CHAPTER I
INTRODUCTION

Ethylene oxide (C_2H_4O, EO) is an important industrial chemical, which is used as an intermediate in the production of various useful chemicals. The ethylene oxide is colorless flammable gas or refrigerated liquid with a faintly sweet odor, and it is the simplest molecule of an epoxide (www.en.wikipedia.org/wiki/Ethylene_oxide). Its major use is in the production of ethylene glycol, which is used to produce polyester polymers. Ethylene oxide can be polymerized to form polyethylene glycol or polyethylene oxide. It can also be used to produce detergents by a process called ethoxylation, sterilants for foodstuffs, solvents, antifreezes, adhesives, sterilants for medical equipment and supplies, and cosmetics (www.prlog.org).

Commercially, ethylene can be oxidized to ethylene oxide with high selectivity by using silver catalyst supported on low-surface-area alpha-alumina (Ag/(LSA)α-Al_2O_3) (Matar *et al.*, 1989). Addition of few ppms of chloride to gaseous reactant as moderator in the form of chlorine-containing hydrocarbon species, such as dichloroethane ($C_2H_4Cl_2$) and vinyl chloride (C_2H_3Cl), has been reported to significantly increase the selectivity to ethylene oxide by 15-20 % (Law and Chitwood., 1942; Campbell, 1984; Tan *et al.*, 1986; and Yeung *et al.*, 1998). Alkali and alkali earth, such as Cs and Re, also provided the improvement of selectivity for ethylene oxide by 10 % (Iwakura, 1985; and Bhasin, 1988). In a recent work, silver catalyst supported on high-surface-area alpha-alumina (Ag/(HSA)α-Al_2O_3) and Au/TiO_2 were proved to exhibit good selectivity for ethylene oxide (Rojluechai *et al.*, 2006). Moreover, adding small amount of Au to form Au-Ag bimetallic catalyst over high-surface-area alpha-alumina support favored the epoxidation of ethylene to ethylene oxide (Rojluechai *et al.*, 2006). However, ethylene conversion obtained from these catalysts could not be detected at any temperature below 493 K. Even though the reaction temperature was raised up to 543 K, ethylene conversion was still low at 1-4 %. Consequently, this limitation results in high energy consumption for catalyst activation at high temperature, which is a disadvantage for industrial application. In addition, several catalytic problems,

e.g. catalyst deactivation, catalyst regeneration, and catalyst replacement, are the other drawbacks of the conventional catalysis processes. The non-conventional catalysis technique is, therefore, expected to overcome these constraints. One of potential techniques is to use non-thermal plasma for the ethylene epoxidation.

Non-thermal plasma is one kind of electric gas discharges, of which electrons in electrodes gain enough energy from applied voltage to overcome the potential barrier of metal surface electrodes (Eliasson, 1991). Then, they are directly transformed to chemically excited or dissociated gaseous species by colliding with the gaeous components present in the plasma zone. The non-thermal plasma is in general any plasma that possesses non-equilibrium properties, either because the ion temperature is different from the electron temperature or because the velocity distribution of one of the species does not follow a Maxwell Boltzman distribution. Typically, the generated excited or dissociated species have much higher reactivity than neutral species at the ground state. Moreover, the main characteristic of non-thermal plasma is that the electrons in plasma zone have a higher energy than neutral gas at relatively low temperature near room temperature. The examples of chemical synthesis using non-thermal plasmas are oxidations of olefin and aromatics, and so on (Suhr *et al.*, 1988; Patiño *et al.*, 1996; Chavadej *et al.*, 2008; Sreethawong *et al.*, 2008; and Sreethawong *et al.*, 2010).

The corona discharge, which is one type of non-thermal plasmas, is employed in this research since it is capable of operating at low temperature and atmospheric pressure (Eliasson, 1991). The characteristic of corona discharge is to use a pair of inhomogeneous pin-plate metal electrode geometries, which can stabilize the discharge generated. There are many industrial applications involving the utilization of corona discharge, such as NO_x and SO_x reduction in flue gas, toxic compound destruction, and ozone production (Chang *et al.*, 1991; Yan *et al.*, 1998; and Bröer and Hammer, 2000). In previous works, it was also worked with photocatalyst for degradation of ethylene (Harndumrongsak, 2002; and Chavadej *et al.*, 2007) and for VOC removal (Chavadej *et al.*, 2007). In addition, Chavadej *et al.* (2008) studied the ethylene epoxidation using a corona discharge system; however, it was found that the ethylene oxide selectivity was still relatively low, probably

because the ethylene and oxygen gaseous reactants were simultaneously fed into the system, resulting in the formation of several by-products.

The objective of this research was to improve the ethylene epoxidation performance using the corona discharge system by initially producing O_2 plasma prior to reacting with ethylene, which was directly fed into the system at various positions in the plasma zone. The effects of various operating parameters, including distance between plate electrode and ethylene feed position, O_2/C_2H_4 feed molar ratio, applied voltage, input frequency, total feed flow rate, and gap distance between pin and plate electrodes, on the activity of ethylene oxidation were examined. Moreover, the optimum conditions for a maximum ethylene oxide production were determined.