## CHAPTER I



## INTRODUCTION

Over the last quarter of a century, the chemical industry has mainly depended on petroleum as the raw material for organic But nowadays, natural gas and associated gas are synthesis. alternative resources to a petrochemical industry. Since the oil crisis, petroleum has been replaced by coal in the steel industries and in the electric power and other large scale energy generation systems. Accordingly, the excessive supply of petroleum has become serious problem, and development new routes for effective utilization of petroleum has become of great significance. "C1 chemistry" is one of the new routes which rised to replace petroleum chemistry, this new route is the catalyst-chemistry which uses syngas as starting materials. C1 chemistry is a general term for synthesis technology starting from compounds contained one carbon atom such as CO, methanol (CH3OH), CO2, and methane (CH4) from natural gas. This contrasted with "petrochemistry" which can be regarded as C2 chemistry because ethylene (C2H4) is used as a primary material in this industry as mentioned above.  $C_1$  chemistry is not a particularly new technological concept. The methanol synthesis from syngas has the oldest history. In 1913, BASF of Germany had developed the technology for methanol synthesis from water gas, and industrialized it. After this success, France, Italy, and Japan

successively started to produce methanol. The Fischer-Tropsch (F-T) synthesis (hydrocarbon synthesis from syngas) an epoch-making result in the history of catalytic synthesis, was industrialized in Germany and Japan more than 40 years ago.

The most striking recent inovation in C2 chemistry was the success in the synthesis of a shape selective zeolite catalyst. ZSM-5 [1] by Mobil Oil of the U.S.A.. They had developed and commercialized the zeolitic cracking catalyst from the 1950's. They discovered in this sequence of studies that the product selectivity can be changed remarkably by changing the shape and size of the zeolite pores. In order to increase the thermal stability and hydrophobic property, synthesis of zeolite richer in silica than conventional synthetic zeolites was investigated. Thus, a novel zeolite ZSM-5 which has a micropore opening with 10 oxygen-rings (pentasil structure) was synthesized. ZSM-5 is different from conventional zeolites having 12-rings (X, Y, and Mordenite) and 8 rings (A and Erionite). The middle size pore dimension of ZSM-5 and its three-dimensional pore structure produced a marvelous result in methanol conversion to gasoline. A long-pending question in conventional F-T synthesis is "How does one avoid having a product distribution determined by the rule of carbon polymerization probability ?". Product distribution is spread over a wide range of carbon numbers in the F-T synthesis. Therefore, both the yield of the gasoline fraction and its research octane-number are low. On the other hand, when ZSM-5 was used as the catalyst, methanol converted completely to hydrocarbons, and the gasoline fraction amounted to about 80% with a fixed bed reactor operation.

Coke formation, i.e., the deposition of carbonaceous residued, on solid acid catalysts such as silica-alumina and zeolites usually leads to rapid deactivation [2]. This is the serious problem of methanol conversion process because it makes lower in activity and selectivity of solid acid catalysts. It is of paramount interest to relate coke formation on various catalysts to their actual rate of deactivation. Coke deposits can indeed lower the catalytic activity by site coverage (poisoning) and/or by pore blocking which prevents the access of the reactants to the active sites.

In general, this work aims to investigate the catalytic behaviour of catalysts (Pt/H-ZSM-5, Re/H-ZSM-5 and Re-Pt/H-ZSM-5) in methanol conversion reaction. Moreover, the deactivation of H-ZSM-5 and the modified H-ZSM-5 loading with Pt and Re-Pt by carbonaceous compounds for methanol conversion were studied.