



CHAPTER I

INTRODUCTION

Hydrogen production has received much attention in recent years because hydrogen is considered as a clean energy source. There are several processes by which hydrogen can be produced, including steam reforming (SR), partial oxidation (POX), auto thermal reforming (ATR), gasification of coal or biomass, and the electrolysis or thermo chemical decomposition of water. Catalytic partial oxidation of methane is an interesting alternative for the production of synthesis gas since the reaction is mildly exothermic. It can produce a H_2/CO ratio of 2 more favorable with respect to downstream processes such as methanol synthesis or Fischer-Tropsch synthesis. One of the most common problems to the successful industrial applications is carbon deposition via the slightly exothermic Boudouard reaction (or CO disproportionation) ($2CO \rightarrow C + CO_2$) and/or methane decomposition reaction ($CH_4 \rightarrow C + 2H_2$), which leads to catalyst deactivation, plugging of the reactor, and breakdown of the catalyst.

In general, the deposition of carbon would occur over the metallic sites as well as the acid sites of the support. Therefore, the studies of developing highly active and stable catalyst have been focused. Many additives were successive to reducing carbon deposition for $Ni/Ce_{0.75}Zr_{0.25}O_2$ (Pengpanich *et al.*, 2004, 2006, and 2007). Nickel-based catalysts are attractive for partial oxidation reaction due to their low cost, but nickel is deactivated easily by carbon deposition and/or its sintering. However, by considering the high cost and limited availability of noble metals, it is more practical to develop Ni-based catalysts which are resistant to carbon deposition and exhibit high activity for such a reaction. The inhibition of carbon deposition over Ni catalysts was proposed by using perovskite-type metal oxides as catalysts since they are reported to give high thermal stability in a broad range of temperature as well as very low carbon deposition (Muller *et al.*, 1994; Qi *et al.*, 2005).

Perovskites can be represented by the general formula ABO_3 structure where the cation A large size is responsible for the thermal resistance of the catalyst, while the cation B small size is associated with the catalytic activity and stability of

the whole crystal configuration. These compounds also offer the possibility to partially substitute A and B cations by other kinds of cations, thus allowing to tailor both thermal stability and catalytic performance (Falcon *et al.*, 2002).

In this study, Ceria-Nickel and Ceria-Zirconia-Nickel mixed oxides prepared via the co-precipitation method were investigated for their catalytic activities and suppression of carbon deposition on methane partial oxidation (MPO) in the temperature range of 400-800°C at atmospheric pressure. The catalysts were carried out in a packed-bed quartz micro reactor (i.d.Ø 6 mm), then characterized by X-ray diffraction (XRD) analysis, BET surface area measurements, H₂ chemisorption, temperature programmed reduction and oxidation, and transmission electron microscopy.