

# Characterization of direct ethanol fuel cell (DEFC) fabricated with porous SiC ceramic membrane

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## ABSTRACT

Direct ethanol-proton exchange membrane fuel cells have been fabricated with ceramic membranes. A porous silicon carbide (SiC) membrane having approximately 30% has been applied for application in a direct ethanol proton exchange membrane (DE-PEM) fuel cell. A horizontal type cell having Pt (40 vol.)/C catalyst layer on both side of the ceramic membrane was used for the demonstration test. The ethanol oxidation based-fuel cell stack showed very high voltage (1.289V) and measurable current level (1.10A) even though at room temperature. Even if, the porous ceramic membrane was utilized for the direct ethanol-proton exchange membrane fuel cell, very low permeability of the ceramic membrane for ethanol as compared to the conventional polymer electrolyte would occur. It was inferred the porous ceramic membrane, the current material rarely exhibit alcohol cross-over to the cathodic electrode greatly decreasing cell efficiency.

**Keywords:** Direct ethanol fuel cell (DEFC), Ceramic membrane, Membrane electrode assembly (MEA)

## 1 INTRODUCTION

Direct Ethanol Proton Exchange Membrane Fuel Cells (DEPEMFCs) are promising candidates as power sources especially at small-scale applications. The last decade polymer electrolyte membrane fuel cells (PEMFCs) directly fed by ethanol have been receiving more and more attention, due to their advantages despite highly advanced-direct methanol fuel cell technology [1]. Direct ethanol fuel cells (DEFCs) belong to the family of proton exchange membrane fuel cells (PEMFCs), in which ethanol is directly used as the fuel. It is widely recognized that proton exchange membrane fuel cell (PEMFC), known also as solid polymer electrolyte fuel cell (SPFC), possesses the advantages of quick start-up due to its low working temperature, compactness, no corrosion problems and flexibility at any orientation.

Over the last years, several experimental works [2-5] have been devoted to the direct use of ethanol in fuel cells (DEFCs). Almost all the above-mentioned scientific works deal with the problem of ethanol electro-oxidation kinetics at the anode compartment, and the research conducted in

order more effective catalysts to be found. However, due to the fact that for the complete ethanol electro-oxidation 12 electrons must be exchanged, it makes the oxidation kinetics inherently slower for a DE-PEMFC in contrast to hydrogen PEMFC.

Considering that H<sub>2</sub> is not a naturally existing gaseous fuel, to date, most of hydrogen used for PEMFCs operation is from the external reforming process, leading to a bulkier and heavier system and increasing simultaneously its complexity and cost. Moreover, although the lower operation temperature is one of the advantages of PEMFC, the fuel requirement is very rigorous.

Once the CO content in the hydrogen-rich gas is more than 10 ppm, an obvious decay in the cell performance will be observed. All of these problems and issues stimulate scientists and researchers to use directly the liquid fuels to feed fuel cells, avoiding in this way the difficulties and hazards associated with handle, storage, transportation and distribution of gaseous reactants such as hydrogen.

This can also provide considerable weight and volume advantages over the external reforming PEMFC. Among all the investigated possible fuels up to a few years ago, methanol was the most favorite due to its relatively higher electrochemical activity compared to the other liquid fuels and much progress has been made in the different pertinent issues, including electrocatalysts, electrolyte, membrane electrode assembly and fuel cell stack of the PEMFC directly fed by methanol (DMFCs).

Methanol is toxic for human beings and specially for optical nerve. Moreover, the sluggish anode kinetics and methanol crossover are still the main challenges to DMFC's commercialization despite that extensive efforts have been devoted. During the attempts to identify fuels for fuel cells, ethanol is considered to be an attractive and promising one at least for the following reasons: (i) its non-toxicity, (ii) natural availability, (iii) renewability, (iv) a higher power density, and (v) zero green-house contribution to the atmosphere.

The major problem in the fabrication and commercialization of DEFCs is connected to the swelling and distortion of the polymer membrane. The present polymer membrane should be replaced by porous SiC

ceramic membrane. The establishment of the DEFC membrane technology would be able to applied to the field of small electronic devices and their power source. DEFC system have some merits in the view of fuel supply & storage as well as replacement compared to other fuel cells. DEFC system shows low operation temperature and high current density as well as rapid starting and response to load variation, the utilization and dissemination of DEFC system in small or portable electronic devices is expected through this research program [6-8].

The present research is to develop a porous ceramic membrane as an important part in direct ethanol fuel cell (DEFC) application using silicon carbide material. The present polymer electrolyte membrane has some problems such as a swelling and a distortion with a geometrical resistance in the crossover of ethanol. The objective of this research is to optimize the core technology of the porous ceramic membrane to substitute the present membrane and synthesize the constituent material of the DEFC. This research is to produce the membrane electrode assembly (MEA) using silicon carbide (SiC) membrane, catalyst, anode and cathode and characterize the efficiency and durability of DEFC system.

## 2 EXPERIMENTAL PROCEDURE

Commercially available  $\alpha$ -SiC powders (mean sizes of 10  $\mu\text{m}$ ) were adopted as the starting materials for the fabrication of porous SiC ceramics. During the heat treatment process, silicone resin experienced an organic-inorganic transformation and acted as the bonding material between SiC particles at a low temperature of 1000 $^{\circ}\text{C}$ . The powders were ground and passed through a 100 mesh sieve and then were pressed under 15 MPa. The green specimen was calcined at 250 $^{\circ}\text{C}$  for 6 hrs in the oven, and then fired at 1000 $^{\circ}\text{C}$  for 1hr under nitrogen atmosphere.

A porous silicon carbide (SiC) membrane having approximately 30% has been applied for application in a direct ethanol proton exchange membrane (DE-PEM) fuel cell. A horizontal type cell having Pt (40 vol.%) / C catalyst layer on both side of the ceramic membrane was used for the demonstration test.

## 3. Result and discussion

### 3.1 SiC membrane and DEFC stack

This research is to produce the membrane electrode assembly (MEA) using silicon carbide (SiC) membrane, catalyst, anode and cathode. Fig. 1 shows XRD pattern and FE-SEM morphology of porous SiC ceramic membrane.

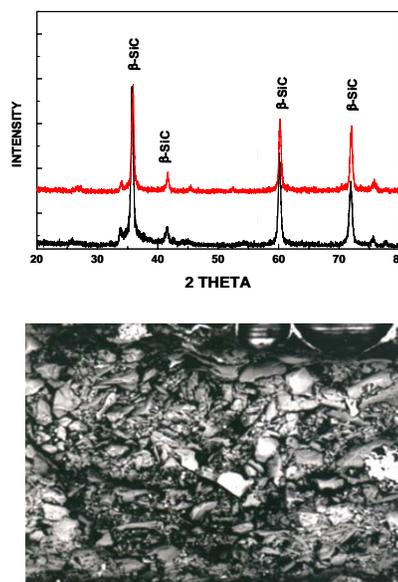


Fig. 1. XRD pattern and FE-SEM morphology of porous SiC ceramic membrane.



Fig. 2. Porous SiC ceramic membrane for DEFC.

Fig. 2 shows image of the porous SiC ceramic membrane. A horizontal type cell having Pt 40 vol.% / C catalyst layer on both side of the ceramic membrane was used for the demonstration test. Fig. 3 shows the DEFC stack fabricated with SiC ceramic membrane. The loading amount of Pt catalyst was about 40 vol. %. A porous silicon carbide (SiC) membrane having approximately 30% has been applied for application in a direct ethanol proton exchange membrane (DE-PEM) fuel cell.

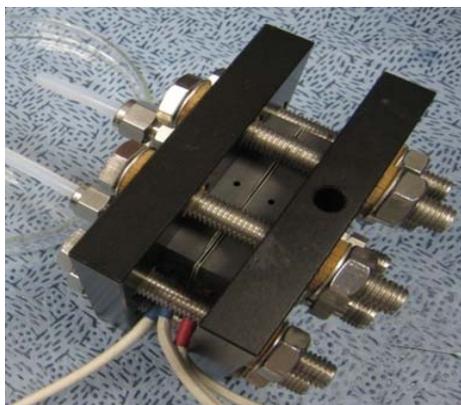


Fig. 3. DEFC stack fabricated with SiC ceramic membrane.

The working principle of direct ethanol fuel cell (DEFC) can be described in equation (1-3). The ethanol-water mixture aqueous solutions are fed to the anode compartment, where with the aid of the electrocatalysts, ethanol is oxidized to produce carbon dioxide releasing simultaneously protons and electrons. Protons are then transported to the cathode through the electrolyte and electrons flow through an external circuit, and then arrive at the cathode (Fig. 4).

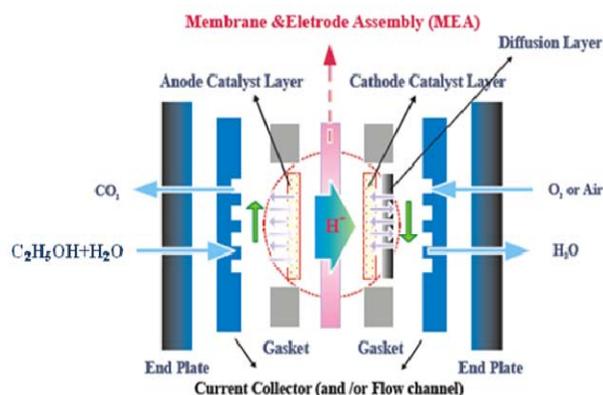
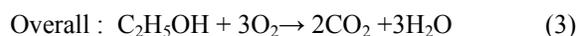
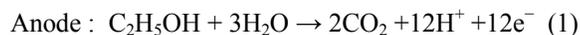


Fig. 4. Schematic of direct ethanol fuel cell.

The diffusion layer is made by porous and electrically conductive material, through which, the electrons generated in the anode catalyst layer are transported to the current collector.

The catalyst layer is the place where the ethanol electro-oxidation takes place, and in the ideal case of the complete electro-oxidation of a molecule of ethanol, 12 protons and 12 electrons would be released. There, the oxidant (always air or pure oxygen) reacts with the protons and electrons transported from the anode to produce water



However, in DEFCs below 100 °C the electro-oxidation of ethanol does not proceed all the way to carbon dioxide (CO<sub>2</sub>), but rather to acetaldehyde (CH<sub>3</sub>CHO), acetic acid (CH<sub>3</sub>COOH), and CO<sub>2</sub> depending on the nature and structure of the catalyst used, the applied potential, and the temperature field established. Acetaldehyde was reported to be the main product of ethanol oxidation in early investigations

### 3.2 Performace of ethanol fuel with aqueous

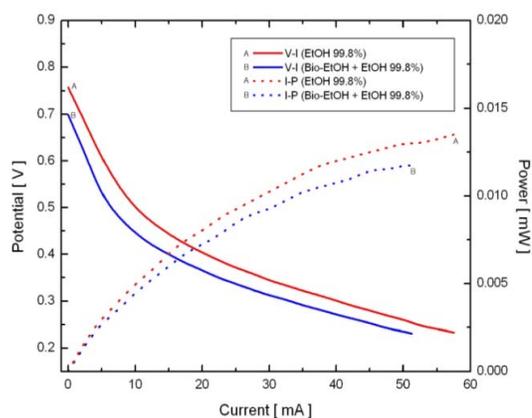


Fig. 5. Performance of ethanol fuel with ethanol fuel.

Fig. 5. shows the voltage-current and power-current of direct ethanol fuel cell. The voltage drop of the direct ethanol fuel cell was observed with increasing current in Fig 5. The sufficient feeding of aqueous ethanol solution into direct ethanol fuel cell stack exhibited very high voltage and high current level of 1.289 V, 1.10A using simple multi-meter. The excess fuel feeding would induce some problems of alcohol cross-over and accumulation of liquid fuel in membrane.

It should be noted that the adsorption and decomposition of ethanol and its intermediate reaction products happen on Pt active sites, while the dissociative adsorption of water occurs over Sn sites to form oxygen-containing surface species.

It provides the active surface and plays the role of reactants and products transportation channels and simultaneously the electron and proton passages. The electrolyte membrane acts as the proton carrier, and the separator of the anode and the cathode compartments.

Furthermore, considering that ethanol is a liquid fuel, the anode electrode structure is much different from the conventional gas-diffusion type. It should be very porous and capable of all being wetted by the liquid solutions, and simultaneously provide the pathway for effectively

releasing carbon dioxide evolved at the anode. On the other hand, the liquid solution at the anode could create the problem of the catalyst layer readily peeling off from the solid polymer electrolyte (SPE) membrane, i.e. “electrode delamination”, due to the different swelling degree between SPE and the electrode.

Hence, this would inevitably result in the discontinuity in the electronic and ionic conductivity, consequently leading to an increased cell internal resistance and irreversibly decreased cell performance and longevity.

A desirable electrode should be prepared in a way that can both maximize the active surface area per unit of the electrocatalyst mass and per unit of electrode area, minimize the obstacles to reactants/products transport to/from the catalyst and exhibit stable desirable performance during the cell’s life test under the actual operation conditions. This study is concerned with fabricating new membrane electrode assembly (MEA) showing active Pt/C catalyst layer, porous anode electrode and porous ceramic membrane.

However, there is no report about the modification or adopting new electrolyte to avoid or at least decrease ethanol crossover to some degree.

From the fundamental research to the development of electrode materials and fuel cell stack assembly until the introduction of fuel cell into commercialization, the investigation on all possible factors affecting the DEFC’s performance and stability are just at their very initial stage, and there is still much room to improve for direct ethanol PEMFC fuel cell development. In order to realize the direct use of ethanol as the fuel for fuel cells, the breakthrough in the electrolyte and electrocatalyst development is the rate-determining step.

The ethanol oxidation based-fuel cell stack showed very high voltage and measurable current level even though at room temperature. Even if, the porous ceramic membrane was utilized for the direct ethanol-proton exchange membrane fuel cell, very low permeability of the ceramic membrane for ethanol as compared to the conventional polymer electrolyte would occur. It was inferred the porous ceramic membrane, the current material rarely exhibit alcohol cross-over to the cathodic electrode greatly decreasing cell efficiency.

## 4 CONCLUSION

A porous silicon carbide (SiC) membrane having approximately 30% has been fabricated for application in a direct ethanol proton exchange membrane (DE-PEM) fuel cell. A horizontal type cell having Pt (40 vol.)/C catalyst layer on both side of the ceramic membrane was used for the demonstration test. The ethanol oxidation based-fuel

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## REFERENCES

- [1] J. Larminie, A. Dicks, *Fuel Cell Systems Explained*, 2nd ed., Wiley, 20-50, 2003.
- [2] S. Song, P. Tsiakaras, *Appl. Catal. B: Environ.* 63, 187–193, 2006.
- [3] Z. Wang, G. Yin, J. Zhang, Y. Sun, P. Shi, *J. Power Sources* 160, 37–43, 2006.
- [4] W. Zhou, Z. Zhou, S. Song, W. Li, G. Sun, P. Tsiakaras, Q. Xin, *Appl. Catal. B: Environ.* 46, 273–285, 2003.
- [5] X. Zhao, L. Jiang, G. Sun, S. Yang, B. Yi, X. Qin, *Chin. J. Catal.* 25, 983–988, 2004.
- [6] E. Spinace, M. Linardi, A. Neto, *Electrochem. Commun.* 7, 365–369, 2005.
- [7] F. Colmati, E. Antolini, E. Gonzalez, *J. Power Sources* 157, 98–103, 2006.
- [8] F. Colmati, E. Antolini, E. Gonzalez, *Appl. Catal. B: Environ.* 73, 106–115, 2007.

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