

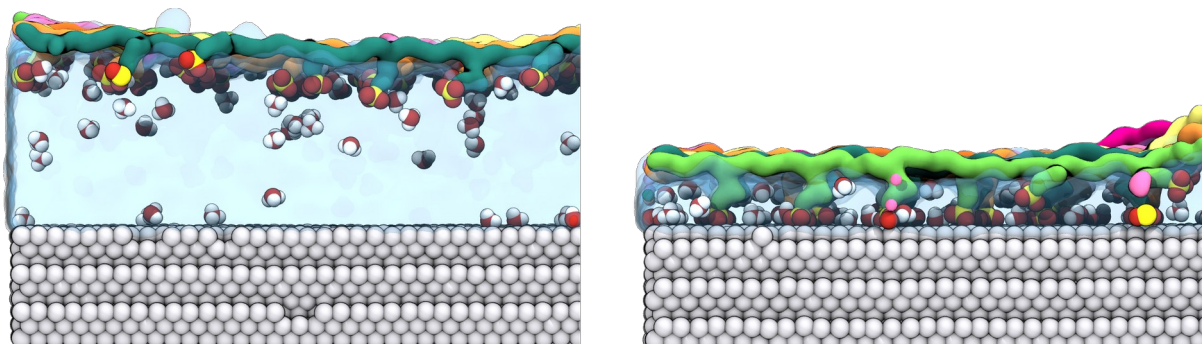
# Exploring the ionomer-platinum interface in the cathode-catalyst layer of polymer electrolyte membrane fuel cells via molecular dynamics simulations

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Polymer electrolyte membrane fuel cells (PEMFCs) play a vital role in establishing a sustainable hydrogen economy, making research into their performance and durability an important endeavor. For the performance of PEMFCs, the cathode-catalyst layer (CCL) plays an important role. This layer consists of a mixture of a proton-conducting ionomer phase and platinum catalyst supported on carbon substrate. At the platinum surface, the oxygen reduction reaction (ORR) takes place reducing the supplied oxygen gas to form water. The ionomer plays an important role in facilitating the transport of protons to the catalyst surface, as required for the electrochemical reaction. While most product water molecules evaporate and escape from the interface, some residual water remains and forms a water film at the ionomer-platinum interface, which impacts the transport of the ORR reactants (both oxygen and protons) and, thus, the CCL performance [1].

Currently one of the most used ionomers is Nafion, which features a perfluorinated alkane backbone. In line with current efforts to reduce usage of such molecules extensive research is performed in order to identify novel ionomers with similar or even better characteristics for usage in PEMFCs.



Using molecular dynamics simulations, we investigate the interface between Nafion and platinum to elucidate structure-function relationships and identify materials descriptors determining their performance as components in CCLs. In this study we focus our efforts on the influence of the water film that is present between the catalyst and the ionomer layer. The thickness of the water film is dependent on multiple factors such as the operating conditions, the local electrochemical environment, and hydrophilicity of the adjacent layers [1,2,3]. From our simulations of water films of varying thickness, we analyze the distributions of reactant species and orientation of water dipoles at the catalyst surface, and assess influence of the water film on the local ionomer structure and the transport of protons within the boundary layer.

These analyses provide insight into the structural and dynamical properties of CCLs at an atomistic scale. The developed workflow will guide future simulations exploring non-fluorinated ionomers as Nafion alternatives with better environmental friendliness to be used in CCLs for PEMFCs.

[1] W. Olbrich, *et al.*, *J. Electrochem. Soc.*, **2022**, 169, 054521.

[2] V. M. Fernández-Alvarez, *et al.*, *J. Electrochem. Soc.*, **2022**, 169, 024506.

[3] D. Damasceno Borges, *et al.*, *ACS Nano*, **2013**, 7, 6767-6773.