In Situ Formed Phosphoric Acid/Phosphosilicate Nanoclusters in the Exceptional Enhancement of Durability of Polybenzimidazole Membrane Fuel Cells at Elevated High Temperatures

Most recently, we developed a phosphotungstic acid impregnated mesoporous silica (PWA-meso-silica) and phosphoric acid doped polybenzimidazole (PA/PBI) composite membrane for use in high temperature fuel cells and achieved exceptional durability under a constant current load of 200 mA cm−2 at 200°C for over 2700 h. In this work, the fundamental role of PWA-meso-silica in enhancing the stability of the PA/PBI membrane has been investigated. The microstructure, the PA uptake, swelling ratio, mechanical property and conductivity of PA/PBI/PWA-meso-silica composite membranes depend on the loading of PWA-meso-silica. The results indicate that the optimum limit of PWA-meso-silica loading in the PA/PBI membranes is 15 wt%. Detailed analysis indicates that the mesoporous structure of the PWA-meso-silica framework disintegrates, forming phosphosilicate phases within the PBI polymeric matrix during fuel cell operation at 200°C. The in situ formed phosphosilicates can immobilize a significant amount of PA, forming PA/phosphosilicate nanoclusters that possess high proton conductivity (e.g., 7.2 × 10−2 S cm−1 at 250°C) and stability and substantially inhibits acid leaching out of the membrane. The substantially reduced acid leaching also alleviates the excess acid in the catalyst layer, reducing the detrimental effect of excess acid on the agglomeration of Pt catalysts especially in the cathode catalyst layer. These phenomena are responsible for the exceptional stability in proton conductivity as well as the significantly reduced agglomeration of Pt nanoparticles in the anode and cathode catalyst layers of PA/PBI/PWA-meso-silica composite membrane fuel cells.

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