



Enhanced Performance Of Direct Methanol Fuel Cell Using Talc Modified Nafion Membrane

Lepakshi Barbora

Centre for Energy, Indian Institute of Technology Guwahati, India

Anil Verma

Department of Chemical Engineering, Indian Institute of Technology Guwahati, India

Abstract:

Magnesium silicate hydroxide, $Mg_3Si_4O_{10}(OH)_2$, commonly known as talc, is employed to modify nafion for application in direct methanol fuel cell. Improved proton conductivity and reduced fuel permeability are shown by the talc/nafion composite membrane. Highest proton conductivity is achieved with 5% talc/nafion membrane, which is 27.14% higher than that of pure cast nafion membrane. However, 1% talc/nafion membrane reduced methanol permeability by 51% compared to nafion 117 and by 42% compared to pure cast nafion. Fuel cell performance was evaluated at varying methanol concentrations and temperatures. The talc/nafion membranes exhibited higher open circuit voltages and current density than both pure cast nafion and nafion 117. The highest current density is achieved with 1% talc/nafion with 3 M methanol at 80°C. At a constant cell voltage of 0.28 V, 155.95 mA·cm⁻² of current density is obtained with 1% talc/nafion, 6.4 mA·cm⁻² with pure cast nafion and 6.82 mA·cm⁻² with nafion 117.

Keywords: Direct methanol fuel cell; Nafion composite; Methanol permeability; Proton conductivity; Talc

Introduction

Magnesium silicate hydroxide, commonly known as talc, with chemical formula $H_2Mg_3(SiO_3)_4$ or $Mg_3Si_4O_{10}(OH)_2$, consists of about 63.37% SiO_2 and 31.88% of MgO [1]. There are previous reports in the literature where SiO_2 has been successfully employed to modify nafion for reduction of methanol crossover (MCO) and enhancement of direct methanol fuel cell (DMFC) performance [2]. Similarly MgO , though not previously employed for nafion modification, has been successfully used for modification of Pt/C catalyst for direct oxidation of ethanol in direct alcohol fuel cell (DAFC) [3]. The presence of MgO in the electrocatalysts improves the kinetic processes, giving $1.8 \times 10^{-2} \text{ mA}\cdot\text{cm}^{-2}$ exchange current density for ethanol oxidation on Pt– MgO/C instead of $3.3 \times 10^{-4} \text{ mA}\cdot\text{cm}^{-2}$ on Pt/C. The best result is found on Pt– MgO/C electrode with Pt to MgO weight ratio of 4:1. According to Xu *et. al.* (2005) MgO plays a similar role as does Ru in Pt/Ru/C catalyst during alcohol oxidation by the bi-functional mechanism in DAFC. Pt acts as main catalyst for catalyzing the dehydrogenation of alcohol during the oxidation reaction and oxygen-containing species (OH_{ads}) can form on the Ru surface at lower potentials. These oxygen-containing species react with CO-like intermediate species on Pt surface to release the active sites for further alcohol oxidation [4]. Thus the coexistence of MgO with SiO_2 in talc and their application for nafion modification is anticipated to be advantageous for fuel cell operations. In this work, synthesis and characterization of talc/nafion composites, a novel combination, for application in DMFC is reported.

Experimental

Pure cast nafion and nafion composite membranes were prepared by casting method and characterized by SEM, XRD, and for proton conductivity and MCO [5]. DMFC performance was also evaluated using the membranes. For the DMFC, anode catalyst was 40%/20% Pt/Ru/C and the cathode catalyst was 40% Pt/C. The electrode area of the single cell was 5 cm^2 and the catalyst loading was $2 \text{ mg}\cdot\text{cm}^{-2}$. Membrane electrode assembly was fabricated by hot pressing the electrodes with the membrane at a temperature of $100 \text{ }^\circ\text{C}$ and pressure of $70 \text{ kg}\cdot\text{cm}^{-2}$ for 3 min. The flow rate of methanol and oxygen was maintained at $0.5 \text{ ml}\cdot\text{min}^{-1}$ and $0.7 \text{ L}\cdot\text{min}^{-1}$, respectively.

Results And Discussion

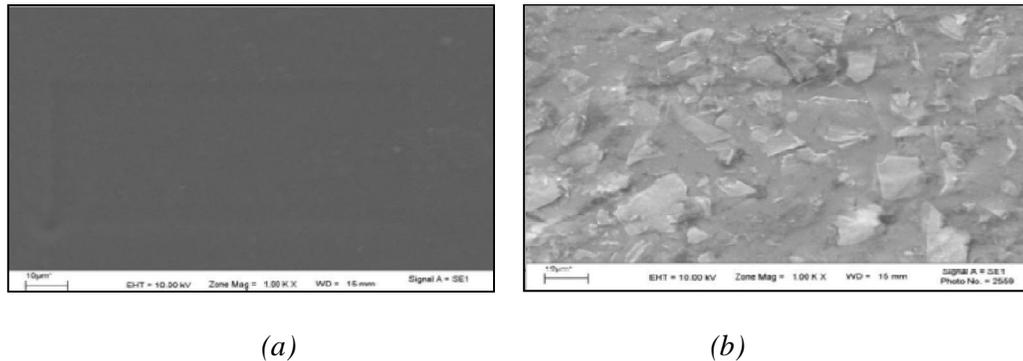


Figure 1: SEM images of (a) Pure cast nafion and (b) 5% Talc/nafion membrane

Modification of pure cast nafion with talc shows noticeably physical appearance in fig.1. The flaky particles of talc are distinctly visible in fig. 1(b). The distribution of talc within nafion is quite uniform and the membrane appears dense with no evidence of pores or cracks.

The XRD pattern of the talc/nafion membrane [fig.2] shows the characteristic peak at 25.24° of talc powder as well as the broad peaks of nafion between $14-18^\circ$. Thus the talc has contributed its crystallinity to the nafion composites.

Proton conductivity of talc/nafion membranes [fig.3] is higher than pure cast nafion attributed to the presence of SiO_2 . SiO_2 in talc exist as Si-OH, confirmed by FTIR spectra [fig. not shown]. Si-OH is hydrophilic, has affinity to proton adsorption and subsequently enhances the proton conductivity of the nafion composites [6]. The proton conductivity value was highest for the 5% talc/nafion membrane. Further increase in talc loading reduced conductivity attributed to the decrease in the effective number of proton exchange sites owing to non-hydrogen bonding between adjacent OH groups in the composites [5].

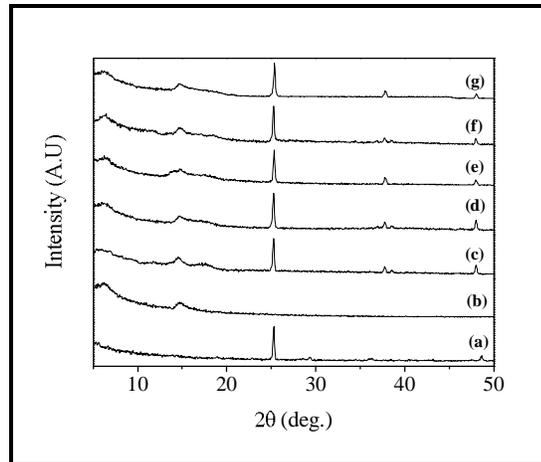


Figure 2: XRD of (a) talc powder, (b) pure cast nafion, (c) 1%, (d) 3%, (e) 5%, (f) 7%, and (g) 9% talc/nafion

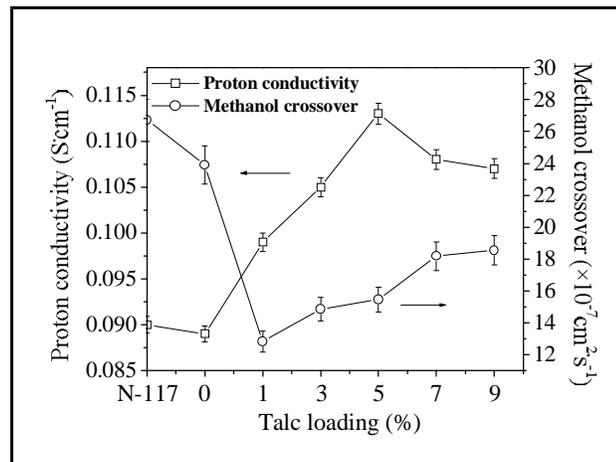


Figure 3: Proton conductivity and MCO profile of nafion 117, pure cast nafion and talc/nafion membranes

The MCO of 1% talc/nafion was lower than pristine nafion, attributed to the crystalline nature of talc. Further increase in talc loading promoted MCO, which may be due to the poor physical strength of the composite membranes as was confirmed by low tensile strength (fig.4).

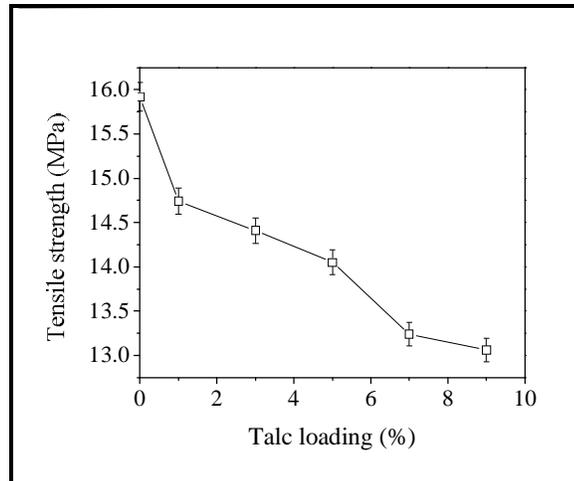


Figure 4: Tensile strength of pure cast nafion and talc/nafion membranes

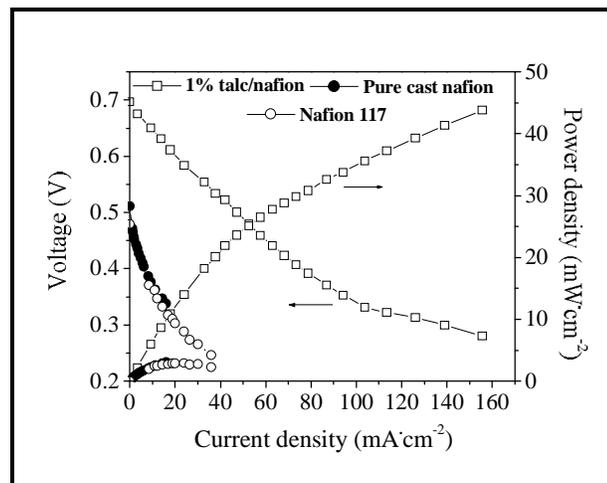


Figure 5 : DMFC performance of using different membranes with 3 M methanol at 80 °C

Amongst the talc/nafion composites, 1% talc/nafion membrane showed the best performance, which may be attributed to low MCO while maintaining proton conductivity. A maximum current density of $155.95 \text{ mA}\cdot\text{cm}^{-2}$ at 0.28 V was obtained at 80 °C with 3 M methanol, which is 59% higher than pure cast nafion and 56% higher than nafion 117 [fig.5].

Conclusion

Reduced MCO was obtained with 1% talc/nafion membrane while retaining the proton conductivity. The DMFC performance with talc/nafion membrane was improved tremendously.

At a constant cell voltage of 0.28 V, $155.95 \text{ mA}\cdot\text{cm}^{-2}$ of current density was obtained with 1% talc/nafion, $6.4 \text{ mA}\cdot\text{cm}^{-2}$ with pure cast nafion and $6.82 \text{ mA}\cdot\text{cm}^{-2}$ with nafion 117. The performance was reproducible and the possible cause of the enhanced performance is being investigated.

Acknowledgement

The authors gratefully acknowledge the financial support of the Department of Science and Technology, Government of India for the above project (SR/FTP/ETA-29/2007).

Reference

1. <http://www.aamineralspecimens.com> (23.07.2012)
2. Kim, D.J., Scibioh, M.A., Kwak, S., Oh, I.H., Ha, H.Y. (2004). Nano-silica layered composite membranes prepared by PECVD for direct methanol fuel cells. *Electrochemistry Communications*, 6(10), 1069-1074. D.J. Kim,
3. Xu C., Shen, P. K., Ji, X., Zeng, R., Liu, Y. (2005). Enhanced activity for ethanol electrooxidation on Pt–MgO/C catalysts. *Electrochemistry Communications*, 7(12), 1305–1308.
4. Prabhuram J., & Manoharan, R. (1998). Investigation of methanol oxidation on unsupported platinum electrodes in strong alkali and strong acid. *Journal of Power Sources*, 74(1), 54-61.
5. Barbora L., Singh, R., Shrotri, N., Verma, A. (2010). Synthesis and characterization of neodymium oxide modified nafion membrane for direct alcohol fuel cells. *Materials Chemistry and Physics*, 122(1), 211–216.
6. Tang H., Wan, Z., Pan, M., Jiang, S.P. (2007). Self-assembled Nafion–silica nanoparticles for elevated-high temperature polymer electrolyte membrane fuel cells. *Electrochemistry Communications*, 9(8), 2003-2008.