

## CHAPTER I

## INTRODUCTION

In recent times, more emphasis has been laid on the expansion of hydrogen energy because of its high-energy efficiency and clean burning properties. Hydrogen can be used directly as a fuel in internal combustion engines or indirectly to supply fuel cells. Widespread use of hydrogen fuel cell vehicles would reduce the usage of oil, thereby reducing environmental pollutants, such as oxides of nitrogen, sulfur and hydrocarbon, since hydrogen produces only energy and water vapor when utilized as a fuel. In addition, it can be utilized in proton exchange membrane fuel cells (PEMFC), in which electricity is generated by electrochemical oxidation of hydrogen across a proton conducting polymer membrane. With the development of the technology of polymer electrolyte fuel cells, a hydrogen fuel cell exhibits a thermal efficiency of up to 35-40%, in comparison with the 25-30%, which is typical of a normal petrol-fuelled engine.

The direct use of hydrogen for fuel cell application is hindered by problems of on-board storage of hydrogen, related to mobile applications, is associated with many disadvantages due to the safety, the cost of storage and handling, refueling, etc. These problems can be overcome by the production of hydrogen on the vehicle from a suitable high energy/density liquid fuel such as methanol, ethanol, methane, dimethylether etc. Methanol is currently considered one of the best candidates: besides having a high hydrogen/carbon ratio, it can be converted into hydrogen at moderate temperature and it does not contain carbon-carbon bonds, hence reducing the risk for coke formation. Moreover, methanol can be produced from plants, renewable sources, causing no net addition of CO<sub>2</sub> to the atmosphere. One of the methods to extract hydrogen from methanol source is through the steam reforming reaction.

CH<sub>3</sub>OH (g) + H<sub>2</sub>O (g) 
$$\rightarrow$$
 3H<sub>2</sub> (g) + CO<sub>2</sub> (g)  $\Delta$ H°<sub>R</sub> = 49 kJmol<sup>-1</sup> (298 K)

Unfortunately, the steam reforming of methanol process produces CO as a by-product in appreciable amounts (>100 ppm) during the reaction. The precious

metal electro catalyst at the fuel cell anode is poisoned by CO in concentrations exceeding a few ppm (20 ppm). Hence, present-day reforming technologies require some type of catalyst to reduce the CO-content in the hydrogen production from steam reforming of methanol.

The current investigation of hydrogen production from methanol mainly focuses on Cu-based and Pd-Zn catalysts. However, Cu-based catalysts deactivated quickly and Pd-Zn catalysts have a high CO content in the product stream. Therefore, they are not suitable for on-board production of hydrogen. Hence, many extensive research efforts are focused on the development of new catalysts to replace Cu-based and Pd-Zn catalysts.

The purpose of this research is to study the methanol steam reforming reaction over Au/ZnO and Au/ZnO-Fe<sub>2</sub>O<sub>3</sub> catalysts. The catalysts were prepared by a deposition-precipitation method and characterized by X-ray diffraction (XRD), Transmission Electron Microscopy (TEM), BET Surface Area Measurement, Temperature Programmed Reduction (TPR), and Temperature Programmed Oxidation (TPO). The effects of Au contents (1%, 3%, and 5%), calcination temperature (200, 300, 400, and 500°C), catalyst pretreatment (H<sub>2</sub> and O<sub>2</sub> pretreatment), reaction temperature (250-450°C), and ZnO-Fe<sub>2</sub>O<sub>3</sub> molar ratio (9:1, 3:1, 1:1, 1:3, 1:9) on the performance of Au/ZnO and Au/ZnO-Fe<sub>2</sub>O<sub>3</sub> catalysts for steam reforming of methanol were studied in detail. Moreover, the optimum condition for Au/ZnO and Au/ZnO-Fe<sub>2</sub>O<sub>3</sub> catalysts are chosen for further study in the presence of decomposition of methanol (DM) and water gas shift reaction (WGS) reactions. Finally, the deactivation tests of both catalysts were also performed for 24 hours.