

CHAPTER I

INTRODUCTION



During the last few decades the vast reserves of natural gas (which the predominant component is methane) have been driving chemists to pursue the chemical utilization of this resource. Many investigations have been carried out on the conversion of natural gas especially methane into high molecular weight hydrocarbons. This is because methane is the main component and it can be readily purified to yield a very clean and H-rich source of hydrocarbons. So it is interesting for the production of clean fuel products [Chen *et al.* (1996)].

Research in this field was spread over a wide range which can be arranged the tendency in three part ; (i) new reactions of C_1 compounds ; (ii) development of efficient catalysts for such reactions ; (iii) development of the industrial processes. Various processes, all based on natural gas conversion, are either already commercial or at an advanced stage of development. There are two general types for methane conversion. One is methane derivation and the other is direct self-coupling (figure 1.1).

For methane derivation, this reaction is two steps. The first step requires the steam reforming to produce synthesis gases (CO , H_2) at a high temperature. Subsequently hydrocarbons can be produced in a low temperature exothermic process from synthesis gas, either by Fischer-Tropsch (FT) synthesis or via methanol and the methanol-to-gasoline (MTG) process. Some of the typical and most desirable reactions of the hydrogenation of CO are summarized in table 1.1 . Other products such as acids, ketones, aldehydes and aromatics can also be produced by similar reactions. Since water is a primary product in most of the synthesis reactions, the water-gas shift reaction can occur between this water and CO from the feed , which changes the O-containing by-products from water to carbon dioxide. As a result,

considerable research into alternative direct routes has been carried out as to avoid this necessity [Fierro (1993)].

On direct self-coupling process, there are two routes for converting methane into higher hydrocarbons, oxidative and non-oxidative coupling. Oxidative coupling of methane (OCM) has been the main route considered recently for the direct catalytic conversion of methane into more useful chemicals [Belgued *et al.* (1992)]. It can only operate at very high temperature, above 927°C. To increase the catalyst lifetime and the selectivity for C₂₊ hydrocarbons, it is interesting for processes operation at lower reaction temperature. Non-oxidative coupling is an alternative route. Furthermore, in the oxidative coupling process the combustion reaction could be occurred. Part of methane loses in this reaction. Although the chemical conversion of methane into more valuable chemicals was studied over the last decade. Nevertheless the non-oxidative coupling of methane (NOCM) was observed in the last few years.

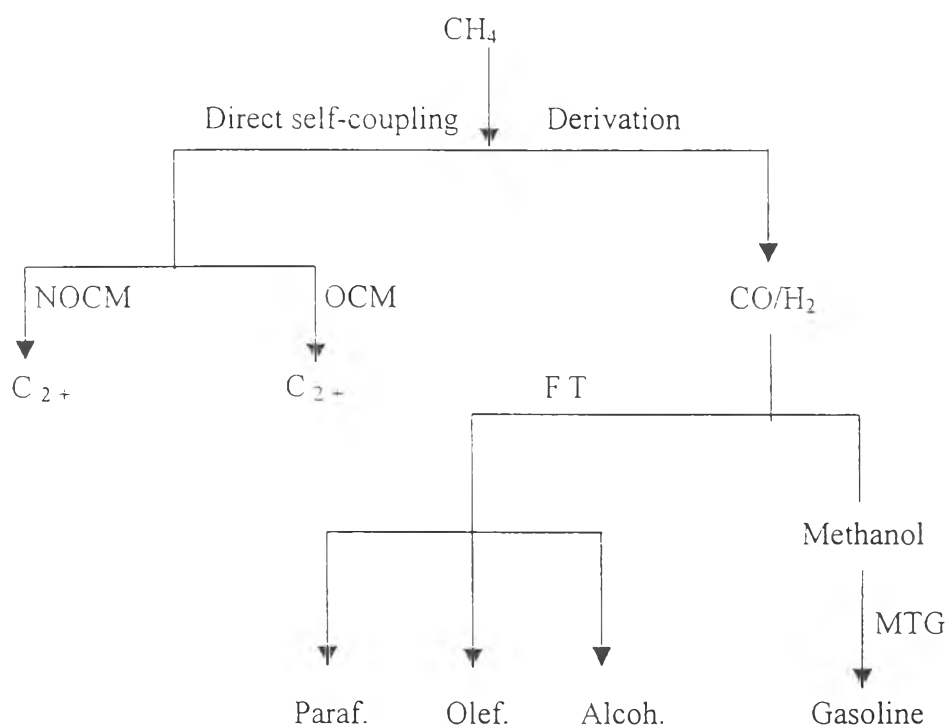


Figure 1.1 Schematic representation of several catalytic developments of which the origin lies in natural gas [Fierro (1993)].

Table 1.1 Reactions which can occur during hydrogenation of CO [Fierro (1993)].

Desirable	Methanation:	$3\text{H}_2 + \text{CO} \longrightarrow \text{CH}_4$
	Hydrocarbons:	$2n\text{H}_2 + n\text{CO} \longrightarrow \text{C}_n\text{H}_{2n}(\text{C}_n\text{H}_{2n+2})$
	Alcohols:	$2n\text{H}_2 + n\text{CO} \longrightarrow \text{C}_n\text{H}_{2n+1}\text{OH}$
Undesirable	Water-gas shift:	$\text{CO} + \text{H}_2\text{O} \longrightarrow \text{CO}_2 + \text{H}_2$
	Boudouard reaction:	$2\text{CO} \longrightarrow \text{C} + \text{CO}_2$
	Coke formation:	$\text{CO} + \text{H}_2 \longrightarrow \text{C} + \text{H}_2\text{O}$

Recently, series of experiments was carried out using transition metals supported on silica for the non-oxidative methane coupling reaction. Subsequently zeolite Y was used in this reaction as a support of transition metals. It gave a better yield in higher hydrocarbons and the temperature at which methane activation took place was lower.

There are few reports in which studied the effective utilization of zeolite Y catalyst containing Pt and Co. Therefore, this study aims to investigate the catalytic performance of various transition metals containing zeolite Y catalyst for the non-oxidative methane coupling.

The present work is arranged as follows:

Chapter II presents the literature reviews of investigation the non-oxidative methane coupling reaction.

The theory of this research, the theoretical consideration on zeolite Y and the mechanism of non-oxidative coupling of methane on metal catalysts are given in chapter III.

Following by the description of the experimental systems and the operational procedures in chapter IV.

The experimental results obtained from a laboratory scale reactor and standard measurement are reported and discussed in chapter V.

Chapter VI gives overall conclusions emerged from this work and presents some recommendations for any future works.

Finally, the sample of calculation of catalyst preparation, the calibration curves of methane, ethane and propane and the data of experiments are included in appendix at the end of this thesis.