CHAPTER I

INTRODUCTION

A fuel cell (FC) is an energy conversion device that produces electricity directly from a gaseous fuel by electrochemical combination of the fuel with an oxidant. A solid oxide fuel cell (SOFC), one of the most popular type of fuel cells nowadays, has attracted many researchers' interest worldwide due to its widest range of applications, flexibility in fuel choice and high system efficiency (Yamamoto, 2000; Steele, 2001; Stambouli and Traversa, 2002). Most of previous studies have been focused on the SOFC fuelled by hydrogen or carbon monoxide which can be either directly fed or produced through a direct or indirect internal steam reformer (Achenbach, 1994; Bessette *et al.*, 1995; Nakagawa *et al.*, 2001; Aguiar *et al.*, 2002; Yamada *et al.*, 2002). Although most of the studies are aimed at electricity generation, some research groups have considered another potential area of application on chemicals and electric power co-generation. Farr and Vayenas (1980) first demonstrated this type of operation for the case of NH_3 conversion to NO. Later many reactions have been applied to SOFC reactors. They are, for example, styrene production from ethylbenzene (Michaels *et al.*, 1984), oxidation of H_2S to SO_2 (Pujare *et al.*, 1987; Yentetakis and Vayenas, 1989), oxidation of methanol to formaldehyde (Neophytides and Vayenas, 1990) and oxidative coupling of methane to ethane and ethylene (Pujare and Sammells, 1988).

During the past few decades, there are many efforts to upgrade methane, an abundant component in natural gas, to other valuable chemicals. There are two routes of methane conversion to other chemicals; i.e., indirect and direct routes. A conventional approach uses an indirect method to convert methane to synthesis gas which is then reacted to form other chemicals such as C_1-compounds (methanol and formaldehyde), C_2-compounds (ethane, ethylene and acetylene) and oxygenate compound. More recent studies have been focused on the direct method. Among the various schemes of direct conversion of methane into more valuable feed stocks, the oxidative coupling of methane (OCM) to C2 hydrocarbon (i.e., ethane and ethylene) is a promising process. However, as found from the OCM kinetic, the combustion of methane to carbon dioxide, one of the most important greenhouse gas, and carbon monoxide are the major side reactions encountered in this system. Selective catalyst for the methane coupling reaction and reactor design to suppress the formation of carbon oxides are of primary importance for the implementation of this reaction to industrial-scale production.

The use of SOFC reactor for co-generation of C2 hydrocarbons and electric power was probably first reported by Pujare and Sammells (1988). A solid electrolyte was used as an oxygen separator and an oxygen distributor to achieve higher C2 selectivity. Although this type of reactor is more attractive than other conventional reactors due to the achievement of high selectivity and possible use of generated electricity, there are still not many works on this subject. Recent works mainly focused on the selection and preparation of selective anode catalysts for the system and effects of operating parameters on the reactor performance (Tagawa *et al.*, 1998; Liu *et al.*, 2001). One of the active and selective catalysts for OCM was amorphous $Ln_{1-x}Al_xO_3$ catalyst (Ln = rare earth metal) (Imai *et al.*, 1990; Spinicci *et al.*, 2001). Tagawa *et al.* (1998) using temperature-programmed desorption (TPD) technique to specify the surface oxygen species. Various LnAlO powder samples (Ln = La, Sn, Nd, Dy, Ce, Tb, Pr) were investigated. The results suggested that the lanthanum containing catalyst (LaAlO) was active and oxygen species were capable of moving around at the operating temperature of SOFC. From the literature survey, it was found that only one work (Guo *et al.*, 1999) is available on modeling of the SOFC reactor for this OCM reaction although there are numerous works on modeling of conventional SOFC fed by hydrogen or carbon monoxide.

From the above reasons, this research is focused on the use of SOFC reactor for co-generation of C2 hydrocarbons and electric power. The objectives of the study were to

1. characterize oxygen species on the anode catalyst by using O_2 temperature-programmed desorption (O_2-TPD) technique,
2. study the steady state oxygen permeation through the LSM/YSZ/LaAlO cell in the SOFC reactor,
3. model the OCM in the SOFC reactor, and

4. determine effects of operating conditions such as temperature, feed molar composition, feed flow rate, external load on performances of the SOFC reactor.