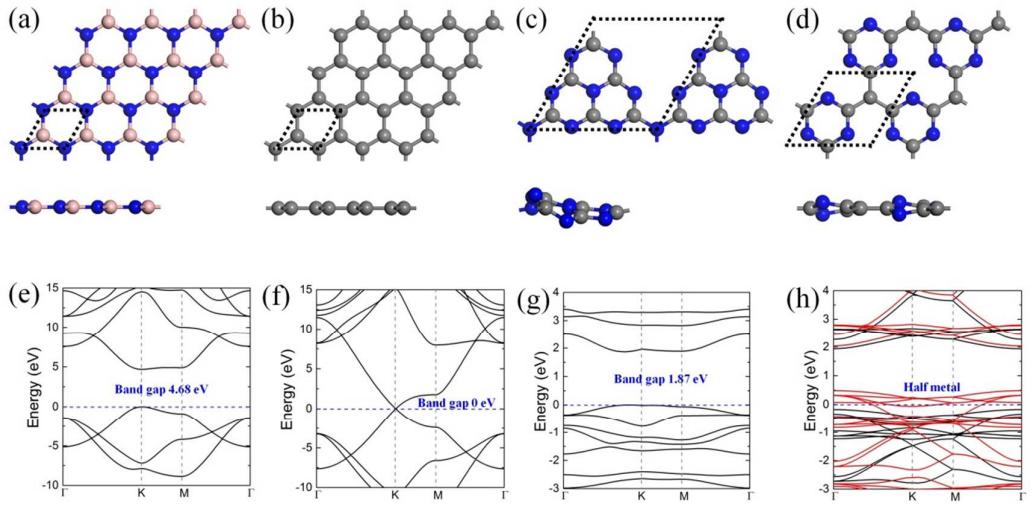

Figure S1. Top (upper) and side (lower) views of (a) a (4×4) *h*-BN supercell, (b) a (4×4) graphene supercell, (c) a (2×1) buckle g-C_3N_4 supercell, and (d) a (2×2) reconstructed g-C_4N_3 supercell. The light magenta, blue and grey balls represent B, N and C atoms, respectively, and the unit cells of each supercell are indicated by black dot lines. The calculated band structures of (e) a (1×1) *h*-BN, (f) a (1×1) graphene, (g) a (1×1) buckled g-C_3N_4, and (h) a (2×2) reconstructed g-C_4N_3. The blue dashed line denotes the Fermi level. The red and black lines in (h) denote the spin-up and spin-down states, respectively.

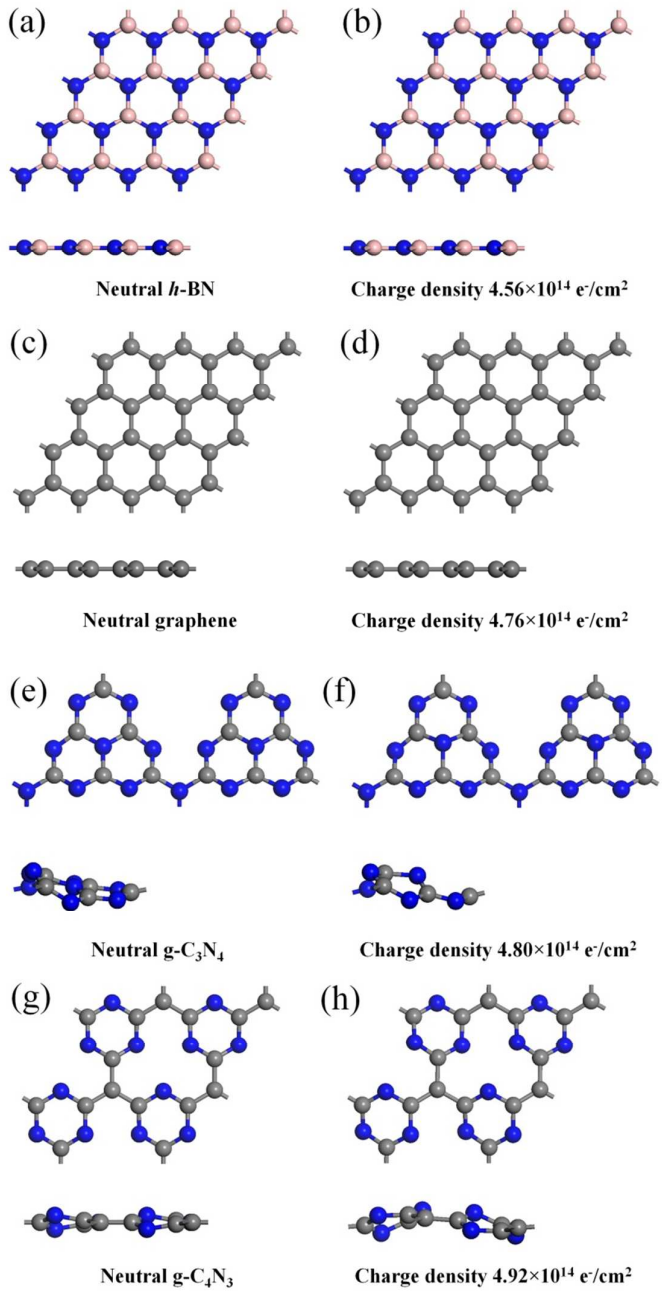

Figure S2. The optimized geometries of the (a) neutral and the (b) charged *h*-BN, the (c) neutral and the (d) charged graphene, the (e) neutral and the (f) charged g-C_3N_4, and the (g) neutral and the (h) charged g-C_4N_3. The charge densities are listed in the figures.

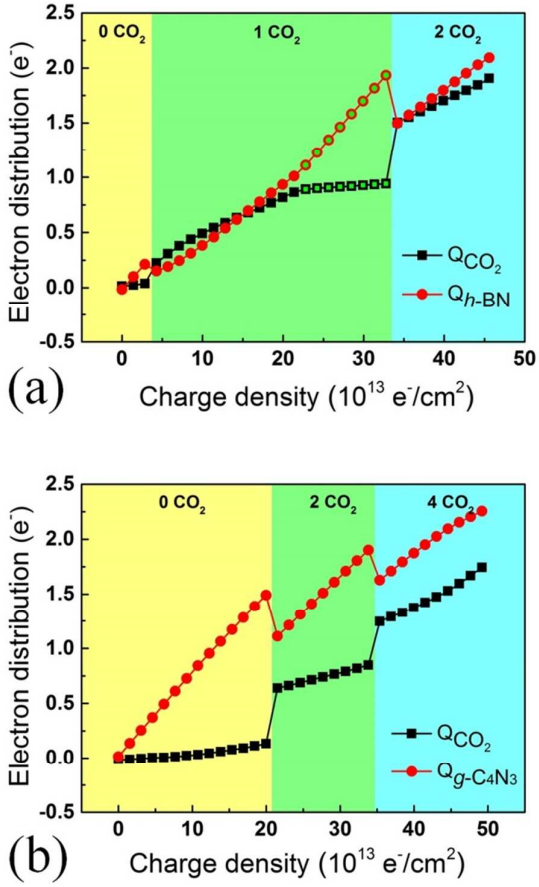

Figure S3. The electron distributions of CO_2 molecules on (a) *h*-BN and (b) g-C_4N_3 as a function of charge density. Q_{CO_2}, Q_{h-BN} and $Q_{g-C_4N_3}$ indicate the number of excess electrons of CO_2, *h*-BN and g-C_4N_3, respectively. The yellow, green and cyan regions in (a) denote 0, 1 and 2 CO_2 chemisorption. The yellow, green and cyan regions in (b) denote 0, 2 and 4 CO_2 chemisorption.