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First-principles study of dihydrogen interaction of porphyrin-like nitrogen-doped graphenes WOON IH CHOI, National Renewable Energy Laboratory, SEUNG-HOON JHI, Department of Physics, Pohang University of Science and Technology, YONG-HYUN KIM, National Renewable Energy Laboratory —
We have studied electronic structure and dihydrogen (H_2) binding characteristics of porphyrin-like nitrogen-doped graphenes (PNGs) by performing first-principles total energy calculations based on the density functional theory. The stability of PNGs and the H_2 binding ability of the PNG metal centers (Ca and $3d$ transition metals from Sc to Zn) have been analyzed within the generalized gradient approximation. We have found that Ca, Sc, Ti, Co, and Ni can be incorporated relatively easily into the nitrogen-doped graphenes, while V, Fe, Cu, and Zn are less likely to be. The PNGs can be used for active building blocks of hydrogen storing metal-organic frameworks. Due to the unique crystal field splitting of the planar PNGs, approaching dihydrogen exclusively interacts with the dz^2 orbitals of the core metals. We also found that intra-d-orbital charge transfer plays a key role in the dihydrogen binding. Finally, we will discuss how such dihydrogen binding can be modified by external strain.

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