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Evidence for hydrated channels and connected water clusters in proton conductive membranes based on sulfonated syndiotactic polystyrene

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Syndiotactic polystyrene (sPS) is able to form different kinds of co-crystalline phases with guest molecules of various size, shape and property. Several advanced materials have been already produced based on sPS co-crystalline films with fluorescent, photoreactive, chiral and paramagnetic guest molecules. In particular, sulfonated sPS (ssPS) can be used as proton-conductive membrane (PCM) for fuel cells, as it presents high proton conductivity (comparable with Nafion), it shows a high chemical and thermo-mechanical stability and it has a low cost. In spite of extensive studies the microstructure of PCMs is still subject of active debates and many structural models have emerged to describe the hydrated membranes. In the current study the morphology of different sPS co-crystalline films (clathrate with guest molecules or as empty crystalline form) and the structural features of ssPS upon hydration were thoroughly understood by combining WAXS, FT-IR and TEM with SANS. By exploiting, on one hand, the neutron contrast variation enabled by the selective hydrogenation and deuteration of different regions and constituents of the sPS and ssPS films and, on the other hand, the separation of scattering features from different constituents on specific directions and sectors of the detection plan, using uniaxially stretched films, an unambiguous structural and morphological characterization of such complex systems could be achieved by SANS. The systematic composition and structural investigation on PCMs based on sPS films, starting from their co-crystallization with guest molecules through the sulfonation and the guest exchange processes (for varying the SANS contrast) and followed by subsequent hydration (using H_2O or D_2O for the contrast variation) from liquid phase or vapor phase using a humidity cell, has evidenced in the amorphous regions hydrated channels that connect water clusters and are aligned along the stretching direction. The high conductivity shown by the membranes is supported by these channels, while the mechanical reinforcement is provided by the oriented lamellar staples in the co-crystalline regions.

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