CHAPTER I
INTRODUCTION

1.1 Introduction

In the past, people have consumed energy from burning fossil fuels that are expected to be completely used in the near future. Scientists have intensively developed an efficient technological alternative that will increase the efficiency of fuel-energy conversion thus minimizing air pollution and the greenhouse effect. Fuel cell is an alternative way. It can give higher efficiency when compared with conventional fossil-fuel power sources, which are based on burning petroleum products. Moreover burning petroleum products creates pollutants like carbon dioxide and carbon monoxide. These residues account for a large amount of the air pollution in our world. In contrast, fuel cells are relying on chemistry but not on combustion. Therefore fuel cells are friendly to environment which emits only water waste product.

Fuel cell is an electrochemical device that converts chemical energy between a fuel and an oxidant directly and continuously to electrical and thermal energy. Fuel cells are making for reliable, inexpensive and environmentally sound sources of energy. In principle, a fuel cell operates like a battery. Unlike a battery, a fuel cell does not run down or require recharging. It will produce energy in the form of electricity and heat as long as fuel is supplied.

Fuel cell consists of two electrodes sandwiched around an electrolyte as shown in Figure 1.1. Hydrogen acts as a fuel passing one electrode and oxygen over the other. Hydrogen molecules dissociate to hydrogen atoms when pass through platinum catalyst anode. The hydrogen atom splits into a proton and electron, which take different paths to the cathode. Proton passes through an electrolyte, while electron travels through an external circuit.

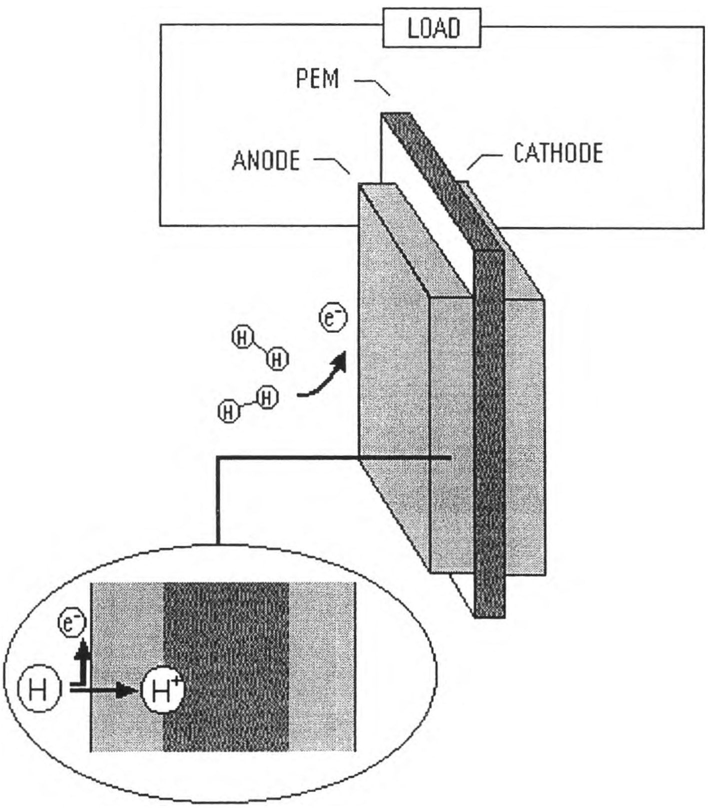

Figure 1.1 Operation of a polymer electrolyte fuel cell (PEFC).

Proton and electron react with oxygen from the air to form water generating electricity and heat. This movement of ions is illustrated in equations 1.1 and 1.2.

Anode: $2H_2 \longrightarrow 4H^+ + 4e^-$ (1.1)

Cathode: $O_2 + 4e^- + 4H^+ \longrightarrow 2H_2O$ (1.2)

Fuels for fuel cells are hydrogen, methanol or natural gas. Hydrogen is one of the most important fuel sources but it is not convenient and safe to store large amount of hydrogen in a vehicle. In contrast, methanol or natural gas could be easily stored in the car. Methanol or natural gas is converted to hydrogen, which can further be used in the fuel cell to generate power to drive the car.

Steam reforming is an important method for converting methanol or natural gas to hydrogen that can be used in the fuel cell. Unfortunately, with the current technology the hydrogen-rich reformed gas contains approximately up to 1% carbon monoxide which can poison the platinum anode and shorten the life of the platinum electrode in the fuel cell. Reducing the carbon monoxide content in the reformed gas to less than 10 ppm before it enters the fuel cell and electrocatalysis tolerating carbon monoxide of such content can solve this problem.

Removal of CO from reformed gas has long received significant attention. Many processes for CO/reformed gas separation such as methanation, pressure swing adsorption and liquid N_2 scrubbing require high capital investment and/or operation cost. Membrane separation using polymeric membrane or Pd/porous-glass composite membrane has been studied but has not been practical. It is desirable to develop low-cost and effective processes that are applicable at various capacities.

Thus far a catalytic oxidation of carbon monoxide in fuels has been proposed for diminishing the carbon monoxide content. Most catalysts for this use have been Pt supported on Al_2O_3. However, these less selective catalysts for the oxidation of carbon monoxide require more than 2% oxygen addition to oxidize 1% carbon monoxide in hydrogen-rich fuels, although it should take only 0.5% oxygen to oxidize 1% carbon monoxide stoichiometrically from equations 1.3 and 14.

$$2CO + O_2 \longrightarrow 2CO_2 \qquad H^0_{298} = -283 \text{ KJ/Mol} \qquad (1.3)$$

$$2H_2 + O_2 \longrightarrow 2H_2O \qquad H^0_{298} = -286 \text{ KJ/Mol} \qquad (1.4)$$

The remainder of oxygen added causes hydrogen combustion leading to the fuel loss. Furthermore, the addition of such a high content of oxygen involves great risks of incident explosion. There is great room, therefore, for the improvement of conventional catalysts. More selective catalysts are desired, which are active for the oxidation of carbon monoxide without consuming fuel hydrogen.

Selective carbon monoxide oxidation has been studied by large number of investigators in several aspects. Noble metals; notably Platinum (Pt), Palladium (Pd) and Rhodium (Rh) have been reported to be especially well suited for selective catalyst oxidation reactions. However, Pt and Rh are too expensive for practical use. Pd is relatively cheap and better oxidizing compared to other metals (Noh *et al.*, 1999).

The mixed oxide between CeO_2 and other rare earth oxides were investigated by some authors who believed that the oxygen atoms were from bulk. Amount of oxygen given off by the mixed oxide was higher than that of the pure CeO_2. An addition of the rare earth oxide was believed to induce the oxygen vacancies in bulk, which accelerated the movement of oxygen atoms through the lattice. Fornasiero *et al.* (1995) incorporated the ZrO_2 into a solid solution with CeO_2. The TPR indicated that the OSC (Oxygen Storage Capacity) of Rh loaded solid solution was higher than Rh/CeO_2 sample. Therefore this research focused on Pd supported on CeO_2, ZrO_2 and mixed CeO_2-ZrO_2.

1.2 Research Objectives

The goal of this research work is to identify alternative catalysts, which are more effective than currently used catalysts such as Pt/Al_2O_3 for oxidation of carbon monoxide in the presence of hydrogen for fuel cell applications. This work focused on Pd metal catalysts prepared by co-precipitation and impregnation on sol-gel methods.

In this study, BET, XRD, and AAS were used to identify structural properties with catalytic performance.