CHAPTER I
INTRODUCTION

Natural gas occurs in reservoirs beneath the surface of the earth. Methane is the main component of natural gas, landfill gas and a by-product from oil refining and chemical processing. Methane is used as a fuel for electrical generation and vehicle fuel in the form of compressed natural gas but it acts as a greenhouse gas resulting in atmospheric pollution if released. Therefore methane conversion is essential and beneficial to produce high valuable products such as olefins and aromatics. Olefins are one of the most important basic petrochemicals, which are used to produce plastics, fibers and other chemicals. Generally, they can be produced by steam cracking of higher hydrocarbons in natural gas.

Methane can be converted into higher hydrocarbons via direct or indirect methods. The indirect method involves the production of syngas (CO and H_2) which requires a consumption of either large amounts of energy or expensive pure dioxygen (Aboul-Gheit *et al.*, 2011). Therefore, the direct methods have been studied to overcome this requirement. However, these processes have not yet reached the commercial stage because of low selectivity of hydrocarbons as a result of the oxidation reactions that occur in the presence of oxygen (Ren *et al.*, 2008; Alvarez-Galvan *et al.*, 2011; Aboul-Gheit *et al.*, 2012). This problem could be solved by operating in the absence of oxygen.

Previously, many research groups attempted to activate methane under non-oxidative conditions and to convert it into higher hydrocarbons and aromatic compounds on zeolite catalysts with transition metal ions (Aboul-Gheit *et al.*, 2012; Vosmerikov *et al.*, 2007). Mo/HZSM-5 has been used as typical catalyst for this reaction (Majhi *et al.*, 2013) to provide aromatics rather than olefins. In order to improve the ethylene selectivity and yield, a series of HZSM-5 supported bimetallic Ni-Mo catalysts prepared by polyol mediated process was studied for methane dehydrogenation and coupling under non-oxidative conditions.

The study of catalytic performance over Ni/HZSM-5 was not favored for methane conversion. In the previous research, the Ni/HZSM-5 provides the high ethylene selectivity but was the lower of methane conversion less than 1 % (Xu *et* *al*., 1995). Some research group tried to using the bimetallic Ni-Mo catalyst (Aboul-Gheit *et al*., 2012; Vosmerikov *et al*., 2007). The result indicated that the incorporation of Ni over Mo/HZSM-5 can enhance the ethylene selectivity and suppress the aromatics selectivity.

The polyol mediated process is a method to synthesize catalyst by use ethylene glycol as solvent. Ethylene glycol is used to dissolve solid precursor and to prevent the agglomeration. These catalysts were tested for dry reforming of methane and the results showed that Ni nanoparticle supported on mesoporous ZSM-5 highly active for reforming of methane with CO_2 (Sarkar *et al*., 2012). The catalyst that prepared by polyol mediated process provides small particle size and high particle dispersion on surface of catalyst resulting in high catalytic activity.

The present work aims to find the optimal conditions for enhancing the production of ethylene by methane dehydrogenation and coupling using bimetallic Ni-Mo catalysts, study on the properties of catalysts characterized by XRF, XRD, SEM, TPD, TPR and surface area analysis. Moreover, the effects of Ni and Mo loading on zeolite methane feed concentration and reaction temperature on the gaseous products yield and selectivity were also studied.