

“Quantum Modeling and Classical Simulations of Hydrogen-Bonded Oligomers: New Candidates for Proton Conduction”

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There is widespread interest in developing *anhydrous* proton exchange membranes (PEMs) that may raise operating temperatures of hydrogen fuel cells to higher values, significantly higher than the ~90 °C limit found for hydrated polyelectrolyte materials. Several groups have studied PEMs composed of amphiprotic organic groups such as imidazole and triazole tethered to polymeric backbones, only to find exponentially low conductivities. This points to a lack of fundamental understanding into how tethering of amphiprotic groups influences hydrogen bond networks and proton transfer dynamics in such materials. In the present work, we have applied a variety of molecular modeling approaches to address this problem. We have applied electronic structure calculations to long chains of amphiprotic species in the gas phase, to investigate how hydrogen bond strength in long “wires” varies with functional group properties such as the deprotonation energy (pK_a) and proton affinity. We have also applied classical molecular dynamics simulations and cluster statistics to investigate the nature of hydrogen bond networks formed from imidazole groups tethered to oligomeric backbones. Finally, we have also applied reactive molecular dynamics simulations to understand how both local and cooperative properties – i.e., functional-group and network properties, respectively, impact proton diffusion in glassy solids composed of oligomeric amphiprotic groups. Our results provide new insights and design principles for the next generation of anhydrous proton conductors.