

CHAPTER I

INTRODUCTION

Methane is the major component of the natural gas, which is considered as a low-price and abundant energy resource. A substantial amount of natural gas has been discovered in very remote areas. Consequently, the large investment are required for transportation of the gas to the places where the utilization is established. Plenty of natural gas and associated gas has been flared or vented to the atmosphere due to this reason. Moreover, nowadays it is not acceptable for the environmental point of view due to the greenhouse effect. The alternative way for utilization of natural gas is the conversion of methane to liquid compounds or other more valuable chemicals. The conventional method is converting methane to synthesis gas followed by (a) catalytic conversion to oxygenated compounds such as methanol or (b) Fischer-Tropsch synthesis to liquid hydrocarbons.

Methanol is an attractive valuable liquid product. It also is an important industrial chemical and a raw material for the production of chemicals such as formaldehyde, acetic acid and methyl-tertiary-butyl-ether (MTBE). The world annual production and demand increase steadily every year as shown in Figure 1.1 (Struis *et al.*, 1996 and Gesser and Hunter, 1998).

Conventionally, the conversion of methane to methanol involves the intermediate production of synthesis gas by steam reforming, followed by catalytic conversion of the synthesis gas to methanol. Current steam reforming technology involves an energy intensive process, which operates at approximately 65% thermodynamic efficiency (Foulds and Gray, 1995). The steam reforming of methane and methanol formation from synthesis gas are as following reactions:

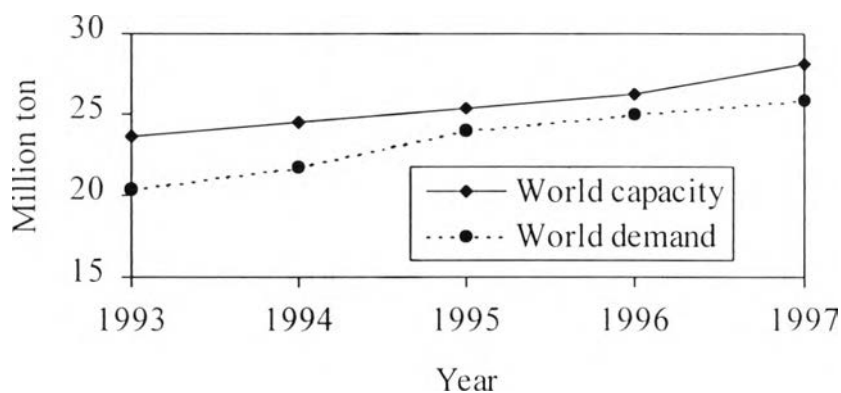


Figure 1.1 The world total capacity and demand for methanol production



The direct partial oxidation of methane to methanol is an alternative route of methane utilization. This reaction has been investigated by a large number of researchers especially during the last two decades of this century, because the reaction can be performed in the region that is more energy efficient than the conventional synthesis. On the other hand, it bypasses the energy intensive endothermic step of synthesis gas formation.



Many techniques have been employed in order to carry out this reaction. The direct conversion of methane to methanol has been initiated by laser as well as by a heated filament. The oxidation of methane to methanol has also been studied in supercritical water. Homogeneous reaction shows a reasonable yield of methanol and formaldehyde. Nevertheless, the reaction

must be operated at severe conditions and long contact time (Gesser and Hunter, 1998). Due to the less stability of these desired products at high temperature, they can be easily further oxidized to undesired products (CO_2 , CO and H_2O). Catalysis is the answer to inhibit this deep oxidation. There are several types of catalysts for methanol synthesis process such as oxides of Cu, Zn, Al, Cr, Ga, and Mo. This work was focused on low pressure catalyst. Among the most well known of them, Cu/ZnO is a good catalyst for producing methanol at low pressure. As the same trend with homogeneous reaction, if the temperature required for the catalytic reaction can be lower, the amount of undesired products can be reduced as well. In the catalytic system, the lattice oxygen (O^- , O^{2-}) acts as an active site for methane activation. However, in order to activate methane molecule at low temperature, such active species is necessary to be provided to activate the methane molecule. The most useful technique for activating methane molecule at low temperature is generating a non-equilibrium plasma in an electric discharges, e.g. corona discharge, environment in order to provide such species.

In this study, the direct partial oxidation of methane to methanol was carried out at low temperature and atmospheric pressure by using a combination of gaseous plasma environment and Cu/ZnO catalyst in packed bed reactor. The aim of this work was to determine methane conversion and products selectivities under studied conditions.