Insights into Proton Permeation Mechanism in 2D Materials by Molecular Simulations

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Graphene and graphene oxide (GO) are promising materials in fuel cell applications due to their selective proton transport and exceptional mechanical and thermal stability properties [1-2]. We have explored the electrochemically driven proton permeation mechanisms through graphene and graphene oxide using ReaxFF Molecular Dynamics simulations. Our findings reveal that the primary mechanism for proton transport in graphene and graphene oxide is the flipping mechanism (Mechanism 2), in which the proton first adsorbs onto the surface and flips through the basal graphene ring. Our results show that the proton conductivity increases with temperature due to the high thermal energy and a less negative surface charge density. In the case of graphene, as the temperature increases from 300 K to 350 K, the proton conductivity rises from 0.014 mS/cm to 0.020 mS/cm, showing a 43% improvement. In the case of graphene oxide, the proton conductivity at 300 K is twice that of graphene. The proton conductivity for graphene oxide improves from 0.031 mS/cm to a maximum value of 0.038 mS/cm, showing a 23% improvement as the concentration of hydroxyl groups increases. The epoxy groups significantly influence the surface geometry and charge density of graphene oxide, offering a potential for tunable proton conductivity in graphene oxide.
To advance the efficiency of the proton exchange membrane fuel cell within the temperature range of 100-250ºC, we employed HTFSI electrolyte dissolved in polyethylene glycol (PEG). The study focuses on pristine graphene and h-BN as 2D membranes due to their thermal and structural stability at high temperatures. In addition, we have demonstrated the role of single vacancy (SV) defects in proton permeation without compromising selectivity. By systematically varying the concentration of SV defects and analysing the projected permeation area, including the distorted hexagonal rings surrounding the defective region, we aim to provide a deeper mechanistic understanding of proton transport through 2D materials.
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References

[1] O. J. Wahab *et al*., *Nature*, vol. 620, no. (Aug 2023), pp: 782-786
[2] Z. F. Wu *et al*., *Nature Communication*, vol. 14, no. 7756 (Nov 2023)

Figures

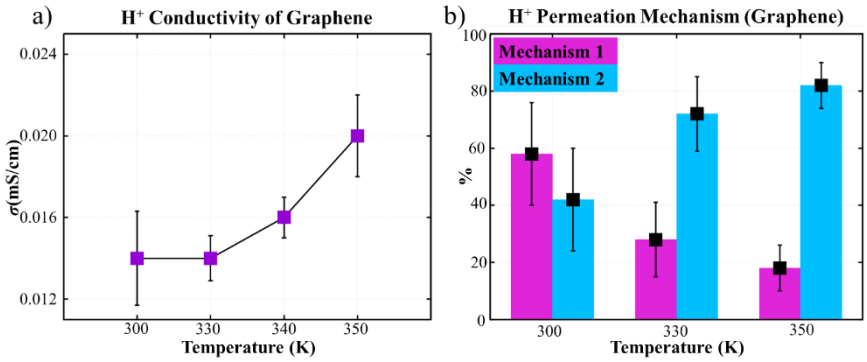

Figure 1: a) Proton conductivity. and b) permeation mechanism. of Graphene.

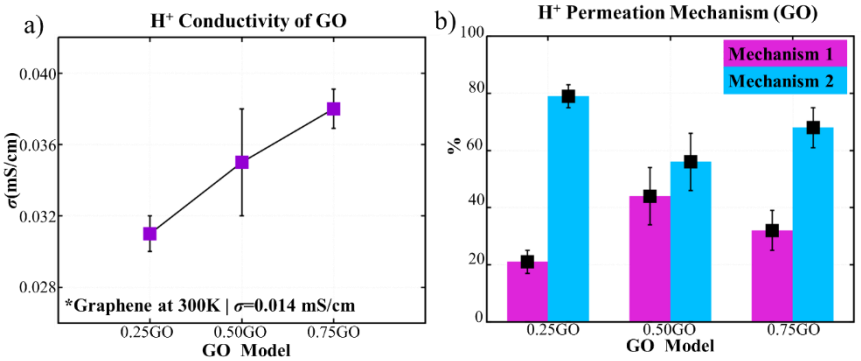

Figure 2: a) Proton conductivity. and b) permeation mechanism. of Graphene Oxide.