CHAPTER V
CONCLUSIONS AND RECOMMENDATIONS

5.1 Conclusions

Bimetallic Au-Pt supported on A zeolite catalysts catalyze the preferential oxidation of CO in the presence of H_2 over the temperature range of 50-310°C had been studied in this work. Many effects on the catalytic activities are presented; including the effect of Au on the Pt/A zeolite performance, sequential metal-loaded, metal-loaded ratio, catalyst pretreatment, catalyst support and pore-size aperture. Additionally, the performance of the best catalyst was also investigated under the realistic reformate which containing CO_2 and H_2O. The important conclusions of this study can be summarized as follows:

TEM investigations indicated that larger gold crystallites were formed on the A zeolite surface. Au metal crystallites weakly interact with the Pt metal crystallites; therefore, Au metal did not form an alloy with Pt metal because two metals appear to be severely phase separated.

The catalytic activities of all prepared catalysts depended strongly on the metal crystallite size and the actual total metal loading. However, their catalytic activities not correlate with the surface area of the prepared catalysts, measured by BET surface area analyzer.

A small amount of Au was added to the Pt/A zeolite catalyst, the CO selectivity was improved at low temperatures. Additionally, the temperature at the maximum CO conversion of AuPt/A zeolite was shifted approximately 30°C to lower temperature.

The appropriate catalyst preparation method was co-impregnation method because the sequential order of metal loading did not show the significant effect on both CO and O_2 conversions. In addition, the appropriate catalyst pretreatment condition was the He pretreatment which achieved higher maximum CO conversion and higher CO selectivity. In addition, He pretreatment at 110°C was necessary to remove adsorbed species on the catalyst surface.

The AuPt/A zeolite catalyst containing the Au to Pt ratio of 1:2 gave the best performance which can completely remove CO from the H_2-rich stream and achieved a high selectivity. Moreover, the temperature at maximum CO conversion was also shifted to lower temperatures.

The catalytic activities of 1%(1:2)AuPt/commercial A zeolite catalyst were much better than 1%(1:2)AuPt/synthesized A zeolite catalyst because the small in crystallite size and better morphology of synthesized A zeolite which facilitated the metal dispersion to the deeper of the pore of A zeolite resulting in the temperature at the maximum CO conversion of 1%(1:2)AuPt/synthesized A zeolite was shifted 60°C to higher temperature.

The different pore aperture of A zeolite catalyst supports were used, including 3A zeolite, 4A zeolite and 5A zeolite. It was found that the CO conversions were in the order of AuPt/4A ~ AuPt/5A > AuPt/3A. The CO was completely converted to CO_2 by the AuPt/4A zeolite at 170°C while the AuPt/5A zeolite gave a maximum temperature at about 190°C. In contrast, the AuPt/3A zeolite could not achieve 100% CO conversion which gave only about 90% CO conversion. Additionally, the AuPt/4A zeolite and the AuPt/5A zeolite achieve a higher CO selectivity at low temperatures, but at high temperatures it achieves a lower CO selectivity than AuPt/3A zeolite. The AuPt/4A zeolite represented the best performance due to its high CO conversion in combination with a high CO selectivity.

Addition of CO_2 to the feed stream leads to negative effect on the CO conversion in the temperature range of 110-210°C. The effect of CO_2 is almost independent of the temperature because it has a positive effect on the CO conversion in the temperature range of 50-90°C and did not have significant effect on the CO conversion at high temperatures (230-310°C).

Addition of H_2O vapor to the feed did not has significant effect on the CO conversion but has significant positive effect on the CO selectivity at high temperatures compared to the preferential oxidation of CO in CO_2-containing feed stream, which is caused by suppressing the competing H_2 oxidation reaction, including promoting the WGS reaction and as a result more CO is converted to CO_2 at high temperatures resulting in an increase the CO selectivity. Additionally, a small

amount of Au was added to Pt/A zeolite catalyst resulting in the activities of the 1% (1:2)AuPt/A zeolite did not show much difference between the presence and the absence of H_2O vapor in the feed stream.

Under realistic condition, the presence of CO_2 and H_2O in feed stream had a negative effect on the CO conversion. However, the CO selectivity of the catalyst was not significantly impacted.

1% (1:2)AuPt/4A zeolite catalyst exhibited a stable catalytic performance during 12 hours of testing time, suggesting that no sintering or coke occurrence on the catalyst during the reaction performing.

5.2 Recommendations

The metal crystallite sizes strongly affected the catalytic activities; therefore, further study is necessary in order to improve the catalyst preparation method. The zeolites were used as catalyst supports which has a highly porous material and a good thermal stability that can provide a highly metal dispersion and a stable catalytic performance, working on many kinds of zeolite is also promising.