CHAPTER I
INTRODUCTION

The upgrading of lower hydrocarbons, especially light alkanes and alkenes, is an important subject of heterogeneous catalysis. In recent years, much attention has been focused on ZSM-5 catalyzed-transformation of light hydrocarbons into higher hydrocarbons (benzene, toluene and xylene; BTX) with high conversion and selectivities, which represents one route of converting the cheap raw materials into more valuable compounds. Alternatively, a newly emerging technology is the catalytic cracking of C_4^+ alkenes to propylene and ethylene, in which catalyzed by ZSM-5 zeolite, has been reported to increase the total yield of olefins (mainly propylene) in the process.

About 52 million tons per year of PG/CG (polymer and chemical grade) propylene is currently produced in the world, the majority of which is produced by steam crackers as a by-product of ethylene. About 70% of propylene is generated by steam crackers, 28% by refinery FCC (Fluid Catalytic Cracking) units, and 2% by propane dehydrogenation, olefin metathesis and methanol to olefins. Propylene demand is expected to grow faster than supply. New options must be considered to meet the future demand for propylene because conventional steam cracking and FCC technologies will be unable to close the growing gap between supply and demand. The alternative olefin cracking technologies (MOI ® (ExxonMobil), SUPERFLEX ® (Lyondell/Kellogg), Propylur ® (Lurgi)) will potentially quadruple propylene yield. These processes are characterized by the use of ZSM-5 catalysts to convert higher molecular weight olefins and paraffins to lighter olefins in a fixed or fluidized bed reactor. The processes are compatible with crackers and FCCs and, unlike methathesis, do not consume ethylene.

Unfortunately, a common problem with zeolite catalysts used in hydrocarbon conversion is deactivation by coke deposits which are formed from undesirable side reactions. It has been generally concluded that the formation of carbonaceous deposits and their action upon the zeolites depends, not only on the characteristics of the zeolite catalyst, but also on the nature of the reactants involved and the related operating conditions.

Therefore, this research will focus on coke formation and life cycle of the catalyst. The following compounds, i.e., *n*-butane, 1-butene, *i*-butene, *n*-pentane, are individually used as feedstocks in order to analyze the coke precursors. After the reaction testing, the characteristics and amount of coke deposited on the spent catalyst will be determined by TPO and TEM technique. The regeneration of the catalyst will be also studied by oxidative treatment with varying flow rate of oxidant, regenerating temperature and duration of regeneration.

On the other hand, in terms of average annual growth rates for demand during 2002-2008 period, demand for benzene is expected to increase 3.8%, toluene 4.1% and xylene 6.3%. Half a century, the catalytic reforming process has been used to convert the alkanes and cycloalkanes to aromatic products as selectively as possible, and most often carried out on a chlorinated platinum/alumina catalyst. Although at present catalytic reforming of naphtha is the main source of BTX, the standard $Pt/Re/Al_2O_3/Cl$ catalyst is, however, not very effective for converting C_6 alkanes to benzene.

It was unquestionable that ZSM-5 zeolite can be modified by incorporation of metal or metal oxides, such as Ga, Zn and Pt, in order to increase the rate and selectivity of aromatization reaction and inhibits cracking side reaction that lead to loss of carbon to undesirable products. This promotion of ZSM-5 has led to the recent use of these materials in moving-bed reactors in the Cyclar process jointly developed by BP and UOP. This process offers a unique ability to produce BTX from excess LPG which is lower value feedstock. The principal Cyclar operating variables are feedstock composition, pressure, space velocity and temperature to encourage the acceptable BTX yields. For example, space velocity is optimized against conversion within this temperature range to obtain high product yields with minimum operating costs.

Accordingly, the second part of this work will concentrate on the activity and selectivity of aromatic products on the aromatization reaction of mixed C_4 feedstock (main composition : 37% *i*-butane and 55% butene) by using ZSM-5 zeolite. Attention is paid to vary reaction temperature, space velocity and diluent. The effect of introduction of metal (Ag and Ga) into ZSM-5 will be also investigated.