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# Novel PBI/SGO composites as alternative proton exchange membranes for fuel cells

## C. Membranes for energy and the environment

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**Abstract**

The urgency of developing new materials able to outperform the conventional electrolyte employed in proton exchange membrane fuel cells (PEMFCs), i.e., Chemours' Nafion®, is growing more and more. The reason must be searched in the four issues afflicting Nafion®: (i) its expensiveness (\$0.24 $cm^{-2}$) due to the fluorination process; (ii) the severe loss of mechanical resistance when exposed to temperatures higher than 80 °C; (iii) the sharp drop in proton conductivity when conditions of dehydration are reached; (iv) the belonging to the larger class of per- and polyfluoroalkyl substances (PFASs), which are being strongly restricted due to their dangerousness toward the environment and human health. In such a context, this work deals with the development of a simple and reproducible procedure to prepare self-assembling PBI/SGO composites, whose characterization aims at assessing their potentiality as non-fluorinated proton exchange membranes able to work within the so-called "conductivity gap", which is identified between 80 and 120 °C. Such operating conditions would foster an enhanced reaction kinetics, the use of less expensive electrocatalysts, and an easier water management in the device. Five samples with PBI-to-SGO mass ratios of 3:1, 2:1, 1:1, 1:2, and 1:3, which were never explored in the literature before, are prepared via an innovative procedure that combines the GO sulfonation with sulfuric acid and a conventional solution casting method. The investigation of the prepared samples via surface and cross-sectional scanning electron microscopy reveal the uniformity of all the composites and the absence of macroscopic defects. From the X-ray diffraction patterns and the attenuated total reflection Fourier-transform infrared spectra, both PBI and SGO contributions are identified, suggesting the correct combination of the constituents within the self-assembled membranes. Thermogravimetric analysis demonstrates the remarkable thermal stability of the composites thanks to the effect of the PBI moieties, especially in the desired temperature range. The samples with higher SGO content exhibit excellent water retention capability, attributed to the influence of the hydrophilic oxygenated and sulfonic acid groups of SGO, while maintaining a reasonable swelling ratio. The electrochemical impedance spectroscopy tests are performed with the in-plane and through-plane configurations to evaluate the proton transfer ability in the two directions without any kind of doping. PBI/SGO 1:2 achieves the highest in-plane proton conductivity of 0.115 S $cm^{-1}$ at 120 °C, overcoming the value typically demonstrated by Nafion® at 80 °C (≈ 0.1 S $cm^{-1}$). Concerning through-plane, PBI/SGO 1:3 displays the best performance of 0.025 S $cm^{-1}$ at 120 °C. In light of the obtained outcomes, the PBI/SGO composites with higher SGO content seem to possess great potential as alternative non-fluorinated electrolytes for PEMFC applications, filling the "conductivity gap".