

CHAPTER V

CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

In this research work, the catalytic activities of Au-based catalyst (Au/CeO₂-ZrO₂) were investigated for the preferential CO oxidation reaction (PROX) in the presence of H₂-rich stream over the temperature range of 50 °C to 190 °C. The catalysts were supplied in a simulated and realistic reformat gas stream in order to study the catalytic behavior. Moreover, CO oxidation and water gas shift reactions were also investigated and compared with preferential CO oxidation reaction. Then the stability was tested in the simulated dry condition (without H₂O + CO₂) and wet condition (with H₂O + CO₂).

The results showed that the catalytic activity of Au/CeO₂-ZrO₂ catalyst was related to the atomic ratio of the supports, which the activity increased with decreased the Zr content. The 1 wt% Au/Ce_{0.75}Zr_{0.25}O₂ catalyst exhibited the highest catalytic activity of 94.06% CO conversion and 49.09% PROX selectivity at 50 °C, which the activities decreased with increasing reaction temperature. These results revealed that it was favorable H₂ oxidation at high temperature reaction. The incorporation of Zr enhanced the catalytic activity due to the formation of solid solution, which increased the amount of oxygen vacancies and active oxygen species, as were confirmed by XRD and FT-Raman characterizations. It can be attributed to the synergetic effect between the support and gold particles at the interface. From TPR analysis, the reduction peak shifted to lower temperature in the presence of Au and it obtained a higher reducibility at low content of Zr, suggesting a higher concentration of surface oxygen in conversion CO to CO₂. The coexistence of Au⁰ and Au^{δ+} species could be observed and these species were active sites for the PROX reaction. Moreover, the 1 wt% Au/Ce_{0.75}Zr_{0.25}O₂ catalyst contained the highest amount of Au, corresponding with the activity. The presence of CO₂ and H₂O in H₂ stream from a reforming process is significant, and the effect on

catalyst performance was studied by addition of either CO₂ or H₂O to the feed stream. The presence of H₂O had insignificant effect on the catalytic performance, whereas the presence of CO₂ in the feed drastically induced a loss in the activity, indicating that CO₂ had a significantly negative impact on the catalytic activity. Furthermore, CO oxidation and water gas shift reactions were performed on Au/CeO₂ and Au/Ce_{0.75}Zr_{0.25}O₂ catalyst. The Au over mixed support produced better activity than the Au supported on pure support for both reactions, suggesting that CO oxidation was almost suppressed by the strong competition between H₂ and CO for adsorption in presence of excess of hydrogen. Doping with Zr could help to improve the thermal stability and resistance the sintering at high temperature reaction. In addition, the activity of both catalysts was exhibited excellent catalytic stability in the simulated dry condition, and then it drastically dropped in the wet condition. Nevertheless, they had the same activity as the fresh catalysts, confirmed by the XRD results that they have a little change in the crystalline size and not see the Zr segregation or gold sintering peaks.

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 chemical structure of Au is an important parameter to determine the catalytic performance of the catalyst, which X-ray photoelectron spectroscopy (XPS) is a technique well-known for characterizing in order to explain the role of the oxidation state of Au on the catalytic behavior.

The efficiency of catalyst for PROX reaction depends on various factors including, e.g., the size Au particles, preparation procedures, supported catalyst. Beside these factors, the bimetallic containing Au might be interesting.