SIMULATION AND OPTIMIZATION OF GASOLINE AUTOTHERMAL REFORMER FOR FUEL CELL APPLICATIONS

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To my beloved parents and my fiance

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ABSTRACT

Fuel cell systems are being developed for powering clean, efficient automobiles of the future. The proton exchange membrane fuel cell (PEMFC) systems being developed for such use require a fuel gas that is either pure hydrogen, or a gas mixture that contains significant concentration of hydrogen. Thus, the vehicles with gasoline as the on-board fuel use a fuel processor, also referred to as an autothermal reformer, to convert gasoline to a fuel gas and reformate, that contains hydrogen, carbon dioxide, water vapor, and nitrogen, with trace levels of other species, such as carbon monoxide and unconverted gasoline. With the help of Aspen HYSYS 2004.1 the steady state model has been develop to analyze the fuel processor and total system performance. In this case study, the PEM fuel cell system consists of the fuel processing and clean-up section, PEM fuel cell section and auxiliary units. While the fuel processing and clean-up section consists of Autothermal Reformer, High-temperature Shift, Medium-temperature Shift, Low-temperature Shift, and Preferential Oxidation. The purpose of this study is to identify the influence of various operating parameters such as A/F and S/F ratio on the system performance that is also related to its dynamic behaviours. From the steady state model optimization using Aspen HYSYS 2004.1, an optimised reaction composition, in terms of hydrogen production and carbon monoxide concentration, corresponds to A/F ratio of 18.5 and S/F ratio of 9.0. Under this condition, n-octane conversion of 100%, H_2 yield of 42% on wet basis and carbon monoxide concentration of 7.56ppm can be achieved. The fuel processor efficiency is about 80.41% under these optimised conditions.

ABSTRAK

Sistem sel bahan api sedang dibangunkan bagi tujuan memperolehi kuasa yang bersih dan sistem pengangkutan yang cekap untuk masa hadapan. Sistem elektrolit polimer sel bahan api (PEMFC) sedang dibangunkan bagi tujuan tersebut tetapi memerlukan hidrogen tulen atau campuran gas yang mengandungi kepekatan hidrogen pada kadar yang tertentu. Dengan demikian, kenderaan yang menggunakan gasolin sebagai bahan api yang diproses melalui *autothermal reforming* (ATR) telah dibangunkan. *Autothermal reforming* berfungsi untuk mengubah gasolin kepada gas bahan api dan bahan-bahan yang lain seperti hidrogen, karbon dioksida, wap air, dan gasolin yang tidak bartindak balas. Aspen HYSYS 2004.1 telah digunakan untuk membina model yang berkeadaan tetap, bagi menganalisa kecekapan pemproses bahan api dan keseluruhan sistem. Tujuan kajian ini adalah untuk mengenalpasti pengaruh bagi nilai-nilai berlainan operasi parameter terhadap pencapaian sistem yang juga berkait rapat dengan sifat-sifat dinamiknya. Di dalam kajian ini, sistem sel bahan api PEM terdiri daripada bahagian pemproses bahan api dan bahagian pembersihan, bahagian sel bahan api PEM dan unit-unit tambahan. Manakala, bahagian pemproses bahan api dan pembersihan pula terdiri daripada *Autothermal Reformer, High-temperature Shift, Medium-temperature Shift, Low-temperature Shift* dan *Preferential Oxidation*.. Daripada kajian yang telah dijalankan dengan menggunakan Aspen HYSYS 2004.1, nisbah *A/F* dan *S/F* adalah 18.5 dan 9.0 dimana penghasilan hidrogen dan kepekatan karbon monoksida adalah optimum. Di bawah keadaan ini, penukaran n-octane adalah 100%, penghasilan hidrogen sebanyak 42% dan kepekatan karbon monoksida adalah 7.56 ppm. Di samping itu, kecekapan pemproses bahan api adalah 80.41% dalam keadaan optimum ini.

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NOMENCLATURE

AFC	alkaline fuel cells
ATR	autothermal reforming
FCV	fuel cell vehicle
GHG	green house gases
HTS	high temperature water gas shift
ICE	internal combustion engines
LHV	lower heating value
LTS	low temperature water gas shift
MCFC	molten carbonate fuel cell
NRC	naphtha reforming catalyst

PAFC	phosphoric acid fuel cells
PEMFC	proton exchange membrane fuel cells
POX	partial oxidation
PROX	preferential oxidation
SOFC	solid oxide fuel cells
SPFC	solid polymer fuel cells
SR	stoichiometric ratio
SREF	steam reforming
TOX	total oxidation

CHAPTER I

INTRODUCTION

1.1 Introduction

Fuel cells, which have seen remarkable progress in the last decade, are being developed for transportation, as well as for both stationary and portable power generation. A variety of fuel cells for different applications is under development, e.g. solid polymer fuel cells (SPFC), also known as proton exchange membrane fuel cells (PEMFCs) operating about 353K, alkaline fuel cells (AFC) operating about 373K, phosphoric acid fuel cells (PAFC) about for 473K, molten carbonate fuel cell (MCFC) operating around 923K, solid oxide fuel cells (SOFC) for high temperature operation, 1073-1373K (Wang and Zhang, 2005).

The advances in fuel cells and their supporting technology have been spurred by the recognition that these electrochemical devices have the potential for both high efficiency and lower emissions. Automobile manufacturers have decided that, given the state of technology, the PEMFC has the best potential to replace the internal combustion engine for propulsion power. Their decision is based on many considerations, including the ability to fit (size and weight) the power plant under the hood of the car, the ability to start up quickly, the ability to meet the changing power demands (dynamic response) typical in driving cycle, and cost (Ahmed and Krumpelt, 2005).

The fuel for the fuel cell system will vary with different applications. In transportation, it may be methanol, gasoline, or diesel. In stationary systems, it is likely to be natural gas, but it could also be propane. In certain niche markets, the fuel could be ethanol, butane, or biomass-derived materials. All these fuels are hydrocarbons or oxygenate that need to be reformed (Ahmed and Krumpelt, 2005). Partial oxidation (POX), autothermal reforming (ATR) and steam reforming (SREF) are the primary methods used in reforming hydrocarbons to produce hydrogen for use in PEM fuel cells.

Partial oxidation and autothermal reforming processes do not require indirect heating in contrast to steam reforming. Moreover, they offer faster startup time and better transient response. However, the product quality is poor due to low hydrogen concentrations, 70-80% for steam reforming versus 40-50% for partial oxidation and autothermal reforming on a dry basis. Compared with partial oxidation and autothermal reforming, catalytic steam reforming offers higher hydrogen concentrations. The steam reforming reaction on the other hand is a highly endothermic reaction and requires heating (Ersoz et al., 2006).

Majority the automobile manufacturers and oil industry accounts hydrogen as the ideal long-term fuel cell systems, but it is not yet clear, what will be the best fuel for the introduction of fuel cell systems. The use of hydrogen results in high efficiencies and a simple system design. Liquid fuels like methanol or gasoline on the other hand show advantages in terms of high energy density, easy fuel handling and-in the case of gasoline- in an existing fuel infrastructure (Wang and Zhang, 2005).

On board reforming of gasoline, which already presents a well-developed distribution network, is particularly interesting for a more efficient utilization of energy in vehicles, compared to internal combustion engines. Besides that, it also have the following advantages of higher heat value, large amounts of storage hydrogen and steady state supply as well as convenient transportation. Moreover, the method of producing hydrogen from gasoline through autothermal reforming combined of partial oxidation and steam reforming enjoys the merit of low energy requirement, due to the opposite contribution of the exothermic hydrocarbon oxidation and endothermic steam reforming (Wang and Zhang, 2005).

For the hydrogen production by gasoline reforming most researchers, prefer autothermal reforming concepts to steam reforming and partial oxidation because they enable

- a high hydrogen yield because of the addition of water to the feed
- minimization of NO_x – and soot-production by the addition of water and the low reaction temperatures (800-1000 °C)
- dynamic operation through in-situ provision of the required energy because of exothermal reactions.

The operating parameters of autothermal reformers are the stoichiometric ratio (SR) which is defined as the amount of oxygen in the feed divided by the amount of oxygen necessary for complete combustion and the feed temperature. In order to achieve high system efficiencies, a suitable system configuration were developed with the help of the steady state simulations (Aspen HYSYS 2004.1).

With respect to the above mentioned requirements there is also the need to optimise the dynamic behavior of a fuel cell system based on gasoline reforming. The quicker the system is able to follow load changes the smaller, cheaper and less heavy is the necessary battery in the car. For that purpose, the components of a fuel cell system with autothermal reformer have to be described by appropriate dynamic simulation model. These models have to be implemented in a simulation program which is able to solve differential equations with the help of a numeric solver (Sommer et al., 2004).

1.2 Problem Statement

The amount and concentration of hydrogen generated from a given amount of gasoline, and the quality of the raw reformat (i.e. CO, CO_2, CH_4 and other hydrocarbons, H_2O, and N_2 contents), are influenced by the reforming conditions. The amount of H_2 produced determines the efficiency of the fuel processor; the greater this amount, the higher is the fuel processor efficiencies. Thus, this study will be covering the following area:

1. What are the optimum Air/Fuel (A/F) and Steam/Fuel (S/F) molar ratios to get the high hydrogen production with CO concentration less than 10 ppm?

2. What are the optimum A/F and S/F molar ratios to get the low inlet temperature of PEM fuel cell (70-80°C) with CO concentration less than10 ppm?

1.3 Objectives and Scope of Study

The objective of this study is to find the optimum of A/F and S/F molar ratios of hydrogen production for fuel cell applications from gasoline via autothermal reforming. In order to achieve that objective, several scopes have been drawn:

1) Development of base case simulation

The base case simulation had been developed using the combined reforming of n-octane that involves a complex set of chemical reactions. From these parallel reactions, we got the stoichiometry of the n-octane ATR and calculate the input and output molar flow of the reformate.

2) Validation of base case model

At this stage, the output from the simulation will be compared with the results that from the base case simulation.

3) ATR optimization

The variations of operating parameters such as A/F molar ratios and temperatures will be using to investigate the influence to the hydrogen production and autothermal reformer efficiencies.

4) Heat integration

The heat integration process is very important in order to obtain the most economical reformer because the utilities cost can be reduce.

5) CO clean up

Whereas high temperature fuel cells (MCFC and SOFC) are capable of converting methane, CO and alcohols, etc. in the anode chamber by internal reforming, the PAFC and PEM cells do not tolerate excessive amounts of CO. The PEMFC does not tolerate more than in the order of 50 ppm CO; the lower the CO concentration, the higher the efficiency of the cell.

5.1) WGS

The water gas shift reaction is an inorganic chemical reaction in which water and carbon monoxide react to form carbon dioxide and hydrogen (water splitting). This reaction will reduce the amount of CO besides of producing hydrogen.

5.2) PROX

The preferential oxidation is a parallel reaction in which CO and oxygen react to form carbon dioxide while hydrogen and oxygen react to form water.

6) Plant wide optimization

6.1) WGS

The variations of S/F molar ratios will be using to investigate the influence of these parameters to hydrogen production, temperature and CO concentration.

6.2) PROX

The variations of air that will be injecting to PROX will be using to investigate the influence to the hydrogen production, temperature entering the fuel cell and CO concentration.

1.4 Thesis Organizations

The important of this study is to identify potential design issues and obtain preliminary estimate of the expected system efficiency. So, the simulation of a gasoline autothermal reforming had been constructed in order to identify the autothermal reforming operating conditions and their effect on the overall system performance or efficiency. Therefore, the objective of this study is to simulate and optimize a gasoline autothermal reformer for fuel cell applications using Aspen HYSYS 2004.1. The remainder of this paper is organized as follows, Chapter II describes the literature review of this study and the methodology of this research was described at Chapter III. Chapter IV and Chapter V discussed about steady state simulation of hydrogen production and results and discussion. Lastly, the conclusion and recommendations for future works are drawn in Chapter VI.

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