Structure of water around graphene nanoribbons from ab initio machine learning simulations

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Water-graphene interactions are fundamental to a range of applications, from catalysis to nanofiltration and biosensing [1]. In this study, we investigate the structure and reactivity of water at the surface of graphene nanoribbons using molecular dynamics simulations driven by ab initio machine learning models for the potential energy surface. We first studied the electronic structure of armchair- and zigzag-terminated ribbons using density functional theory (DFT) calculations with the Quantum Espresso suite [1]. Afterwards, we performed molecular dynamics simulations at room temperature employing both the MACE-MP foundation model [2] and a specialized model trained using the DeePMD-kit [3]. For the latter, we used energies and forces derived from DFT by adopting the Strongly Constrained and Appropriately Normed (SCAN) exchange and correlation functional [4]. Analysis of the radial distribution function and water density profiles around the nanoribbon (see Figure 1) reveals that the SCAN-based model better reproduces the experimentally observed structure of water, underscoring its accuracy in capturing hydrogen-bonding interactions. Notably, we observe that the graphene nanoribbon surface interacts more strongly with the hydrogen atoms than with the oxygen atoms of water molecules, indicating a preferential orientation of water at the interface. These findings highlight the importance of functional choice in accurately modeling water-carbon interfaces and suggest that graphene nanoribbons may exhibit distinct properties in water structuring and interfacial chemistry, potentially influencing their behavior in water purification, sensing, and electrochemical applications.

Figures

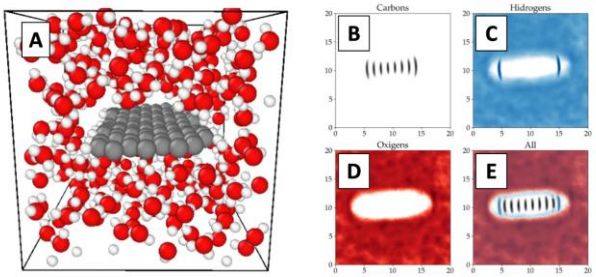

Figure 1. A) Snapshot of a graphene nanoribbon in water. B), C), D), and E) show the density of carbon, hydrogen, oxygen, and all atoms, respectively, at a plane perpendicular to the plane of the graphene nanoribbon.

References

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