Permeation and Recombination of Hydrogen Chemisorbed on Graphene: Insights from Computations

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Recent discoveries that graphene permeates protons[1] and hydrogen atoms[2] have opened up prospects of new applications in hydrogen technologies, energy storage and conversion, isotope separation, etc. In addition, a great interest has risen for uncovering the microscopic mechanisms underlying such observations. In the presentation, we will address two processes involved in hydrogenated graphene, namely, the permeation or flipping of chemisorbed hydrogen atoms through a graphene layer and the recombination of these atoms (desorption and formation of hydrogen molecules, a reaction that can occur after permeation). With the aim to provide some insight into these processes, we have carried out density functional theory computations using large molecular prototypes of graphene. Firstly, we will discuss a new mechanism for the flipping of chemisorbed hydrogen atoms[3] or protons[4] (see Fig. 1), for which the estimated activation energies are of the order of the experimental findings[1,2]. Secondly, we will report reaction paths and rate coefficients for the recombination of hydrogen and deuterium and analyze the large isotopic substitution effects[5] observed in thermal desorption measurements[6,7]. Finally, some findings about the role of Stone-Wales defects on the studied processes will be outlined.

References

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Figures

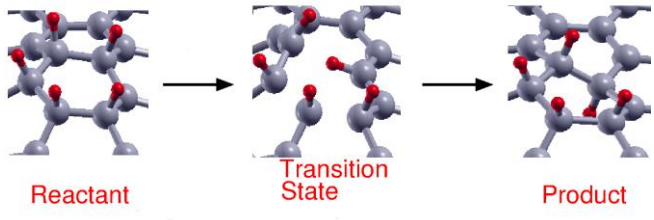

Figure 1: Geometries of the reactant, transition and product states for a hydrogen atom flipping through a highly hydrogenated carbon ring. The activation energy is about 1.5 eV.