

SIMULATION AND OPTIMIZATION OF PROPANE AUTOTHERMAL
REFORMER FOR FUEL CELL APPLICATIONS

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To my beloved father and mother

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ABSTRACT

Autothermal reforming (ATR) is one of the leading methods for hydrogen production from hydrocarbons. Liquefied petroleum gas, with propane as the main component, is a promising fuel for on-board hydrogen producing systems in fuel cell vehicles and for domestic fuel cell power generation devices. In this research, autothermal reforming of propane process is studied and operation conditions were optimized using Aspen HYSYS 2004.1 for proton exchange membrane fuel cell application. Furthermore, heat integration process also applied after the existed stream from the ATR reactor. Besides that water gas shift (WGS) which included High Temperature Shift (HTS), Medium Temperature Shift (MTS) and Low Temperature Shift (LTS) reactor, preferential oxidation (PrOx) were used for the clean up system to reduce the concentration of carbon monoxide. Then, optimization for the ATR, WGS and PrOx reactor were done to get the highest hydrogen produced with the lowest CO. Temperature and component's profile were also investigated for every unit's operations. Based on the final result, 100 kgmole/hr of propane with the ratio of air and water 1 : 7 : 4.3, produced 41.62% of hydrogen with CO concentration lower than 10 ppm, and 83.14% fuel processor efficiency.

ABSTRAK

ATR merupakan salah satu kaedah yang terbaik untuk menghasilkan hidrogen daripada hidrokarbon. Cecair gas petroleum dimana propana dijadikan sebagai bahan utama menjanjikan bahan mentah utama untuk menghasilkan system hidrogen untuk kenderaan *fuel cell* dan untuk kegunaan kuasa jana *fuel cell* peralatan domestik. Dalam kajian ini, reactor ATR menggunakan propana adalah dikaji dan operasi sistemnya dioptimumkan menggunakan Aspen HYSYS 2004.1 untuk *fuel cell* aplikasi. Selain itu, integrasi haba juga diterapkan dan diaplikasi selepas aliran keluar daripada reaktor ATR. Di samping itu, WGS dan PrOx proses dijalankan untuk proses pembersihan bagi merendahkan kepekatan CO. Proses pengoptimum dilakukan untuk setiap reaktor bagi menghasilkan hidrogen yang paling tinggi dan pada masa yang sama kepekatan CO terendah. Butiran yang lebih terperinci bagi suhu dan komposisi bahan turut dikaji pada setiap unit operasi. Berdasarkan kepada keputusan akhir yang diperolehi daripada proses simulasi didapati bahawa sebanyak 100 kgmol/j propana dengan nisbah kepada air dan udara sebanyak 1 : 7 : 4.3 telah menghasilkan sebanyak 41.62% hidrogen dengan kepekatan CO dibawah 10 ppm, dan kecekapan sistem penjana adalah 83.14%

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LIST OF ABBREVIATIONS

ATR	-	Autothermal reforming
AES	-	Aspentech Engineering Suite
CH ₄	-	Methane
C ₂ H ₆	-	Ethane
C ₃ H ₈	-	Propane
C ₄ H ₁₀	-	Butane
CHP	-	Combined Heat and Power
CO	-	Carbon monoxide
CO ₂	-	Carbon dioxide
CPO	-	Catalytic partial oxidation
CH ₃ OH	-	Methanol
CMR	-	Compact methanol reformer
CFBMRR	-	Circulating Fluidized Bed Membrane Reformer-Regenerator
ESEM-EDAX	-	Environmental scanning electron microscopy-energy disperse X-ray analysis
GC	-	Gas-chromatographs
H ₂	-	Hydrogen
H ₂ O	-	Water
HSA	-	High surface area
IIR-MCFCs	-	Indirect internal reformer molten carbonate fuel cells
IPOX	-	Indirect partial oxidation
LPG	-	Liquefied petroleum gas
MCFC	-	Molten carbonate fuel cell
MR	-	Membrane reactor

MSR	-	Methane steam reforming
OSR	-	Oxidative Steam Reforming
PAFC	-	Phosphoric acid fuel cell
PEMFC	-	Proton exchange membrane fuel cell
POX	-	Partial oxidation
SE-SMR	-	Sorption-enhanced steam methane reforming
SOFC	-	Solid oxide fuel cell
SPFC	-	Solid polymer fuel cell
SR	-	Steam reforming
TRs	-	Traditional reactors
η	-	Thermal efficiency
γ	-	Activity coefficient
WGS	-	Water Gas Shift
PrOx	-	Preferential Oxidation
PFD	-	Process Flow Diagram

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CHAPTER I

INTRODUCTION

1.0 Background Research

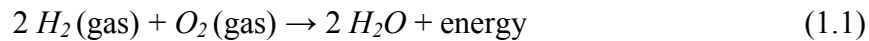
Hydrogen is the most abundant element in the universe. Chemically bound hydrogen is present all over the earth; as part of the Earth's water mass (including fossil substances) (Haussinger et al., 2003). Hydrogen (H₂) is a colourless, odourless, tasteless, flammable and non-toxic gas at atmospheric temperature and pressure. The gas burns in air with a pale blue, almost invisible flame. Hydrogen is the lightest of all gasses, approximately one fifteenth as heavy as air. Hydrogen ignites easily and forms an explosive gas together with oxygen or air.

Hydrogen is used in diverse industries such as a chemical, petrochemical and petroleum refining, metallurgy, glass and ceramics manufacture, electronic and food processing. Hydrogen is used in large quantities as a raw material in the chemical synthesis of ammonia, methanol, hydrogen peroxide, polymers and solvents. It is used in refineries for desulphurization and hydrotreating. It is used in metallurgical industries to provide a reducing atmosphere and in the annealing of steel. The electronic industry uses hydrogen in the manufacture of semiconductor devices. In the food industry, hydrogen is used for hydrogenation of fats and oils. Hydrogen also finds specialty application as a rocket fuel due to its high combustion energy release per unit of weight.(Newson and Truong, 2001).

Recently, there has been a tremendous interest in the development of hydrogen economy based on hydrogen as the energy carrier. Excitement in this field stems from the fact that hydrogen is a potentially non-polluting, inexhaustible and efficient source of energy. Hydrogen can be used in a fuel cell to produce electricity and heat or even be combusted with oxygen to produce energy and the only by product is water (Chris, 2001).

A fuel cell by definition is an electrical cell, which unlike storage cells can be continuously fed with a fuel so that the electrical power output is sustained indefinitely (Reed, 1995). They convert hydrogen or hydrogen-containing fuels, directly into electrical energy plus heat through the electrochemical reaction of hydrogen and oxygen into water. The process is that of electrolysis in reverse.

Overall reaction:



Because hydrogen and oxygen gases are electrochemically converted into water, fuel cells have many advantages over heat engines. These include high efficiency, virtually silent operation and, if hydrogen is the fuel, there are no pollutant emissions. If the hydrogen is produced from renewable energy sources, then the electrical power produced can be truly sustainable. The two principle reactions in the burning of any hydrocarbon fuel are the formation of water and carbon dioxide (Espinal et al., 2005). As the hydrogen content in a fuel increases, the formation of water becomes more significant, resulting in proportionally lower emissions of carbon dioxide. As fuel use has developed through time, the percentage of hydrogen content in the fuels has increased. It seems a natural progression that the fuel of the future will be 100 % hydrogen.

Fuel cells are classified according to the electrolyte use. There are the alkaline fuel cell used in space vehicle power systems, the phosphoric acid fuel cell (PAFC) used in both road transportation and stationary engines, the solid polymer fuel cell (SPFC) also used in both road transportation and stationary engines, the molten carbonate fuel cell (MCFC) used in stationary engines and the solid oxide fuel cell (SOFC) used only in stationary engines (Chen and Elnashaie, 2004)

In general, fuel cells can be used for a wide variety of applications, the most important of which are as a power source for vehicles, as a stationary power source (for example for large-scale power plants generation, power generation in the home or small power plants for larger industrial or residential sites) and as a power sources for portable devices (for example laptops, cameras, mobile phones, for replacement of batteries etc.). Natural gas and propane are attractive for stationary applications since they are low-cost fuels and the infrastructure for their transportation already exists (Wang et al., 2005).

1.1 Problem Statement and Importance of Study

This research aims to develop an optimized model of hydrogen production plant using propane for fuel cell application by autothermal reforming. In order to analyze its performance, well-defined steady state model that will represent the real plant for hydrogen production is required. In order to do that, Aspen HYSYS 2004.1 is utilized. The important to have this optimized model is to analyze design parameter for fuel processor, and also to get preliminary fuel processor efficiency.

1.2 Objective and Scopes of Study

Objective of this study is to develop an optimized model of hydrogen production plant using propane for fuel cell applications by autothermal reforming. In order to achieve that objective, the following scopes have been drawn:

- i). Steady-state Model for Base Case
 - Steady-state model of hydrogen plant from propane was carried out by stoichiometry mathematical analysis calculation and the simulation using Aspen HYSYS 2004.1.

- ii). Steady-state Model Validation for Base Case
 - Steady-state model developed within Aspen HYSYS 2004.1 process simulator was validated using data from stoichiometry mathematical analysis calculation

- ii). Process Heat Integration Model Development
 - Heat integration model was applied for this simulation using Aspen HYSYS 2004.1.

- iii). Clean Up Model Development
 - Clean up model was applied to reduce the concentration of carbon monoxide at the reformer.

- iv). Plant Wide Optimization
 - Every reactor had been optimized to achieve the highest hydrogen and the lowest carbon monoxide.

- v). Temperature and Components Profile Analysis
 - To get the overall overview of the process of the reformer.

1.3 Thesis Organization

The realization of the objective of this thesis involves the culmination of a number of tasks. The first task, introduced in chapter two is to do the literature survey about the synthesis of hydrogen for fuel cell applications. In this chapter, an internal research of hydrogen production using propane by autothermal reforming was been concentrated. This chapter is the most important chapter because we developed the method of hydrogen synthesis are based on the literature survey that we had done.

Chapter three is methodology for this thesis whereby the arrangements for the methods are based on the scopes. Basically, there are five methods that we carried out. Then, chapter four is optimization simulation of hydrogen production plant from propane for fuel cell application. In this chapter we developed the simulation using Aspen HYSYS 2004.1.

Chapter five is the results and discussion whereby the results are based on the methodology that is developed from chapter four. Then, chapter six presents the conclusions and recommendations.

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