

Proton Transport Mechanisms in Graphene and Graphene Oxide: Insights from ReaxFF Molecular Dynamics Simulations

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Graphene and graphene oxide (GO) have emerged as promising materials for fuel cell applications due to their selectivity towards proton transport and exceptional mechanical and thermal stability properties. [1,2,3] In this study, we have investigated the mechanism of electrochemically driven proton permeation through graphene and GO using the ReaxFF molecular dynamics simulations. Our findings reveal two primary mechanisms for proton permeation: (i) a direct permeation through graphene rings and (ii) a flipping mechanism, where the proton first adsorbs onto the graphene surface before flipping through the graphene ring. The latter mechanism dominates in the case of both graphene and graphene oxide. Additionally, we explore the effect of temperature on proton flux, revealing that the proton conductivity increases with temperature. This enhancement in conductivity is attributed to the increased proton thermal energy and reduced graphene charge density, which weakens the interactions between the graphene and protons. Compared to graphene, GO exhibits superior proton conductivity, which is primarily due to the presence of functional groups. At 300 K, the proton conductivity of GO is twice that of graphene. Furthermore, as the concentration ratio of hydroxyl groups to epoxy groups increases, its conductivity improves from 0.031 mS/cm to a maximum value of 0.038 mS/cm. Between the hydroxyl and the epoxy functional groups, the epoxy groups significantly influence the geometry and charge density of graphene oxide, offering a potential for tuneable proton conductivity of graphene oxide.

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References

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Figures

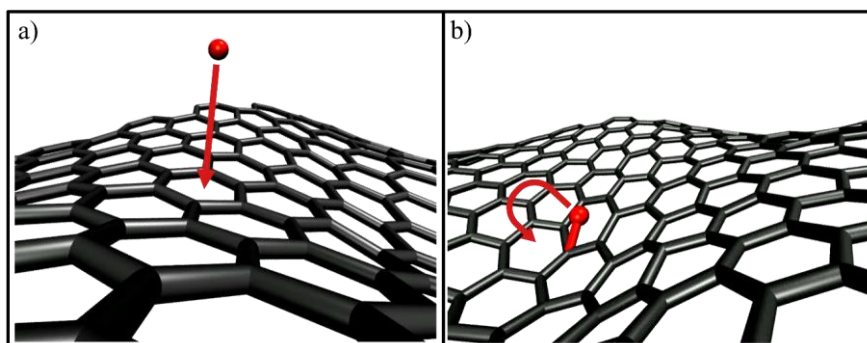


Figure 1: Schematic of proton permeation via a) Mechanism 1 and b) Mechanism 2 in graphene.