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Abstract

Hybrid Membrane Materials Based on Polybenzimidazole and Silica with Grafted Phosphonic Groups for Fuel Cell Applications †

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Abstract: Owing to high thermal and chemical stability and good mechanical properties, polybenzimidazole (PBI) doped with phosphoric acid is a very promising material to be used as an electrolyte in medium-temperature fuel cells. However, PBI use at temperatures below ~160 °C is impeded by the leaching of free H_3PO_4 from the membrane. In order to overcome this problem, one of the possible approaches is the incorporation of inorganic particles capable of stabilizing H_3PO_4 in the PBI matrix. Surface-modified particles are more efficient for this purpose. In this work, we study the properties of proton-conducting membranes based on PBI and silica particles surface-modified with propylphosphonic groups. Composite membranes are obtained by the casting of polymer solution containing tetraethoxysilane and modified silane ((2-diethylphosphatoethyl)triethoxysilane) with hydrolysis by HCl. The mass concentration of the dopant is 5 or 10 wt %, and the mole fraction of functional groups on the oxide surface is varied in the range of 0–100 mol % by changing the composition of the precursor mixture. All films are treated by 75% H_3PO_4. The resulting membranes are characterized using transmission and scanning electron microscopy, IR spectroscopy, and impedance spectroscopy. The grafting of functional $–PO_3H_2$ groups onto the silica surface leads to a significant increase in the uptake of phosphoric acid by hybrid membranes, the content of which determines the conductivity of these materials. An increase in the number of $–PO_3H_2$ groups leads to both an increase in the degree of acid doping and ionic conductivity. The conductivity of the best samples obtained reaches 0.081 S/cm at 160 °C. The introduction of acid groups on the dopant surface is a promising approach from the point of view of reducing the amount of phosphoric acid required to maintain a high proton transport rate.

Keywords: proton conductive membrane; polybenzimidazole; hybrid membrane; fuel cell; proton conductivity; surface modified silica

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