CHAPTER 5

Conclusions

5.1. Variation of Nafion content and solvent types for MEA fabrication

In this research, the two main compositions of catalyst inks, solvent and Nafion solution, were investigated for MEA fabrication. The morphology of MEAs was characterized by SEM technique. The colloid solvent (i.e. THF) agglomerated catalyst particles generating a bigger cluster of catalysts and thinner catalyst layer which is the cause of lowest void space in catalyst layer. Solution solvents (i.e. IPA and Ethanol) assisted catalyst distribution generating more void space and thicker the catalyst layer. This improved mass transportation and Pt surface areas through catalyst layer resulting in the higher polarization of solution ink (IPA and Ethanol) compared to colloid ink.

The best polarization curves of fuel cell were obtained and depended on Nafion content and solvents. MEAs from Ethanol, IPA and THF solvents yielded each of the best performance at the Nafion contents of 20, 25 and 30 wt.%, respectively. The optimum Nafion content in each MEA provides the most utilization of catalyst in catalyst layer because the most catalyst surfaces are utilized to increase the reaction and consequently conduct the electrons releasing from the reaction. This also means the compromising mass (reactant gas and water) and ion (electrons and proton) transport in three phase regions (pores, catalyst cluster and Nafion webs). In the case of the insufficient Nafion content in the catalyst layers, the volume faction between the catalyst cluster and Nafion or pore and Nafion are increased. Although the electrons and reactant gas are easy to transport, protons have not enough Nafion to be their pathway thus this affects the uncompleted electrochemical reaction. The case of the excessive Nafion content, there are two possible problems. Firstly, the excessive Nafion content is possible to penetrate into the pores affecting the mass transport problem. Moreover Nafion favors to adsorb water, byproduct of the reaction. This is the cause of water flooding which are the

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hindrance for reactant gas penetrating to active site. Secondly, Nafion is possible to fully cover the active areas of catalyst cluster affecting the reduced catalyst utilization and pathway of electron.

When considering the MEA fabricated by THF, the thickness of catalyst layers from THF are thinnest and require the highest optimum Nafion content. This means that the compactness of Nafion and catalyst clusters in catalyst layer from THF are highest compared with that from IPA and ethanol. Thus, the pores and catalyst active areas of catalyst layers from THF are smallest. This result was also proved by determining the kinetic parameter and overall resistance from the fitting polarization curve at low and medium current density. The value of i_0 for each MEA were determined. The MEA fabricated by ethanol yielded the highest value of i_0 followed by that by IPA and THF respectively. This means the catalyst layer formed by ethanol has highest ECSAs because the value of i_0 directly relates with ECSAs. The overall resistance of MEA made by THF was highest due to the smallest ECSAs where are used to conduct electron.

The ethanol and IPA solvent which make the catalyst ink formed as solution ink gave the higher performance compared with colloid ink from THF. The comparison between catalyst layer morphology from IPA and ethanol was analyzed by referring from Ref. [17]. The size of Nafion agglomeration from ethanol ink is larger than that from IPA. This is because the value of $|\delta_{ethanol} - \delta_{Nafion\ side\text{-}chain}|$ is smaller than that of $|\delta_{IPA} - \delta_{Nafion\ side\text{-}chain}|$. This means that the ethanol solvent is more compatible with Nafion side-chain than the IPA solvent. The compatibility between Nafion side-chain and solvent results in the dissociation of negative charges ($-SO_3^-$) group from Nafion side-chain to bond with solvent molecules. The higher negative charges from the mixture of Nafion and ethanol provides the higher performance than the lower negative charges from the mixture of Nafion and IPA. Among these MEAs, MEA from ethanol provided the highest current density of 697.02 mA/cm^2 at cell potential of 0.6 V. Therefore, the solvent affects the MEAs both physical morphology and electrochemical reaction during fuel cell operation. Moreover, the optimal Nafion content depends on solvent to

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provide the best fuel cell performance. In this work, Ethanol shows the most balance of mass transport, Pt utilization and gives the best fuel cell performance.

5.2. Various Nafion contents in Pt/C concentration for MEA fabrications

The MEAs with Pt/C catalysts of 10%, 20%, and 40% by weight were fabricated directly onto electrolyte membrane using an ultrasonic spraying technique. At the same Nafion content and Pt loading, the morphology of catalyst layers were characterized and revealed that the lower Pt/C concentration produced larger secondary pores. Moreover, from the cross-section images, the thickness of catalyst layers depended on Pt/C concentration; lower Pt/C concentrations had thicker catalyst layers than the higher Pt/C concentration. The larger pores and thicker catalyst layers from the lower Pt/C concentration due to the lower bulk density and higher amount. The cell resistance in each MEA was also measured. The distribution of cell resistance also depended on catalyst layer thickness, and the thicker catalyst layer exhibited a broader distribution.

At an identical Pt loading of 0.3 mg/cm^2, the MEAs using Pt/C catalysts of 10%, 20%, and 40% by weight required a Nafion content of 25%, 20%, and 15% in catalyst inks, respectively, in order to have the best performance. Consequently, the optimal Nafion content depends on Pt/C concentration, in which the higher Pt/C concentration requires lesser amounts of Nafion (ionomer). The ECSA in each MEA was determined by CV technique to study behavior of polarization curve at activation loss region. MEA fabricated by 10%Pt/C had the highest ECSA at Nafion content of 35wt.% while that by 20%Pt/C and 40%Pt/C have the highest ECSA at Nafion content of 25wt.%. These results were not the same with the optimum Nafion content obtained from polarization curve at medium (ohmic loss region) and high current densities (concentration loss region). This can be attributed that the determining ECSA was performed at activation loss region. When focusing at low current density of polarization curve, the optimum Nafion contents in each MEA were the same trend with those from the data of ECSA. The optimum Nafion contents at activation loss region in each MEA were higher than those at ohmic and concentration loss regions. This can attribute that normally, the increase in Nafion content is beneficial to extend ECSA but it is drawback at medium

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and high current density because of higher production of water. The produced water at medium and high current density is more possible to be captured by Nafion and then occurring flooding phenomena blocking mass transportation. The effect of flooding phenomena was clearly observed at MEA fabricated by 10%Pt/C. The PEMFC performance of 10%Pt/C was improved with reducing RH from 95% to 65%. The determining the proper Nafion content in this work was compared with other studies. Regardless of the fabrication methods, the Pt loadings and the operating conditions, the correlation of Nafion content in this work agrees with other researchers. Therefore, it is important to account for the relationship between Pt/C concentration and Nafion as one of the main parameters for state of the art MEA fabrication and PEM fuel cells.

5.3. Recommendations for future work

5.3.1. To completely determine the most utilization in each catalyst types and to analyst in the economic aspect, the various Pt loading in each types of catalyst should be performed.

5.3.2. Normally, the resistances in PEMFC comprise of mass, ion and contact resistances but in this work, these resistances cannot be individually measured. Thus, the measurement used to evaluate each type of resistance should be applied for better analysis and understanding.

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