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A cooperative multi-proton mechanism for proton permeation in graphene

It has been recently experimentally shown that the rule of impermeability of pristine graphene and some other 2D materials, to any kind of atom or molecule at room temperature is not fulfilled in the case of protons and their isotope deuterium[1,2]. These charged species permeate through the 2D material following a low barrier (~ 0.8 eV) activated process. Most of the theoretical attempts to provide with a reasonable explanation have found that permeation of the H^+ (D^+) involves large energy barriers (around 3.5 eV) and are therefore too high to explain the experimental findings[3]. In most previous models it was assumed an isolated proton permeating the 2D membrane. In this work however, we consider protonated graphene at high local coverage and explore the role played by nearby chemisorbed protons in the permeation process. By using density functional theory calculations applied to large molecular prototypes for graphene we have found[4] that when various protons are adsorbed on carbons belonging to the same hexagonal ring, permeation barrier can be lowered down to 1.0 eV, thus making feasible the permeation of protons through pristine graphene. The proposed insertion mechanism necessarily need to count with the nearby protons and it could be of relevance not only to help in the understanding of experiments from ref.[1,2], but also in many other scenarios.

References

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Figures

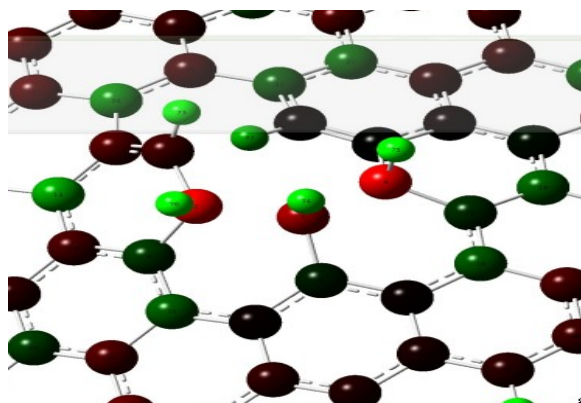


Figure 1: Transition state (TS) for proton flipping in the cooperative multiproton mechanism.