

Proceeding

ISBN 978-979-18845-2-5

**THE 3<sup>rd</sup> SRIWIJAYA INTERNATIONAL SEMINAR ON  
ENERGY SCIENCE AND TECHNOLOGY 2010  
(SISEST - 2010)**

**Theme :**  
**"New and Renewable Energy Development  
for Solving Energy Crisis"**

**PALEMBANG, 3 - 4 NOVEMBER 2010**

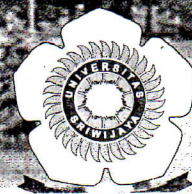
**SRIWIJAYA UNIVERSITY  
SOUTH-SUMATERA - INDONESIA**

**Organized by:**

**The National Strategic Prime Research  
(New & Renewable Energy Development)  
Rusnas PEBT Sriwijaya University**

**Sponsored by:**

**RISTEK**



**BPPT**



ISBN :978 – 979 18845 – 2 – 5

**PROCEEDING  
THE 3<sup>rd</sup> SRIWIJAYA INTERNATIONAL  
SEMINAR ON ENERGY SCIENCE AND  
TECHNOLOGY 2010 (SISEST 2010)**

**3-4 NOVEMBER 2010  
SRIWIJAYA UNIVERSITY  
PALEMBANG, SOUTH SUMATERA  
INDONESIA**

**Organized by :**

Lembaga Pengelola Riset Unggulan Strategis Nasional (RUSNAS)  
Pengembangan Energi Baru dan Terbarukan Universitas Sriwijaya (The  
National Strategic Excellence Research New and Renewable Energy  
Development Sriwijaya University)

## Catalyst Effect of Pt and Co to Conductivity, Morphological and OCV of Membrane Electrode Assembly (MEA) for Proton Exchange Membrane Fuel Cell (PEMFC)<sup>1</sup>

Dedi Rohendi

e-mail: rohendi\_dedi@yahoo.co.id

### ABSTRACT

*The research has been done about electrode and Membrane Electrode Assembly (MEA) manufacture for Proton Exchange Membrane electrolyte Fuel Cell (PEMFC) with Casting method using a catalyst of Pt-Co/C result of impregnation with Pt and Co content varies. The resulting electrode was characterized conductivity of surface and cross section and morphology using SEM-EDX. MEA is made by attaching electrodes on Nafion membrane electrolyte and conductivity of cross section was then characterized and tested its performance using a simple fuel cell equipment. The results of conductivity measurement of electrodes on the concentration of platinum (Pt) remains and Cobalt (Co) varied showed that the conductivity increased with increasing concentration of Co, as well as the Pt concentration varies and Co remains. EDX-SEM observations on the electrode surface showed that the contents of Pt on the electrode Pt-Co/C was detected, PTFE and carbon dominate and cobalt was not detected in the operating potential. Meanwhile, OCV of MEA analysis results at concentrations of Pt remains and Co varied to show the highest OCV of the sample MEA at contents of Pt:Co = 0.6:0.6 mg/cm<sup>2</sup> is 0.871 volts, while OCV with the concentration of Pt is decreased with a fixed Co concentration OCV was not decreased significantly.*

*Key words : Pt-Co/C, impregnation, MEA, PEMFC*

### INTRODUCTION

Proton Exchange Membrane Fuel Cells (PEMFC) is the equipment that produces electrical energy from an electrochemical reaction between hydrogen gas as fuel and oxygen as the oxidant with the aid of proton exchange membrane as an electrolyte and catalyst layer

contained in electrode (Wang, Zuo et al. 2009). PEMFC is widely regarded as one of the promising energy conversion devices for automobiles, residential and portable power systems for power density and high energy conversion efficiency, as well as non-pollution (Du, Wang et al. 2009).

The development of fuel cell was stagnant when the basic design has reconfiguration. The most significant barrier that PEM fuel cells had to overcome was the costly amount of platinum required as a catalyst. The large amount of platinum in original PEM fuel cells is one of the reasons why fuel cells were excluded from commercialization. Thus, the reconfiguration of the PEM fuel cell was targeted rather directly on the electrodes employed and, more specifically, on reducing the amount of platinum in the electrodes. This continues to be a driving force for further research on PEM fuel cell electrodes (Litster and McLean 2004).

One important component of the PEMFC is the electrode, which is a part where the electrochemical reaction occurs. Electrodes consisted of the cathode (where reduction reaction occurs) and anode (where oxidation reaction occurs). From the several methods of making electrodes, casting method is a method of making electrodes that are easy, cheap with the efficient use of materials and satisfactory performance (Harris and Walczyk 2006). Casting Method is done by attach electrode material on the substrate.

Important parameters of the catalyst layer performance is the ability to facilitate the electrochemical reaction and to stream of electrons generated at the anode or required at the cathode. Electrical conductivity of the electrode is one important parameter to determine the



performance of electrodes, which electrodes are arranged on the backing layer, Gas Diffusion Layer and Catalyst Layer must can stream and passes electrons generated at the anode. Several methods have been done to improve the electrical conductivity, among them by Lin (Lin, Chen et al. 2008). Between the cathode and anode are membrane electrolyte, and a combination of electrodes with an electrolyte membrane formed Membrane Electrode Assembly (MEA).

Catalysts are used to facilitate the reaction of oxygen and hydrogen. Selection of alloy catalyst Pt-Co/ C results of the reduction are due to platinum (Pt) alone has the properties that easily poisoned, oxidized, and the price is relatively expensive so the use of other metals from the class of transition metals such as cobalt (Co) to support the role of Pt as a catalyst to be important. The use of metal alloys as catalysts Fuel Cell proven to improve electrode performance, especially cathode (Seo, Lee et al. 2006).

## EXPERIMENT

### 1. Preparations of Pt-Co/C Catalyst

Catalyst Pt-Co/C prepared by impregnation and reduction of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  and  $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$  on the carbon substrate. Impregnation and the reduction is intended to distribute the particles of Co and Pt on the carbon substrate. Catalyst impregnation process was carried out with the mass ratio of Pt and Co are varied. Results of impregnation catalysts were reduced by using  $\text{H}_2$  at a temperature of  $400^\circ\text{C}$  for four hours.

### 2. Preparations of Electrode

Preparation of electrodes covering the stages:

#### a. Preparations of Backing Layer (Ismail, Rohendi. 2003)

Backing layer used was backing layer with composition ratio of carbon (50% wt) and

PTFE emulsion (50% wt) with a size of 2 cm x 2 cm.

#### b. Preparations of Reaction Layer and Casting to Backing Layer (Ralf, et al. 2002)

A certain amount of PTFE powder (20% wt) was dissolved by adding little by little cyclohexane, and stirred until homogeneous. Into the PTFE paste is added a certain amount of active carbon 200 mesh and Pt and Co catalysts with varying content. The mixture was stirred until homogenous to form paste and taped over the backing layer to form electrodes. Electrodes are formed is heated at a temperature of  $350^\circ\text{C}$  for 30 minutes, then cooled and dipped into a mixture of 15% Nafion solution and ethanol. Electrodes are then heated in an oven at a temperature of  $80^\circ\text{C}$  for 30 minutes. Electrodes that have been made analyzed the value of surface conductivity and cross section and characterized the structure and content of the particle surface by EDX-SEM.

### 3. Preparations and Testing of MEA ( Membrane Electrode Assembly)

Nafion membrane was activated by heating for 30 minutes in 1M HCl solution. On both sides of the membrane that has been activated and stretched, placed on one side of the anode and cathode on the other side. MEA is pressed on the appliance press by heating at 75 kg/cm<sup>2</sup> real pressure for 20 minutes. MEA formed was tested the elektrochemical activity on a simple fuel cell equipment.

## RESULTS AND DISCUSSION

### 1. Preparation of Electrode

#### a. Observation Results of Electrodes Conductivity

Observation Results of surface and cross section conductivity of reaction layer on electrode is shown in figures 1 and 2.



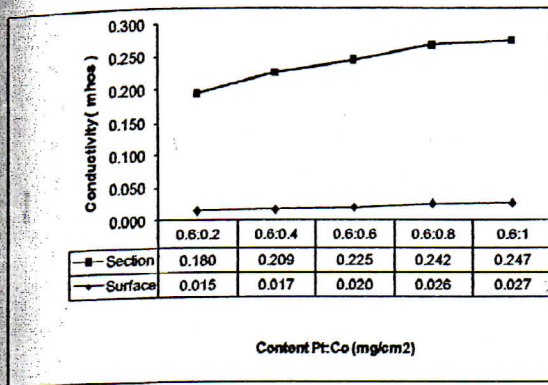


Figure 1. Comparison of surface and cross-section conductivity of the catalyst/reaction layer with fixed Pt and varied Co concentration

Figure 1 shows the contribution of Co catalyst as the supporting Pt catalyst in improving the conductivity of the surface and cross-section reaction layer on the electrode. The higher concentration of Co is added to the Pt concentration fixed, the conductivity increased. This can be understood because the conductivity of Co ( $0.1786 \times 10^8 \text{ Ohm}^{-1} \cdot \text{m}^{-1}$ ) greater than the conductivity of Pt ( $0.0952 \times 10^8 \text{ Ohm}^{-1} \cdot \text{m}^{-1}$ ). Seen a significant increase occurred in the concentration of Co  $0.8 \text{ mg/cm}^2$ . The picture above also shows that the more concentration of Pt and Co were added then the resulting conductivity will increase. The difference is that the conductivity of the electrode cross section is greater than the conductivity of the surface. This is caused by the cross-section measuring electrode distance is shorter (distance measuring electrode according to the thickness of about  $0.75 \text{ mm}$ ) so that the obstacles are smaller and produce a greater conductivity than the surface electrodes on the surface of measuring distance is longer (distance measuring  $1 \text{ mm}$ ).

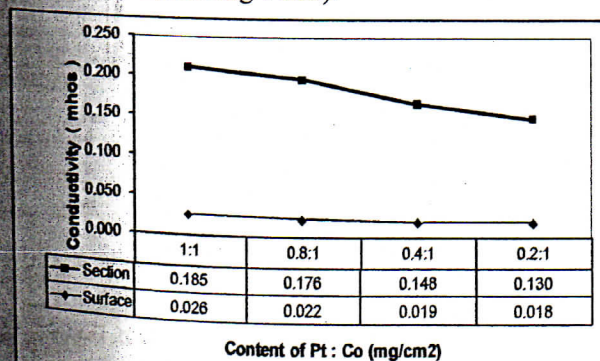


Figure 2. Comparison of the surface and cross-section conductivity of catalyst layer with Pt varied and Co fixed concentrations

Figure 2 shows that at a decreasing Pt concentration and the concentration of Co remains, its conductivity decreases. Figure 2 shows the contribution of Pt catalyst in determining the conductivity of the surface reaction layer on the electrode.

Figures 1 and 2 show the role of each of Co and Pt is significant in contributing to the conductivity of the reaction layer even though the value is not cumulative when compared with the conductivity value respectively.

b. Observation Results of EDX-SEM

Observation analysis using SEM-EDX is intended to look at the surface morphology structure as well as the content of the catalyst particles. EDX data from the surface of the electrode catalyst Pt-Co/C in the form of spectra and SEM images of electrode surface is presented in figure 3 and for comparison, The EDX and SEM data of the electrode surface of the catalyst Pt/C and Co/C is presented in figure 4 and 5.



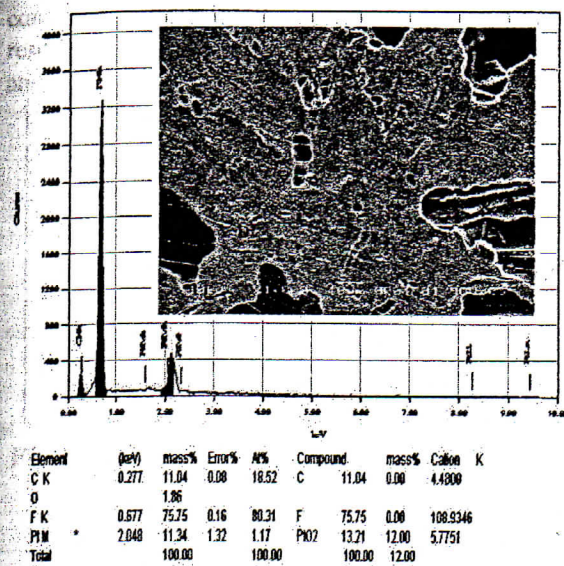


Figure 3. Spectrum data of The EDX-SEM of the Pt-Co/C catalyst with the content comparison of Pt:Co = 0,6:0,2

Figure 3 above shows the percentage data of Pt were detected at 11.34%, C of 11.04% and F of PTFE dominating (75.75%). Meanwhile, Co is not detected by the operating potential applied. This is because the amount of Co is added too little (0.2 mg/cm<sup>2</sup>). The emergence of F in large quantities are very possible because less than perfect mixing between the catalyst Pt-Co/C with PTFE so that the distribution of Pt-Co/C and PTFE uneven.

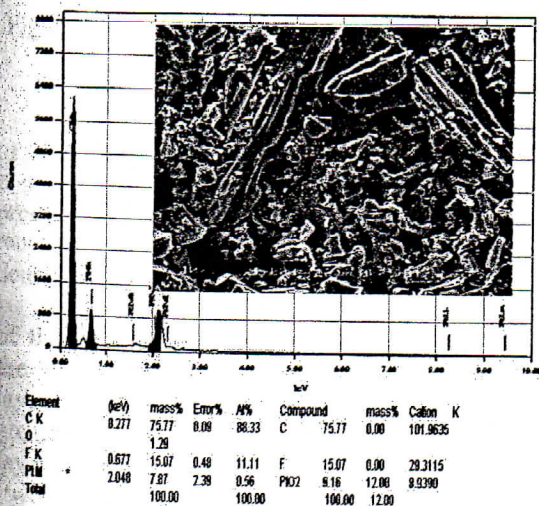


Figure 4. Spectrum data of the EDX-SEM of the Pt/C catalyst with the content comparison of Pt : C = 1: 0,40

Figure 4 shows the content of Pt and C in the form of the detected elements are 7.87% and 75.77%. Content of C looks dominant, this is because content of C that is used is greater than the Pt content and distribution between Pt, C and PTFE are uneven. The picture above also shows the existence of F elements are derived from PTFE. Meanwhile, from SEM images of catalyst Pt / C with magnification 1000x visible grain C is so dominant and the white light images granular that aggregated together with PTFE and carbon.

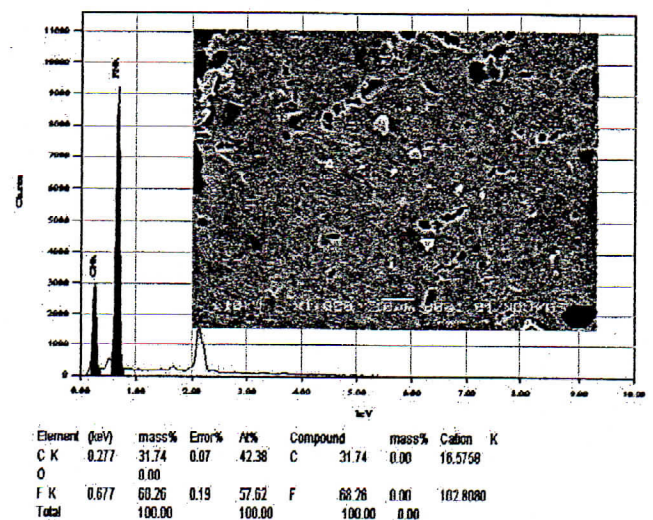


Figure 5. The Spektrum data of EDX-SEM of Co/C catalyst with the content comparison of Co:C = 1:40

Figure 5 showed significant of the PTFE content (68.26%). EDX spectrum data also indicate that the Co remains undetected at the content 1 mg/cm<sup>2</sup>.

2. Observation Results of MEA
  - a. Conductivity Observation

MEA conductivity measured is the conductivity cross-section (thickness) which is a combination of conductivity cross-section of two electrodes (cathode and anode) and the



conductivity of Nafion membrane. Measurement results of MEA conductivity shown in figure 6 and 7.

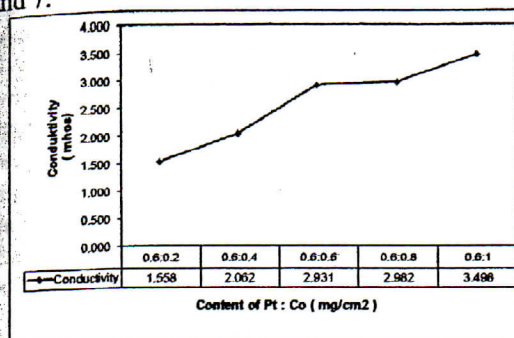


Figure 6. Comparison of MEA conductivity with fixed Pt contents and Co varied.

Figure 6 above shows that the conductivity of MEA produced increases with increasing Co content. This is related to the conductivity of Co is high enough.

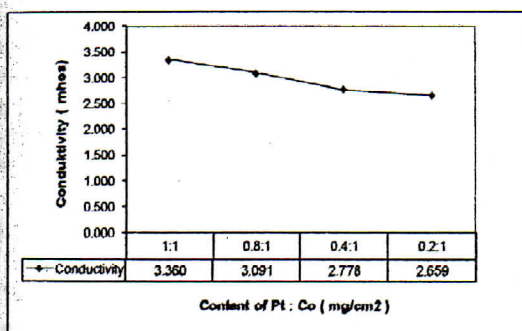


Figure 7. Comparison of MEA conductivity with varying contents of Pt and Co remains

Figure 7 above shows that the conductivity of MEA produced increases with increasing Pt concentration. Comparison of conductivity produced in the picture above does not differ much, because its structure is almost the same density.

Existing data indicate that the conductivity of the cross-section MEA is much smaller than the conductivity of the cross section reaction layer. This can be understood because of the greater thickness factor of the MEA than reaction layer and the many factors supporting layer MEA (two electrodes and an electrolyte membrane layer).

b. The Data of Open Circuit Voltage (OCV) of MEA

OCV of MEA with various catalyst composition measures at the same temperature and time. OCV of the MEA shown in figure 8 and 9.

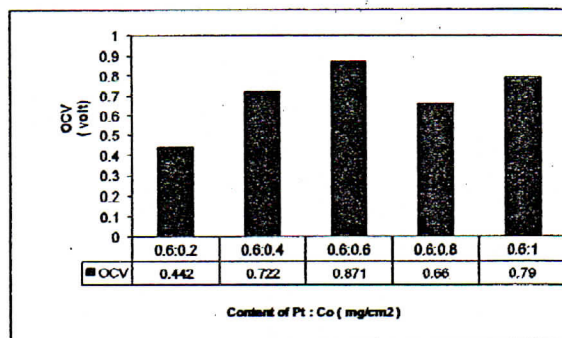


Figure 8. Comparison OCV of MEA at fixed Pt contents and Co varied

Figure 8 shows that the highest OCV of MEA samples with contents of Pt : Co = 0.6 : 0.6 mg/cm<sup>2</sup>. What is interesting is the potential of cells with contents of Pt : Co = 0.6 ; 0.8 mg/cm<sup>2</sup> and 0.6 : 1 mg/cm<sup>2</sup> concentration decreased in comparison with Pt : Co = 0.6 : 0.6 mg/cm<sup>2</sup>. This can occur because the distribution of Pt and Co that may not be evenly distributed, or overlapping, so that the active site can be blocked by the Co Pt. In addition to the distribution of catalyst, the density structure of the MEA and the MEA thickness also affect the resulting OCV.

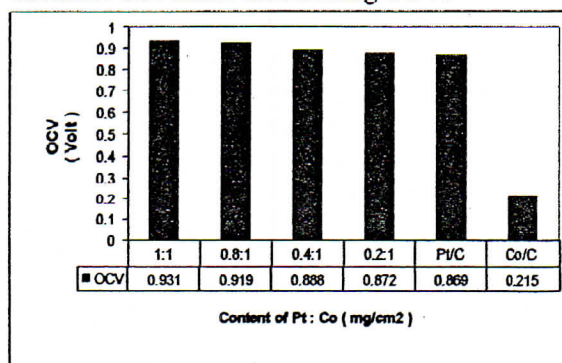


Figure 9. Comparison OCV of MEA at fixed Co contents and Pt varied

Figure 9 shows the role of catalyst of Pt and Co in the alloy catalyst Pt-Co/C. At Pt



concentration diminishing with fixed Co concentration, the cell potential decreased but not significantly. This shows that Co has a role in supporting the role of Pt as a catalyst. From the figure 9 shows that up to 0.2 mg/cm<sup>2</sup> Pt concentration with the concentration of Co = 1 mg/cm<sup>2</sup>, OCV value almost the same with Pt content = 1 mg/cm<sup>2</sup> without Co. For comparison are shown curves of Pt/C and Co/C, which indicates if Co is used as the sole catalyst (without Pt), OCV value so small. This indicates that Co is not suitable for use as a single catalyst.

### CONCLUSIONS

Pt and Co catalyst prepared by impregnation and reduction methods proven to increase the electrical conductivity of the surface and the cross-section electrode and cross-section of MEA. EDX-SEM results showed that the impregnation method is quite successful in placing Pt catalyst on electrode. Co as support of Pt catalysts in the PEMFC is proven to maintain the OCV while content of Pt reduced to 0.2 mg/cm<sup>2</sup>. The highest OCV ratio achieved on the content of Pt: Co = 0.6: 0.6 mg/cm<sup>2</sup>. Although Co in Pt-Co/C catalyst is proven to maintain the OCV of MEA, but the use of Co as the sole catalyst for PEMFC does not show good performance.

### ACKNOWLEDGEMENTS

Thanks and appreciation to the Directorate of Higher Education c.q DP2M which has been finance the research through Research Competitive Grant XIII.

### REFERENCES

- Du, C., B. Wang, et al. (2009). "Hierarchy carbon paper for the gas diffusion layer of proton exchange membrane fuel cells." Journal of Power Sources **187**(2): 505-508.
- Harris, T. A. L. and D. F. Walczyk (2006). "Development of a Casting Technique for Membrane Material Used in High-Temperature PEM Fuel Cells." Journal of Manufacturing Processes **8**(1): 8-20.
- Ismail, S., Rohendi, D. (2003). "Modification on Electrode System to Increase Performance of Polymer Electrolyte Fuel Cell

(PEFC)". Proceedings of Regional Symposium on Chemical Engineering, Metro Manila, Philipines.

- Litster, S. and G. McLean (2004). "PEM fuel cell electrodes." Journal of Power Sources **130**(1-2): 61-76.
- Ralf, Z., et al.(2002). "Gas Diffusion Structures and Gas Diffusion Electrodes for Polymer Electrolyte Fuel Cells." United States Patent Application No. 0041992.
- Seo, A., J. Lee, et al. (2006). "Performance and stability of Pt-based ternary alloy catalysts for PEMFC." Electrochimica Acta **52**(4): 1603-1611.
- Wang, Z.-B., P.-J. Zuo, et al. (2009). "Durability studies on performance degradation of Pt/C catalysts of proton exchange membrane fuel cell." International Journal of Hydrogen Energy **34**(10): 4387-4394.