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journal homepage: <http://www.elsevier.com/locate/memsci>Effect of *N*-cyclic cationic groups in poly(phenylene oxide)-based catalyst ionomer membranes for anion exchange membrane fuel cells

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ABSTRACT

Herein, as a binder (or catalyst ionomer) for AEMFCs, we investigated the effect of two different cationic copolymers based on poly(phenylene oxide) (PPO) with *N*-cyclic quaternary ammonium (QA) groups, including six-membered dimethyl piperidinium (DMP) and bis-six-membered azonia-spiro undecane (ASU). An earlier report on the same polymers for membranes in AEMFCs indicated the better electrochemical performance of PPO-ASU compared with PPO-DMP. Therefore, we would like to investigate these two polymers for catalyst ionomers. The outcome in this study using these two copolymers as catalyst ionomers indicates the opposite result; the electrochemical performance of the PPO-DMP ionomer is much better than the PPO-ASU ionomer. The commercial Fumion ionomer was used for the qualitative comparison. The density functional theory (DFT) calculation of the adsorption energy according to different orientations of the cationic groups on the catalyst surface shows that there is no difference between the adsorption energy of DMP and ASU cations, in compliance with the orientations of the cations. Although the PPO-ASU ionomer membrane has the highest hydroxide conductivity at 60 °C in liquid water, the hydrogen oxidation/reduction (HOR) activity of PPO-DMP and PPO-ASU showed similar values with the Fumion ionomer. While the PPO-DMP ionomer membrane shows relatively large fuel gas (hydrogen) permeability in dry and wet conditions, due to the chain flexibility and the presence of two methyl groups compared to the single methyl groups and lower flexibility of the PPO-ASU and Fumion ionomers. The electrochemical performance of a membrane electrode assembly (MEA) using the PPO-DMP ionomer exhibited an exceptional peak power density of 335 mW cm⁻² compared to lower peak power densities of PPO-ASU and Fumion ionomers under 60 °C and a fully humidified condition (H₂/O₂). The SEM images of MEAs after testing supports the conclusion that the PPO-DMP ionomer forms a uniform catalyst interface that is very well bound between the electrode and membrane, unlike the PPO-ASU and Fumion ionomers. The PPO-DMP ionomer membrane also showed better tensile strength and elongation at break than the PPO-ASU ionomer membrane. Therefore, we conclude that the well-prepared three-phase boundary structure played a critical role for the catalyst ionomer in each electrode, overcoming one of the critical performance-limiting factors.

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