

## CHAPTER I

### INTRODUCTION

Methane is the major component in natural gas and recently it has been discovered abundantly in shale gas. Economically methane has low value, therefore, it is suggested to react with other hydrocarbons to gain higher value higher value chemicals, e.g. methylation of benzene to obtain xylenes. However the stability of C-H bond of methane causes reaction of methane with other chemicals to be difficult.

Current industrial technologies for using methane are dominated by an indirect route via synthesis gas. However, this process used high energy consumption and high cost. On the other hand, direct routes for methane conversion such as a selective oxidative of methane to C<sub>1</sub>-oxygenated and oxidative coupling of methane to ethane (Lin *et al.*, 2009), have shown advantages but these processes are not commercialized yet because of low methane conversion and poor product selectivity.

Halogenation of methane is a process for activation of methane by extracting hydrogen from methane molecule and react with halogen free-radical to produce methyl halide under relatively mild condition, then the methyl halide will achieve to produce a higher hydrocarbon or oxygenates. Halogenation of methane has been intensively in the previously by Weissman and coworker (1984). The Benson process required the use of chlorine as a catalyst but the corrosive nature of the reactant and products was the major drawback in front of the commercialization. Among the halogen, bromine was believed to be the best choice, for the slightly exothermic reaction of bromine and methane has the advantage that one can control the degree of halogenations (Degirmenci *et al.*, 2005).

Oxidative bromination of methane (OBM) reaction is widely used to convert methane to bromomethane or methyl bromine (CH<sub>3</sub>Br) that could be the suitable feedstock for high value petrochemical production. Xu *et al.* (2005), Wang *et al.* (2006) and Yang *et al.* (2008), firstly reported OBM reaction and developed Ru- and Rh-containing impregnated catalysts. HBr/H<sub>2</sub>O (40%wt) and oxygen were used instead bromine which showed high selectivity for CH<sub>3</sub>Br and excellent stability However, Liu *et al.* (2010) studied Rh supported silica sol-gel catalyst and found that the catalytic performance was excellent during 650 h time on stream and

they did not observe any catalyst deactivation. Due to the high cost of Rh and Ru metal, Lin *et al* (2009) focused on non-noble metal oxide supported silica catalyst, prepared by impregnation technique, on OBM reaction and found that BaO/SiO<sub>2</sub> catalyst can initiate the OBM to yield CH<sub>3</sub>Br and CO.

The purpose of this work is to investigate the effect of catalysts preparation between incipient wetness impregnation and sol-gel method of barium oxide or tungsten oxide on silica catalysts for the oxidative bromination of methane.