Development of Nanophase CeO₂-Pt/C Cathode Catalyst for Direct Methanol Fuel Cell

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1. Introduction

Direct methanol fuel cell (DMFC) is in the forefront of its commercialization due to significant advancements in fundamental, engineering and technological aspects leading to an era of their promising applications [1, 2]. In order to use DMFC as a portable power source, it is necessary to match for both high performance and small size. Increasing air utilization in the cathode of DMFC is a viable proposition for portable power applications, since the burden of air pump can be eliminated, reducing the volume, cost and energy consumption of the system. Fuel cell will exhibit lower performance when air is used instead of oxygen due to low partial pressure of oxygen in air. Hence, the incorporation of oxygen storage material such as ceria (CeO₂) [3] into cathode catalyst layer may increase the local oxygen concentration, leading to enhancement of the fuel cell performance.

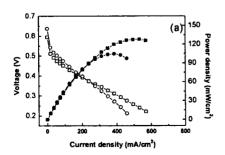
2. Experimental

The CeO₂ doped Pt/C was used as a cathode catalyst and Pt-Ru (50:50 at %) black catalyst (Johnson Matthey Inc) was used as an anode catalyst. The amount of ionomer content in the anode was varied based on the catalyst, but the amount of ionomer in the cathode was fixed at 60 wt% for ceria incorporated catalyst and 30 wt% for

black catalyst. Membrane electrode assembly was fabricated by placing a Nafion® 115 membrane between a PtRu (3 mg/cm²) anode and a Pt (3 mg/cm²) (ceria doped and undoped) cathode followed by hot-pressing at 140 °C, 70 kg_f/cm² for 150 sec. Impedance spectra were obtained on the operating cells with a potentiostat (IM6, Zahner).

3. Results and Discussion

Single cell tests were made for direct methanol fuel cell with 20wt%Pt/C cathode catalyst in the presence and absence of 1 wt% CeO₂ at oxygen or air in cathode compartment (Fig. 1). In oxygen atmosphere, pure Pt/C cathode catalyst exhibited higher performance (130.0 mW/cm^2) than ceria doped Pt/C (103.6 mW/cm^2) at 0.3 V (Fig. When air was used (250 sccm), higher performances were noted for CeO2 doped Pt/C than with Pt/C (Fig. 1b). With air ceria exhibited pronounced effect because of oxygen deficiency in the catalyst layer (Fig. 1b). The performance was higher in oxygen atmosphere compared to air atmosphere, since the oxygen partial pressure in the former was higher. In case of 1wt% ceria doped Pt/C, the power density at 0.3V was 103.0 and 55.0 mW/cm² for oxygen and air atmosphere respectively. The difference arise since in air there was nitrogen blanket formation in cathode catalyst layer that hindered the accessibility of reactant oxygen molecules to the platinum catalytic sites.



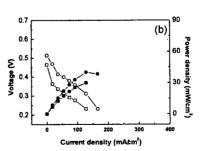


Fig. 1 Single cell performance of DMFC with 20 wt% Pt/C with and without CeO_2 under (a) oxygen 250 sccm (b) air 250 sccm [- \Box - \Box -0C20P, - \bigcirc - \bullet -1C20P]

Impedance studies were carried out at the potential regions of oxygen reduction reaction namely 0.7 and 0.9 V, the Nyquist plots are shown in Fig. 2. The spectra show a single arc and the size decreases for CeO₂ doped Pt/C compared to pure Pt/C at all potentials.

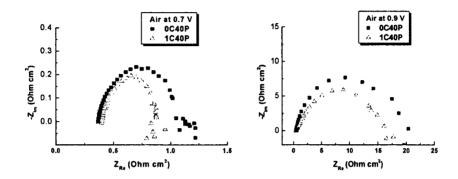


Fig 2. Impedance spectra of cathode with OC40P and 1C40P in air at different potentials

Polarization resistance observed at low potential regions is generally related to mass transfer process [4], and hence a decrease in resistance at low potential (0.7 V) was attributed to easier and more facile mass transfer of oxygen molecules to Pt active sites for the ceria doped Pt/C catalyst.

4. Conclusions

The incorporation of oxygen storage material such as ceria (CeO₂) into cathode catalyst layer may increase the local oxygen concentration, leading to enhancement of the single cell performance of DMFC. The resistance of ceria doped Pt/C electrode is smaller than normal Pt/C electrode in air operation especially at low potentials as found in impedance spectra, indicating the easier and more facile mass transport of reactant oxygen molecules to reactive platinum sites on ceria addition. 1wt% CeO₂ doped 40%Pt/C is found to be an optimum catalyst composition exhibiting highest performance in air atmosphere. Higher loading of ceria may result in adverse effect on

cell performance by decreasing the active surface area of platinum catalyst particulates and by increasing resistance in the catalyst layer due to poor electronic conductivity of ceria.

References

- 1. S. Wasmus and A. Küver. J. Electroanal. Chem, 46, 14, (1999).
- 2. R. Dillon, S.Srinivasan, A.S.Aricò and V.Antonucci, *J. Power Sources*, 127, 112, (2004).
- 3. G. Balducci, M. S. Islam, J. Kaspar, P. Fornasiero, M.Graziani, *Chem. Mater.* 12, 677 (2000).
- 4. A.J. Bard and L.R. Faulkner, *Electrochemical Methods Fundamentals and Applications*, 2nd Edn, John Wiley & Sons, New York, p. 385, (2001).