

Supplementary Information for

Evidence for a Crystalline-Rich Skin on Perfluorosulfonate Ionomer Membranes

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Surface structure of partially hydrated membrane

As the membrane would swell and become not smooth after hydration, it cannot be used in grazing incident experiment as described above. To investigate the surface structure of hydrated PFSI membrane, we have used an alternative but not too standard solution. Firstly, we prepared a wet filter paper and then covered it on a membrane for a short time to wet the membrane surface but without detaching the membrane from the glass slide. After that, we immediately conducted the grazing incident experiment. It's worth noting that this experiment is not a standard procedure but just an alternative solution.

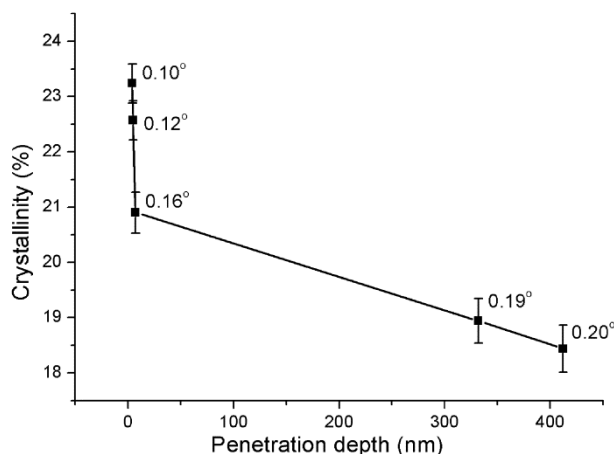


Figure S1. Crystallinity at the surface of partially hydrated membrane as a function of the penetration depth. The corresponding incident angles are displayed beside each data point.

As shown in Figure S1, in the surface layer from 400nm depth to 4nm layer, the crystallinity increased from 18.4% to 23.2%. The variation of crystallinity in the surface region for the partially hydrated membrane have the same trend as dry PFSI membrane in Figure 3 but with lower crystallinity in each layer at the same depth. Therefore the surface structure with high crystallinity including the crystallites concentrated skin may be weakened after hydration. Further work is necessary to investigate the surface structure transformation of PFSI membrane in different circumstance.