Investigation of the Ionic Conductivity and Relaxation of COF/polymer Composite Solid Electrolytes

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With the ever-growing issue of fossil fuels scarcity, the development of clean and renewable energy resources has become the most important global challenge. Proton exchange membrane fuel cells (PEMFCs) are a clean technology employing hydrogen as fuel and yielding only water and heat as byproducts of the electrochemical reaction. Their efficiency mostly depends on the proton conductivity (σ) of the solid polymer electrolyte (SPE) in the membrane electrode assembly (MEA). In a PEMFC operating at 60-80 °C, the ideal SPE should possess high σ also at low relative humidity (RH), apart from being insoluble in water and stable in acids or bases. Indeed, the main challenge in the development of PEMFCs consists in the water management: although σ usually depends on the SPE's hydration, the latter is detrimental for the cell stability and performance, since condensation, followed by flooding, is inevitable when the operating temperature is around 80 °C. A common and simple activation strategy in real-life use of FCs is to steam or boil the MEA prior to use, but SPEs exhibiting good σ preservation at low RH are still lacking. Although covalent organic frameworks (COFs) do not usually exhibit high intrinsic proton conductivity (σ), they have been recently proposed as SPEs in PEMFCs, thanks to their high crystallinity and stability to acids and bases. [1]

Here, a simple strategy to improve the performance of poor COF-based proton conductors through addition of sodium polyacrylate (PANa) superadsorbent polymer is presented. Electrochemical impedance spectroscopy (EIS) investigations after activation at high temperature and RH provide insights into the role of PANa, whose presence is key to preserve high σ at low RH. For the first time a humidity-dependent X-ray diffraction analysis was performed to support the EIS data, revealing a strengthening of the stacking interaction along the COF (100) plane direction via H-bonding, thereby promoting the proton hopping. Furthermore, an extensive investigation of the relaxation mechanism as a function of PANa content enabled us to determine a Debye relaxation regime for the COF/PANa blends with the maximum relaxation frequency increasing with the PANa content, resulting in a more efficient charge carrier transport. [2]

References

[1] R. Sahoo, S. Mondal, S. C. Pal, D. Mukherjee and M. C. Das, Adv. Energy Mater., 11 (2021), 2102300.
[2] S. Gullace, L. Cusin, F. Richard, N. Israfilov, A. Ciesielski, P. Samorì, *submitted*

Figures

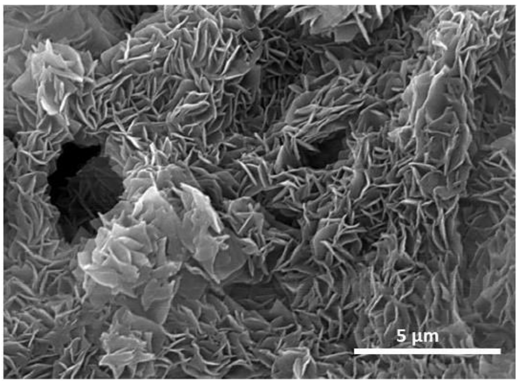

Figure 1: Scanning electron microscopy image of the investigated COF/PANa blend.