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Electrochemical Characterization of Stabilized Heteropolyacid / Ionomer Composite Membranes for High Temperature PEMFCs

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INTRODUCTION

From an applications point of view, several advantages result by operating a PEMFC at elevated temperatures (above 100°C) and low relative humidities. Therefore, a significant economic incentive exists to develop and commercialize PEMFCs that operate under these conditions. Nafion® / heteropolyacid (HPA) composite membranes have been proposed for high temperature applications[1]. However, heteropolyacids are soluble in water and leach out relatively easily from the host matrix.[2] Stabilization of the HPA additive within the ionomeric matrix - which is essential for proper membrane processing and for good interfacial stability in membrane electrode assemblies (MEAs) – has been recently demonstrated in Nafion® based systems.[2, 3] Studies aimed at investigating the water uptake and proton conductivities of the Nafion®/ stabilized HPA composite membranes as a function of membrane processing conditions, operating temperatures and relative humidity were performed. Composite membranes prepared using hydrocarbon host matrixes other than Nafion® were also evaluated to elucidate the role played by the inorganic additive in enhancing proton conductivity.

EXPERIMENTAL

Composite membranes were made using three techniques. The first technique involved addition of an appropriate amount of HPA directly to the solubilized ionomer and preparing the membrane by solvent casting. In the second technique [2], the HPAs were chemically modified and converted from a low surface area, water-soluble compound into a high surface area water insoluble compound by a cation exchange process. The chemically modified HPAs were then incorporated into an ionomeric matrix, and membranes prepared by solvent casting. The final method involved stabilization of the HPA using an in-situ sol-gel technique.[3] Nafion®112, recast Nafion® and recast hydrocarbon membranes were used as blanks. All data were compared against these membranes.

The vapor phase water uptake of the membranes at room temperature was estimated as a function of relative humidity. Humidity control was achieved using saturated salt solutions. Liquid phase water uptake measurements were performed only on the stabilized membranes. Proton conductivity measurements were made using a specially designed 4 point conductivity cell (Bekktech LLC)[4]. Both direct and alternating current techniques were employed. Temperature and humidity were controlled using a fuel cell test system (Scribner Associates, model 890B) and a flow loop (built in-house). Room temperature proton conductivity measurements were obtained using a 2 point conductivity cell placed within controlled (using salt solutions) humidity environments. Finally, membrane electrode assemblies (MEAs) were prepared by applying catalyst onto the membrane using an airbrush. The MEAs were evaluated in a fuel cell environment at different temperature and relative humidity conditions. The proton conductivity of the membranes was estimated *in-situ* from resistance measurements obtained using the current interrupt technique. The conductivities obtained using different techniques were compared. The effect of the stabilization of the additive on proton conductivity of the composite membranes was studied.

RESULTS

The room temperature water uptake and proton conductivities of Nafion® / stabilized PTA composite membranes are shown in Fig. 1. Conductivities obtained from *in-situ* measurements in an operating fuel cell at different temperatures and inlet relative humidities are shown for stabilized Nafion® / PTA composite membranes (Fig. 2). It is seen that while conductivity does depend on temperature, the effect of relative humidity is more pronounced. Further results will be discussed in the paper.

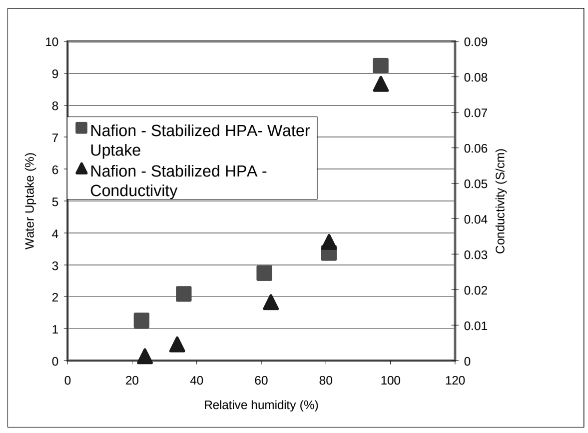

Fig. 1: Room temperature water uptake and proton conductivities of Nafion® / stabilized HPA composite membranes

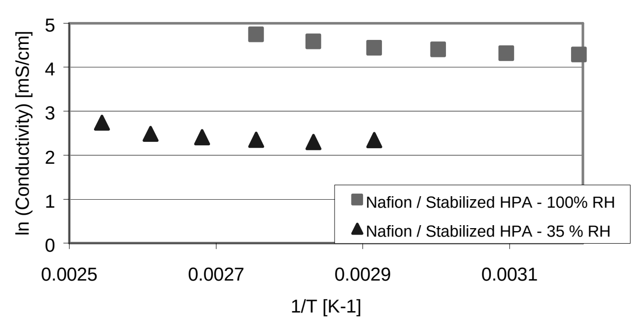

Fig. 2: Proton conductivities of Nafion® / stabilized PTA composite membranes at different temperatures and inlet relative humidities (RH)

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