CHAPTER I INTRODUCTION

Recently, there is an increasing concern with environmental and energy consumption. Thus, renewable energy sources are the way to solve these problems such as solar energy, biomass energy, and hydrogen energy. Hydrogen is an attractive as energy carrier because it has the potential to contribute to the future energy, especially for polymer electrolyte membrane fuel cells (PEMFCs). PEMFCs are currently considered among the most advanced system for application of electric vehicles or residential power-generations. Because they have many advantageous features such as a low-operating temperature (80–120 °C), sustained operation at high current density, low weight, compactness, potential for low cost and volume, fast start up and suitability for intermittent operation (Patil, 1992).

Hydrogen is usually generated from hydrocarbons or alcohols by steam reforming, partial oxidation, or autothermal reforming in combination with the watergas shift reaction. The reformate stream generally contains around 1 vol% of CO from water gas shift reaction and this has a significant negative effect on the anodic Pt-electrode of PEMFCs. From this limitation, they require the CO removal to lower CO concentration below 10 PPM (Schmidt *et al.*, 1994). The most simplest method is the preferential oxidative of CO (PROX) due to a simple and cost-effective way (Haruta *et al.*, 1989). The PROX catalyst should have high oxidation rate, high selectivity with respect to undesired H₂ oxidation and stability with reaction time.

In the PROX process, CO oxidation (Eq.(1.1)) is the desired reaction while H_2 oxidation (Eq.(1.2)) should be avoided.

$$CO + (1/2) O_2 \longrightarrow CO_2 \qquad (1.1)$$
$$H_2 + (1/2) O_2 \longrightarrow H_2O \qquad (1.2)$$

At temperature higher than 150 °C, the oxidative reaction may be accompanied by reverse water-gas shift (Eq.(1.3)) and/or CO methanation (Eq.(1.4)) reactions.

 $CO_2 + H_2 \longrightarrow CO + H_2O$ (1.3)

 $CO + 3H_2 \longrightarrow CH_4 + H_2O$ (1.4)

Many literatures suggest that gold-based catalysts are very effective for the low-temperature selective CO oxidation when compared to Pt-based catalysts, which the range of temperature is suitable for PEMFCs. This application is based on Haruta's work that gold nanoparticles supported on reducible oxide supports are highly active for CO oxidation reaction (Haruta *et al.*, 1989). The activity of gold catalyst depended on the size of gold particles, strong contact between the gold particles and the support, and suitable selection of the support.

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Supported gold catalyst is one of candidate in intense studies for CO oxidation and many studies report that metal oxides are appropriate to be used as the supports because they could be partially reduced. Among supports showed, ceria (CeO₂) support is very interesting and it has been widely investigated for decades for this reaction. Another support as zirconia (ZrO₂), it seems to be an attractive one because it has high oxygen vacancies which increase the reducibility of the catalyst.

The purpose of this research is to study the activity of Au/CeO₂–ZrO₂ for preferential CO oxidation (PROX). In addition, the effects of support composition, composition of feed stream, side reactions (water-gas shift and CO oxidation), and deactivation test on the catalytic performance will be investigated. The catalytic activities are presented in terms of CO oxidation and selectivity toward CO oxidation. Various characterization techniques such as Atomic Absorption Spectroscopy (AAS), X-ray Diffraction (XRD), and Temperature-Programmed Reduction (TPR) will be carried out to understand the relationship of the catalyst properties, activity, and selectivity.