CHAPTER I
INTRODUCTION

1.1 General Introduction

With its inexpensive and plentiful resource, natural gas becomes the most available primary fuel for both community and industry. Among all primary fossil fuels (oil, natural gas, and coal), natural gas is the cleanest energy and is recognized as the most environmentally safe fuel. Natural gas is considered to be not only a predominant primary fuel but also the most crucial source of chemical feedstocks i.e. synthesis gas, methanol, and ammonia for producing many basic petrochemicals, via either indirect or direct chemical processes. There are several conventional reactions to produce useful chemicals from natural gas, e.g. thermal pyrolysis, steam reforming, partial oxidation, and auto-thermal reforming. Nonetheless, the reactions normally require high operating temperatures, due to a very strong C-H bond of methane, the main component of natural gas. From the large external energy requirement, most of these catalytic reactions still have not been succeeded in commercial processes, except for the steam reforming process.

The use of non-thermal plasma processes should be one possible way for solving the constraints of the catalytic reactions to be operated at severe conditions. In the non-thermal plasma, the elementary reactions with electron collision (mainly ionization, excitation, and dissociation of gas molecules) create a variety of chemically reactive species (radicals and excited species) for activating subsequential chemical reactions. The interactions between the accelerated charged particles and other chemical species take place throughout the entire plasma zone to promote the reactions with high productivity, which are not possible to occur in surface catalysis technologies (Mutaf-Yardimci *et al.*, 1998). There are several types of non-thermal plasmas, i.e. glow, corona, microwave, radio frequency, and dielectric barrier discharges. These non-thermal plasmas provide high product selectivities and yields, but at relatively low power levels. A gliding arc is another new discharge type of non-thermal plasmas that is designed to keep a high level of non-equilibrium, high electron temperature, and high electron density to enhance chemical processes with relatively low energy consumption. It therefore successfully provides the most effective non-equilibrium characteristics with simultaneous high productivity and good selectivity (Fridman and Kennedy, 2004).

The non-thermal gliding arc discharge is considered to be a promising approach to facilitate natural gas reforming at low temperatures. Additionally, the concept of the direct utilization of natural gas has a contributing part in reducing the cost of any separation process and the net emission reduction of carbon dioxide as a greenhouse gas. Therefore, to acquire the ultimate profit from natural gas resources in Thailand, this research attempted to convert raw natural gas into more desirable products under the gliding arc discharge reactor.

1.2 Objectives

1. To study the effects of other gaseous hydrocarbons and carbon dioxide present in natural gas on the methane reforming under the AC gliding arc system.
2. To study the effects of electrical discharge parameters, i.e. applied voltage and frequency on the simulated natural gas reforming under the AC gliding arc environment.
3. To study the effects of oxygen adding, i.e. using pure oxygen and air, and hydrocarbons-to-oxygen molar ratio on the combined CO_2-containing natural gas and partial oxidation under the AC gliding arc system.
4. To study the effects of operational parameters, reactor configuration and the presence of catalyst on the methane reforming using the gliding arc microreactor.

1.3 Scope of Work

In this work, simulated natural gas contained a CH_4:C_2H_6:C_3H_8:CO_2 molar ratio of 70:5:5:20 was used as a representative of raw natural gas in Thailand, based on the data source from PTT Public Company Ltd., as shown in Table 1.1. In the first part (Chapter III), the series of experiments were systematically carried out under different feed gas systems: pure methane, CH_4/He, CH_4/C_2H_6/He, CH_4/C_2H_6/C_3H_8/He and CH_4/C_2H_6/C_3H_8/CO_2 (simulated natural gas). The interactions among the gas components of natural gas under the plasma environment were clarified. Furthermore, several possible chemical pathways were proposed also, based on the experimental results. For the next step (Chapter IV), the electrical parameters of applied voltage and frequency were investigated to find out the optimum conditions for the reforming of the simulated natural gas under the gliding arc system. The optimum conditions were selected to study the effect of oxygen addition in feed. The comparative results between using pure oxygen and air as well as the significant role of oxygen on the plasma reactions were discussed. In Chapter V, for the simplicity, methane was used as a feed gas for the preliminarily study of the integrated gliding arc and microreactor system. In addition, the methane reforming was also performed under the studied gliding arc microreactor over the unloaded catalyst plate and nickel-loaded catalyst plate.

Table 1.1 Typical compositions of natural gas in Thailand

Composition	Arawan	Bongkot	Yadana	Yetakun
CH_4	63.565	62.315	69.099	79.110
C_2H_6	8.285	7.318	0.915	7.360
C_3H_8	4.900	4.466	0.167	2.550
i-C_4H_{10}	1.225	1.002	0.018	0.550
n-C_4H_{10}	1.095	0.990	0.028	0.640
i-C_5H_{12}	0.370	0.333	0.008	0.240
n-C_5H_{12}	0.250	0.219	0.008	0.240
C_6^+	0.455	0.340	0.020	0.150
CO_2	15.995	22.287	4.144	6.970
N_2	3.860	0.730	25.598	2.290

Source: PTT Public Company Limited (2001).