CHAPTER VIII
CONCLUSIONS AND RECOMMENDATIONS

8.1 Conclusions

In this work, the catalytic activities of Au-based catalysts were investigated and developed for the preferential CO oxidation reaction in the H_2-rich stream which was supplied from both the simulated reformate and the real methanol-steam reformate. Starting with the simulated reformate, the catalytic activities of AuPt/A zeolite catalyst for the PROX reaction in the single- and the double-stage reactors were studied in the temperature range of 50–300°C. The process efficiency can be enhanced by using the double-stage reactor which gained more process efficiency than that of the single-stage reactor in terms of CO selectivity. In the double-stage process, the effect of reaction temperature is more pronounced than the effect of oxygen split ratio. The best conditions were 170°C and 50:50 which are a reaction temperature in both stages and an oxygen split ratio, respectively. At this condition, it provided the maximum CO conversion combined with a high CO selectivity. Besides, the stability of the catalyst was tested at 170°C in a double-stage process without CO_2 and H_2O, which showed stable performance in the high catalytic activities without any degradation for 60 h. However, the active temperature range is too high for the PROX unit in application directly to on-board fuel processor. Thus, the development of catalyst has been carried out further.

It is well known that the catalytic activity of a Au catalyst is strongly related to the type of catalyst support, which brings about a great difference in Au particle size and interaction with the support. The Au catalyst was developed by using ZnO and Fe_2O_3 as catalyst supports. Our results showed that both the Au/ZnO and the Au/ZnO-Fe_2O_3 catalysts prepared by the deposition-precipitation technique exhibited high catalytic activities, where they achieved an almost complete conversion of CO to CO_2 at 50–70°C with 60–75% selectivity. Moreover, using the mixed-metal oxide support (ZnO-Fe_2O_3) resulted in improved resistance toward the presence of CO_2 and H_2O. Both catalysts were quite stable with time-on-stream. Additionally, the Au-

oxide interface plays an important role in the PROX activity. The deposition of Au on supports with Na_2CO_3 is an effective method for creating smaller Au particles, resulting in an increase of the number of Au-oxide interfaces, which enhances the catalytic activity for this reaction.

However, the deposition-precipitation technique involves many steps for preparing an active catalyst. To reduce these steps, the photodeposition technique was applied to deposit Au in nanometer size on the catalyst support. From the results, it can be concluded that Au/ZnO and Au/ZnO-Fe_2O_3 catalysts can be successfully prepared via the photodeposition under UV-vis light irradiation. Moreover, it revealed that the prepared catalysts (without heat treatment step) exhibited higher catalytic activity, where it achieves a complete conversion of the CO at 30°C and 50–73 % CO selectivity. The presence of a mixed oxidation state of Au is the active site for the PROX reaction. In addition, the mixed metal oxide support (ZnO-Fe_2O_3) can reduce the influence of CO_2, which is present in the simulated stream.

Finally, the fuel processor system consisting methanol steam reformer unit and PROX unit was construct. Methanol steam reforming took place at 250°C providing the highest CH_3OH conversion (100%) over ShiftMax 240, combined with 75% H_2 selectivity, and 65% H_2 yield. At high process performance of MSR, the CO content from MSR is lower than 1 vol%. Experimental results demonstrated that Au/ZnO-Fe_2O_3 prepared by the photodeposition technique can be utilized in a PROX unit under a realistic condition from MSR unit in order to remove the CO content to 0 ppm before feeding the pure H_2 fuel to PEM fuel cell.

8.2 Recommendations

Although the Au/ZnO-Fe_2O_3 prepared by the photodeposition technique showed an excellent activity, the active temperature range and the durability are still too low for being applied to an onboard fuel processor so that further catalyst development is needed. Type of metal oxide catalyst supports and a catalyst preparation technique (irradiation time and power of UV light) play an important role

for achieving the active catalyst that created nanosize Au particles deposit on that support, working on many kinds of metal oxide is also promising.