

Supplementary Information

Insight into oxygen diffusion mechanism in ionomer film on catalyst surface with varying perfluorosulfonic acid and water contents

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Model validation

In classic molecular dynamics (MD) simulations focusing on the hydrated Nafion systems of proton exchange membrane fuel cells (PEMFCs), the models are commonly validated by examining the density of bulk Nafion and the diffusion coefficients of water molecules. Accordingly, we built the bulk Nafion models by using the potential parameters from the modified DREIDING force fields.^{1, 2} As shown in Fig. S1, the simulated results including the density of bulk Nafion and diffusion coefficients of water molecules are in agreement with the previous results,^{1, 3-7} thus providing validation for the accuracy of the applied potential parameters and MD models. Furthermore, the modified DREIDING force fields are widely utilized in the field of PEMFCs.⁸⁻¹⁵ All of these indicate that the simulation results presented in this study are reliable.

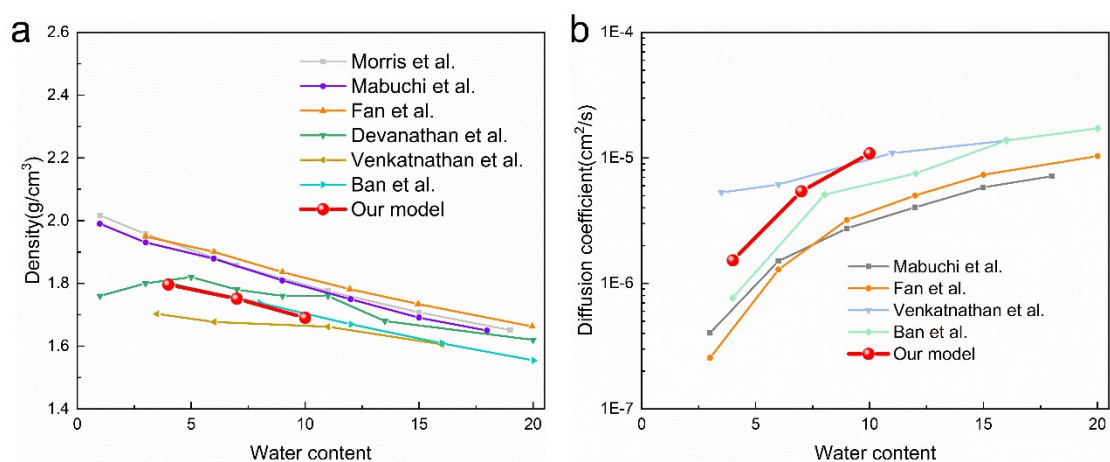


Fig. S1. Simulated (a) density of bulk Nafion and (b) diffusion coefficients of water molecules.²

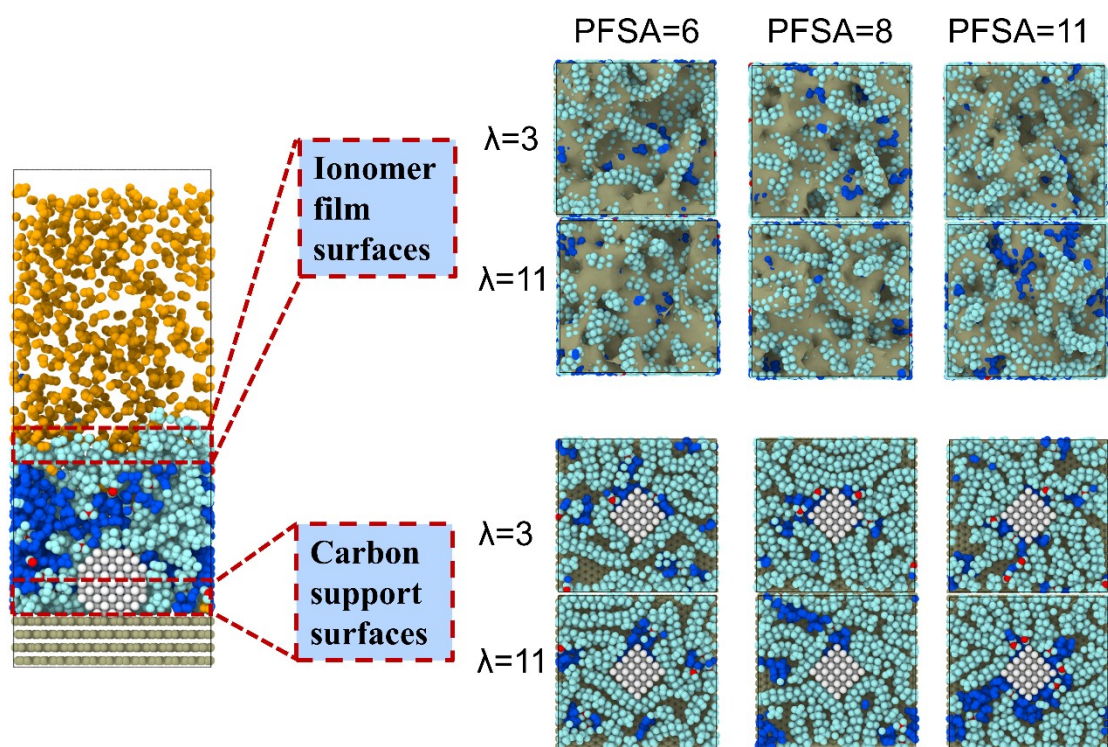


Fig. S2. Snapshots of surface regions and dense layers.

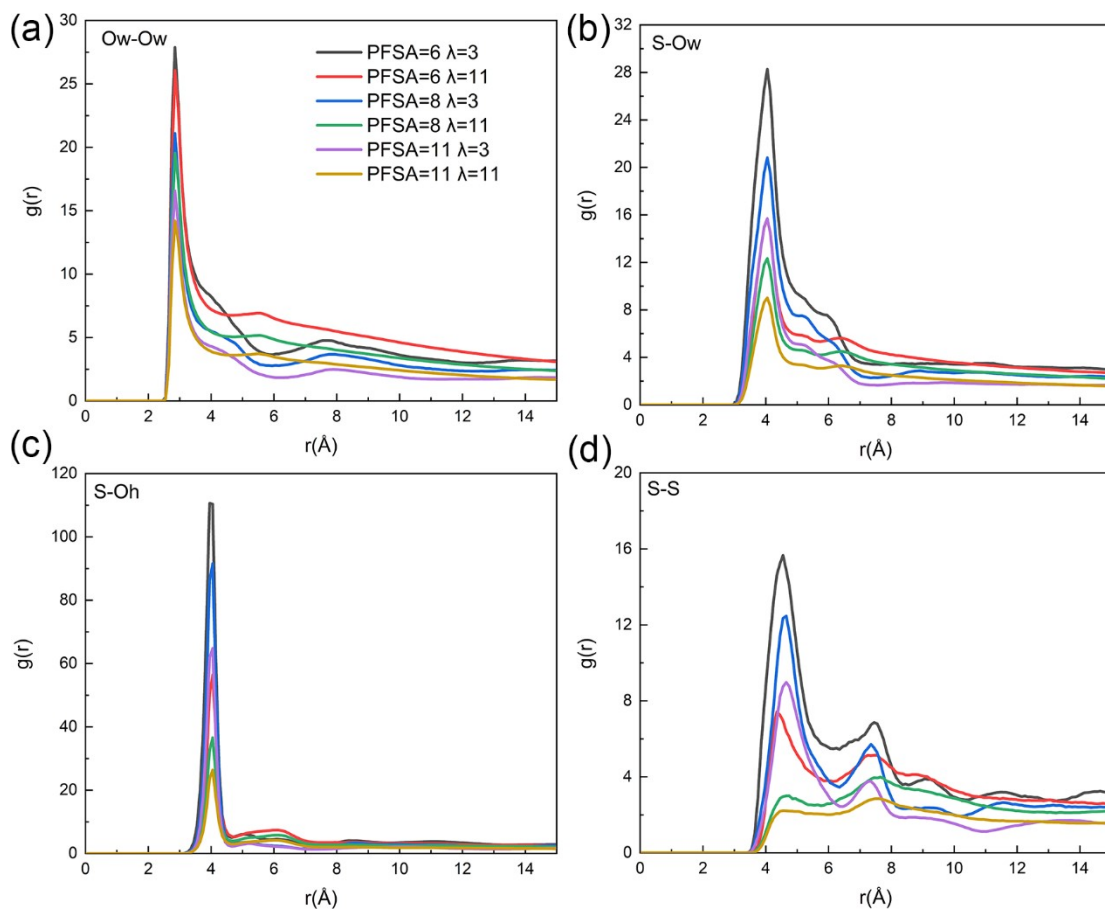


Fig. S3. The RDFs of (a)Ow-Ow (oxygen atoms of water molecules), (b)S-Ow, (c)S-Oh (oxygen atoms of hydronium ions), (d)S-S (sulfur atoms of sulfonic groups).

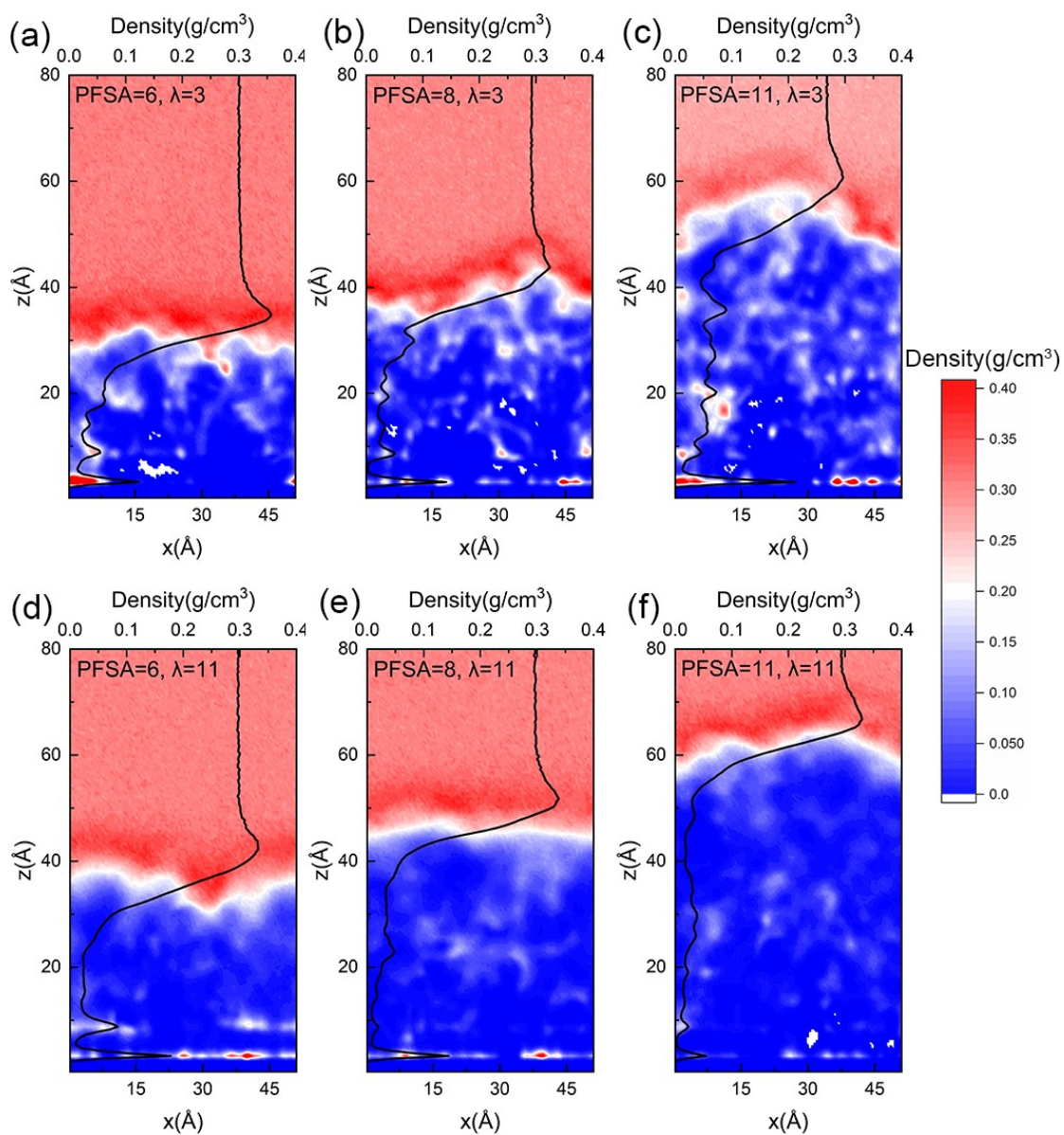


Fig. S4. The density distributions of oxygen molecules.

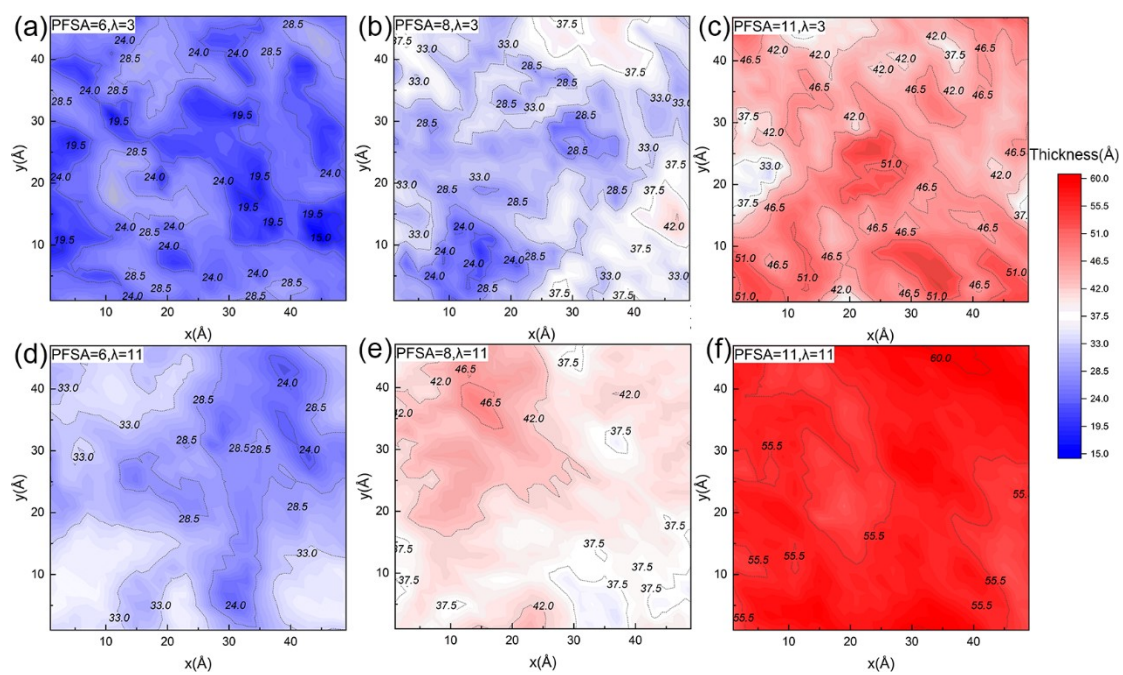


Fig. S5. The thickness distribution of ionomer film through the x-y planes.

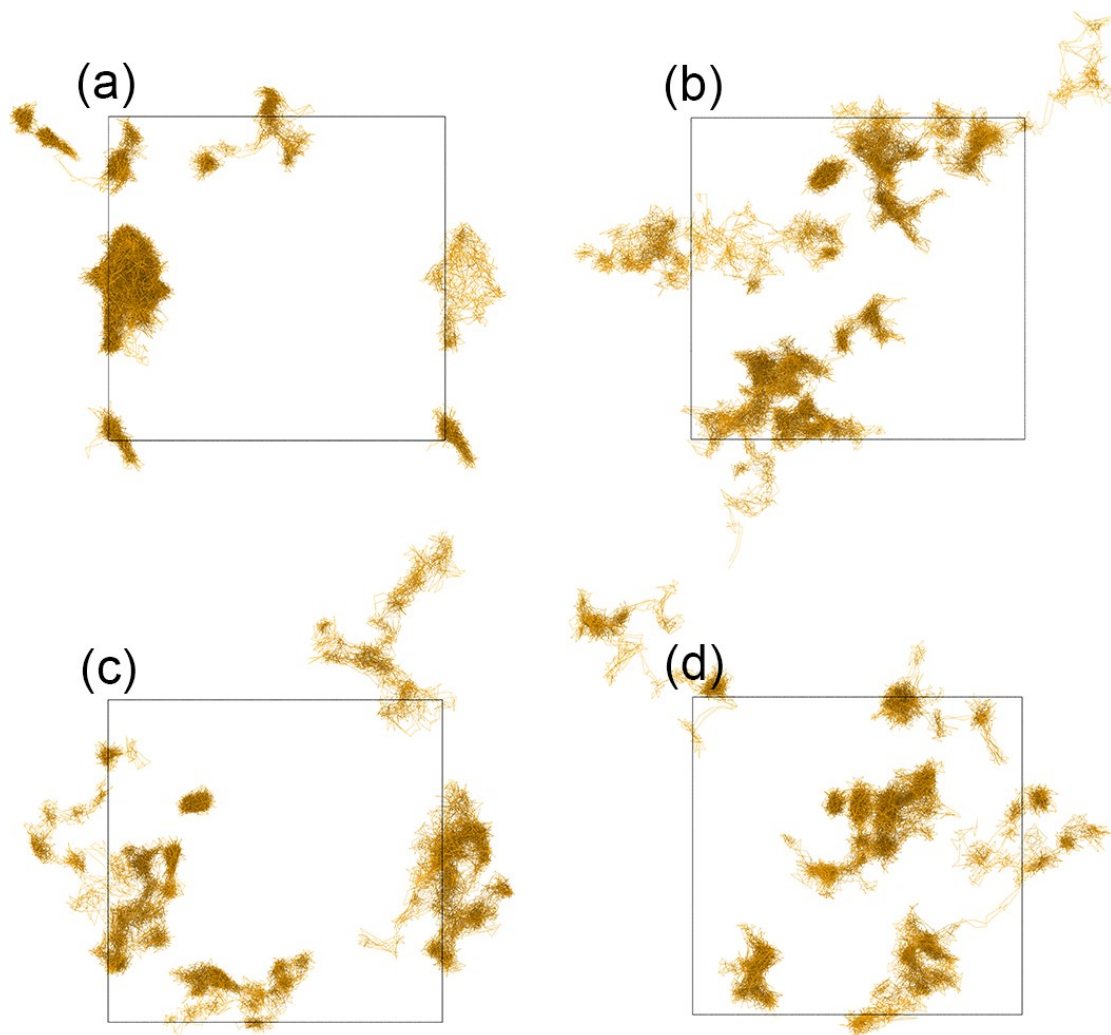


Fig. S6. Typical oxygen diffusion trajectories in ionomer film. (a) PFSA=6, $\lambda=3$; (b) PFSA =11, $\lambda=3$; (c) PFSA =6, $\lambda=11$; (d) PFSA =11, $\lambda=11$.

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