CHAPTER V CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

In this research work, a series of Au-based catalyst (Au/Fe₂O₃-TiO₂) have been used to catalyze the preferential CO oxidation reaction (PROX) in the presence of H₂ over a range of operating temperature of 40 to 180 °C. Several parameters on the catalytic behavior were investigated—including the effect of support composition, calcination temperature, amount of gold Au loading, and the effect of O₂ pretreatment—in order to find the optimum preparation conditions to produce the finest Au/Fe₂O₃-TiO₂ catalyst. In addition, the catalytic behavior of the best Au/Fe₂O₃-TiO₂ catalyst was then studied under the realistic reformate gas stream, which contained CO₂ and H₂O.

From the result demonstrated that the Au/Fe₂O₃-TiO₂ catalyst is highly active than Au/Fe₂O₃ and Au/TiO₂ catalysts. The higher activity of Fe containing catalyst was attributed to the ability to supply reactive oxygen, thereby stabilize active gold species (Au metallic or Au⁰) in the catalyst. The calcination process changes the chemical state and particle size of metal gold. There was considerable increase in CO oxidation with increasing calcination temperature. The catalytic activity at various reaction temperature in the range 40 to 180 °C showed that 1% Au/Fe₂O₃-TiO₂ (1:4) calcined at 400 °C exhibited the highest CO conversion of 94.99% and PROX selectivity of 56.46% at 60 °C, which could be applied in the optimum condition for PEMFC application. The reason behind this fact is that the 1% Au/Fe₂O₃-TiO₂ (1:4) catalyst calcined at 400 °C showed better gold dispersion on the support and high metal-metal interaction, as were confirmed by TPR and TEM characterizations. However, the catalytic activity were decreased with increasing reaction temperature. These results revealed that during CO oxidation reaction at high temperatures other reaction mechanism, such as H₂ oxidation occured.

Furthermore, when O_2 pretreatment was applied to these catalysts, CO conversion decreased from 94.99% to 44.47% at 60 °C. The reason behind this

phenomenon is that the surface oxygen species are formed on gold, then they are occupying reactive sites. Thus the strongly adsorbed surface oxygen species block the adsorption and diffusion of weak adsorbed surface oxygen result, resulting in a decrease in catalytic activity of the catalyst. And also the metal-metal interaction tend to decrease as evidenced by TPR result.

The 1% Au/Fe₂O₃-TiO₂ (1:4) calcined at 400 °C catalyst showed a good result in stability test for 16 hours and the catalyst was able to stand the presence of water up to 10% in the feed stream without any significant decrease in catalytic activity. However, when CO₂ (10%) was injected in the feed stream, the catalyst activity was negatively affected.

5.2 Recommendations

The catalysts are recommended to be prepared in the same batch in order to prevent any errors that may occur, as the catalytic performance is sensitive to many import factors.

The oxidation state of gold presented on the catalyst surface and catalytic performance is the important parameter which can determine the catalytic performance of the catalyst. Therefore the X-ray photoelectron spectroscopy (XPS) is recommended for further characterization, in order to explain the role of the oxidation state of Au on the catalytic behavior.

To accomplish 100% CO conversion and higher PROX selectivity, the mixed support by other techniques, such as photo-deposition, and co-precipitation might be interesting.