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Anew computationalapproach formthe study of atomic hydrogen adsorption and molecular hydrogen formation on graphene and PAHs

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We used a new computational approach to calculate the energetics of adsorption of H atoms and the dynamics of formation of H_2 on graphene/PAHs through the Eley-Rideal process. A simple model is developed to quantify the contribution of tunneling of hydrogen to its diffusion at various temperatures. Although specific calculations have appeared previously, the new approach allows handling in the same framework of adsorption on both physisorption and chemisorption sites, the diffusion of hydrogen between the two, and the formation of molecules through the Eley-Rideal and Langmuir-Hinshelwood processes.

The semi empirical bond order ReaxFF potential, originally developed for hydrocarbons, is trained to reproduce DFT generated adsorption potential energy of hydrogen on graphene as well as Eley-Rideal potential energy formation of H_2 on graphene. We have employed GARFfield software, a genetic algorithm reactive force field optimizer suitable for training of the ReaxFF force field (a reactive force field). The GARFfield algorithm has proven to be more efficient than the traditional optimization programs, such as conjugate gradient or steepest descent. In principle, a CH ReaxFF potential, trained against a rich DFT data base, has the accuracy and transferability of the corresponding first- principles calculations without its limitations. In this study, the CHO ReaxFF potential is trained against the first-principles data generated in the chemisorption and physisorption regions of the adsorption of hydrogen as well as in molecular hydrogen formation on graphene.

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