ENHANCEMENT OF PRODUCER GAS QUALITY THROUGH CO_2 ABSORPTION USING CaO-SAND MIXTURE IN A FLUIDIZED BED REACTOR

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by

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LIST OF ABBREVIATIONS

atm	Atmosphere
BFB	Bubbling Fluidized Bed Gasifier
CFB	Circulating Fluidized Bed Gasifier
CFD	Computational Fluid Dynamics
CO_2 BFBAR	Bubbling fluidized bed CO_2 absorption reactor
CPG	Compressed Producer Gas
FLR	Fluidized limestone reactor
g	Gas
HV	Heating value
HyPr-PING	Hydrogen production by reaction-integrated novel gasification
IEA	International Energy Agency
LNG	Liquefied natural gas
MOX	Malaysia Oxygen Company
P_{eq}	Equilibrium decomposition pressure
pg	Producer gas
2D	Two Dimensional
3D	Three Dimensional
Q	Volume flow rate
RTP	Envergent Rapid Thermal Processing
S	Solid
SPG	Simulated Producer Gas
TCD	Thermal conductivity detector
ΔP	Pressure drop

LIST OF SYMBOLS

A_r	Aspect Ratio
A	Surface Area (m^2)
B	Constant in Syamlal-O'Brien drag model
C_D	Drag factor on single particle system
$Ca(OH)_2$	Calcium hydroxide
$CaCO_3$	Calcium carbonate
CaO	Calcium oxide
CH_4	Methane
CO	Carbon monoxide
CO_2	Carbon Dioxide
d_i	Inner diameter (m)
exp	Exponent
e	Restitution coefficient of solid phase
F_{drag}	The general drag force (kg/ms^2)
g	Gravity (m/s^2)
g_o	The general radial distribution function
h	Height (m)
H_2	Hydrogen
h_s	Static bed height (m)
In	In
$\bar{\bar{I}}$	The unit tensor
K_{sg}	Drag factor of phase s in phase g (kg/m^3s)

L	Length (m)
W	Width (m)
H	Height (m)
N_2	Nitrogen
N	Normal
P	Pressure (Pa)
P_s	Solids pressure (Pa)
Re	The Reynolds number
Re_s	The particle Reynolds number
^{o}C	Degree Celcius
Out	Out
P	Thermal Power (Watt)
T	Temperature (^{O}C)
t	Time (sec)
u_{mf}	Minimum fluidizing velocity (m/s)
ρ	Density (kg/m^3)
η_{diesel}	Efficiency of diesel engine
$d_s^{\bullet}$	Dimensionless particle size
Ar_{numb}	Archimedes number
μ_g	Dynamic viscosity (kg/ms)
m	Mass (kg)
A	Cross sectional area (m^2)
$u_t^{\bullet}$	Dimensionless terminal velocity
ΔP	Pressure drop (Pa)

d_s	Diameter solid particles size (m)
ϕ_s	Sphericity of solid particle
u_t	Terminal Velocity (m/s)
P_{inpg}	Thermal power input (Watt)
%	Percent
(Na_2CO_3)	Sodium carbonate
(NaOH)	Sodium hydroxide
$\nabla . q_s$	Diffusive flux of fluctuating energy (kg/ms^3)
$\vec{V}$	Velocity (m/s)
v_r	The relative velocity correlation
ε_g	Gas phase volume fraction
ε_g	Solid phase volume fraction
γ_{Θ_s}	Dissipation of granular temperature (kg/ms^3)
Δ	Change in variable, Final-Initial
∇	The Dell operator (1/m)
Θ_s	Granular temperature (^{o}C)
λ_s	Bulk viscosity (kg/ms)
μ_g	Gas viscosity (kg/ms)
$\mu_{s,}$	Granular viscosity (kg/ms)
$\mu_{s,col}$	Collisional viscosity (kg/ms)
$\mu_{s,kin}$	Kinetic viscosity (kg/ms)
$\mu_{s,fric}$	Frictional viscosity (kg/ms)
Π	The irrational number Π

P_g	Gas density (kg/m^3)
P_s	Solid density (kg/m^3)
$\overline{\overline{\tau}}$	The stress-strain tensor (Pa)

PENINGKATAN KUALITI GAS TERHASIL MELALUI SERAPAN CO_2 MENGGUNAKAN CAMPURAN CaO – PASIR DI DALAM REAKTOR LAPISAN TERBENDALIR

ABSTRAK

Tesis ini adalah mengenai kajian ujikaji bagi meningkatkan kualiti gas yang dihasilkan dari proses penggasan melalui konsep penyerapan karbon dioksida (CO_2) menggunakan campuran kalsium oksida (CaO) bersama pasir di dalam reaktor penyerapan lapisan terbendalir gelembung CO_2 (CO_2 BFBAR). Selain itu kajian ini juga menggunakan kaedah simulasi berangka. Pada masa kini, penggasan biojisim merupakan satu alternatif yang dapat digunakan untuk mengantikan tenaga bahanapi fosil. Gas terhasil, yang terhasil daripada proses penggasan biojisim boleh digunakan untuk menjanakan kuasa dan elektrik. Walaubagaimanapun, CO_2 yang terdapat di dalam gas terhasil mengurangkan nilai pemanasannya kerana CO_2 bertindak sebagai bahan pencair. Penggunaan batu kapur yang mengandungi terutamanya bahan mineral kalsit (kalsium oksida) sebagai bahan pengerap berasaskan kalsium untuk menyerap CO_2 yang terdapat di dalam gas terhasil boleh menjadikan teknologi biojisim ini lebih berdaya maju. Pengisian dinamik bendalir berkomputer (CFD) 3 dimensa digunakan untuk melakukan simulasi bagi mencari ciri-ciri hidrodinamik bagi bahan lapisan CaO-pasir yang diletakkan dalam CO_2 BFBAR. Keputusan simulasi yang diperolehi bagi CaO bersaiz 100, 500 dan 1000 micron menunjukkan bahawa pembendaliran terhasil dengan baiknya pada kadar alir isipadu 15-55 L/min menggunakan udara. Selain itu, CO_2 BFBAR juga telah dibangunkan dan kajian terhadapnya pada kelakuan bahan campuran CaO-pasir juga dilihat dari sudut ujikaji model sejuk. Kesan-kesan disebabkan oleh

campuran CaO-pasir, saiz zarah CaO, kadar aliran isipadu dan tekanan masukan udara disiasat secara ujikaji. Proses penyerapan-nyahserapan oleh CaO juga dikaji menggunakan Penganalisa Termogravimetri (TGA) terhadap 1 kitar, 4 kitar dan kitar berbagai. Tiga suhu yang berbeza (500, 600 dan 700^{o}C) disetkan sebagai pembolehubah. Kadar tindakbalas CaO diperolehi. Keputusan menunjukkan pada kitar pertama, kadar tindakbalas penyerapan CO_2 adalah cepat semasa di peringkat pertama kemudian diikuti dengan kadar tindakbalas penyerapan yang perlahan. Sebanyak 0.337 dan 0.065 mg/min kadar tindakbalas penyerapan CO_2 didapati untuk kawasan penyerapan pantas dan perlahan. Diperhatikan juga bahawa kadar tindakbalas penyerapan CO_2 berkurangan apabila bilangan kitar penyerapan-nyahserapan ditambah. Selepas ujikaji TGA dijalankan, ujikaji model panas pula dilakukan untuk mengkaji kebolehkesanan penyerapan CO_2 mengunakan nilai-nilai optimum yang diperolehi dari ujikaji model sejuk. Gas simulasi yang terdiri daripada 20% CO_2 dan 80% N_2 digunakan dalam CO_2 BFBAR sepanjang suhu 650-750^{o}C. Pembendaliran terhasil dengan baiknya pada keadaan 50 dan 40 peratus campuran CaO bagi semua tekanan (2-6 bar). Saiz zarah 1000 micron pada campuran CaO-pasir dan kadar aliran isipadu udara antara 15-55 L/min juga memberikan pembendaliran yang baik. Di dalam ujikaji model panas pula, penyerapan CO_2 yang baik terhasil pada keadaan campuran 50% CaO mengunakan tekanan gas simulasi pada nilai 3 bar dengan kadar alir isipadu 45 L/min sepanjang suhu 650-750^{o}C dalam CO_2 BFBAR. Penggunaan gas terhasil simulasi dan gas terhasil bertekanan juga menunjukkan keputusan yang baik. Kepekatan CO_2 dalam gas terhasil simulasi menurun kepada 57.5%, manakala hidrogen dan karbon monoksida pula meningkat kepada 12% dan

6% selepas beroperasi selama 10 minit. Apabila mengunakan gas terhasil bertekanan, kepekatan CO_2 juga menurun sebanyak 77.4%, manakala hidrogen dan karbon monoksida pula meningkat kepada 23.3% dan 21.7%. Oleh sebab itu, nilai pemanasan gas terhasil bertekanan secara langsung juga meningkat dari 4.51 MJ/*N*m^3 kepada nilai semaksimum 6.04 MJ/*N*m^3 iaitu peningkatan sebanyak 38%.

ENHANCEMENT OF PRODUCER GAS QUALITY THROUGH CO_2 ABSORPTION USING CaO-SAND MIXTURE IN A FLUIDIZED BED REACTOR

ABSTRACT

This thesis concerns an experimental study and numerical simulation used to enhance the producer gas quality through carbon dioxide (CO_2) absorption using calcium oxide (CaO) - sand mixture in a CO_2 bubbling fluidized bed absorption reactor (CO_2 BFBAR). Biomass gasification is a thermo-chemical conversion process of solid biomass into gaseous fuel called producer gas that can be used to generate power and electricity. However, carbon dioxide (CO_2) content in the producer gas reduces its heating values as CO_2 acts as a diluent. The use of limestone consisting mainly of the mineral calcite (calcium oxide, CaO) as calcium based sorbent to absorb CO_2 in the producer gas will increase the heating value of the producer gas. 3-D Computational Fluid Dynamics (CFD) software was used to determine hydrodynamic characteristic of CaO-sand bed material in a CO_2 BFBAR. The simulation results show that with 100, 500 and 1000 micron particle size of CaO, good fluidization at 15 – 55 L/min volume flow rate of air was achieved. The CO_2 BFBAR was developed and the behavior of CaO-sand mixtures in a cold model experiment was studied. The effect of the CaO-sand mixtures, the CaO particle sizes, the volume flow rate and the pressure of air intake were investigated experimentally. The absorption-desorption process of CaO was studied with the thermogravimetric analyzer (TGA) over 1, 4 and a muticycle. Three differents temperature (500, 600 and 700^{o}C) were set a variables. The reaction rate of CaO was obtained. Results show that for number 1 cycle the CO_2 absorption

reaction rate was fast at first stage and then followed with slower reaction rate. About 0.337 and 0.065 mg/min CO_2 reaction rate were obtained in rapid and slow absorption regime respectively. It is also observed that the CO_2 absorption reaction rates decreases when number of cycle's desorption-absorption process was increased. After TGA experiment, the hot model experiment was conducted to investigate CO_2 absorption at the optimum condition obtained from the cold model experiment. The simulated gas consisting of 20% CO_2 and 80% N_2 was introduced in the CO_2 BFBAR at temperatures of 650-750^oC. The CaO percentages of 50 and 40 in sand were found to have a good fluidization at all air pressures (2 - 6 bar). In addition to that, the 1000 micron particle size of the CaO–sand mixture and the volume flow rate of air between 15 – 55 L/min were also found to generate good fluidization. In the hot model experiment, the best CO_2 absorption occurs in 50% CaO mixture with simulated gas, at pressure of 3 bar and the volume flow rate of 45 L/min at 650-750^oC in the CO_2 BFBAR. The CO_2 concentration in the simulated producer gas when applied decreases approximately 57.5%, where this resulted in increases of H_2 and CO to approximately 12% and 6%, respectively within 10 minutes of operation. For the compressed producer gas, the CO_2 concentration decreases approximately 77.4%, where H_2 and CO increase approximately 23.3% and 21.7%, respectively. Therefore the heating value of the compressed producer gas increases from 4.51 to maximum of 6.04 MJ/Nm^3 an increase of 38%.

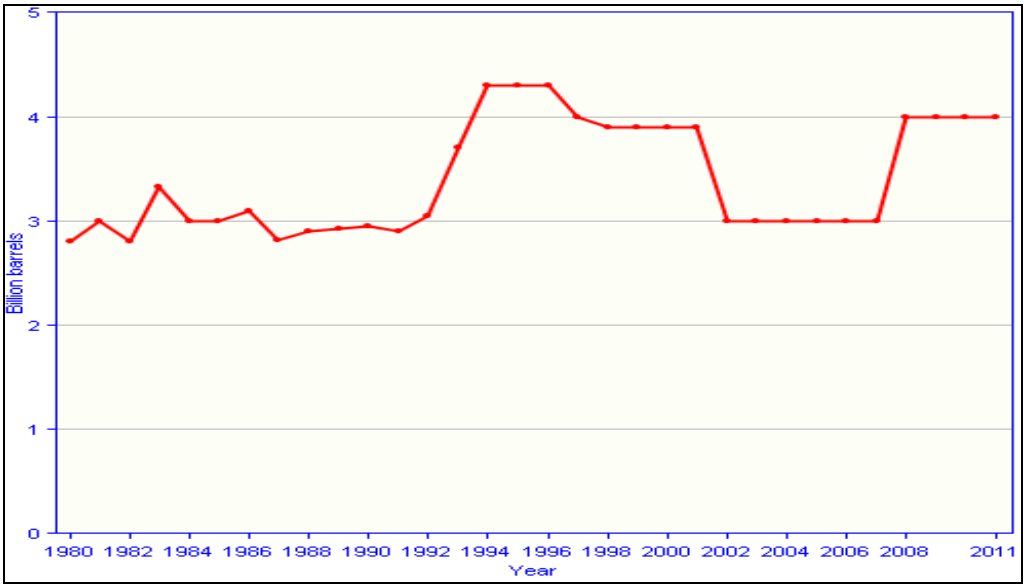
CHAPTER ONE

INTRODUCTION

1.1 Fuel Scenario in Malaysia

Fuel is undoubtedly the most important source of energy. It has been used predominantly in power plants and to generate work in internal combustion engines. However, the extensive exploitation of fossil fuels for power has given rise to a number of serious problems namely depletion of fuel reserves, price inflation of raw materials, adverse effects on the environment due to emissions from combustion devices and increment of greenhouse gas emissions.

Malaysia is currently known as a significant producer of oil and it oil reserves is the third highest in the Asia-Pacific region after China and India. Interestingly, Malaysia is a net exporter of liquefied natural gas (LNG) and is known as the third largest in the world after Qatar and Indonesia in 2010 (Energy Information Administration Report, 2011). According to Oil and Gas Journal (2011), in January 2011, Malaysia had proven to have oil reserves of 4.0 billion barrels. Although the oil reserves are large, it is decreasing from a peak of 4.3 billion barrels in 1996 as shown in Figure 1.1. In term of oil production in Malaysia, it is reducing but from there is an increase in consumption (Figure 1.1, below). These unparalleled situations will cause the country's oil reserves to become exhausted in the future unless new explorations show positive results.



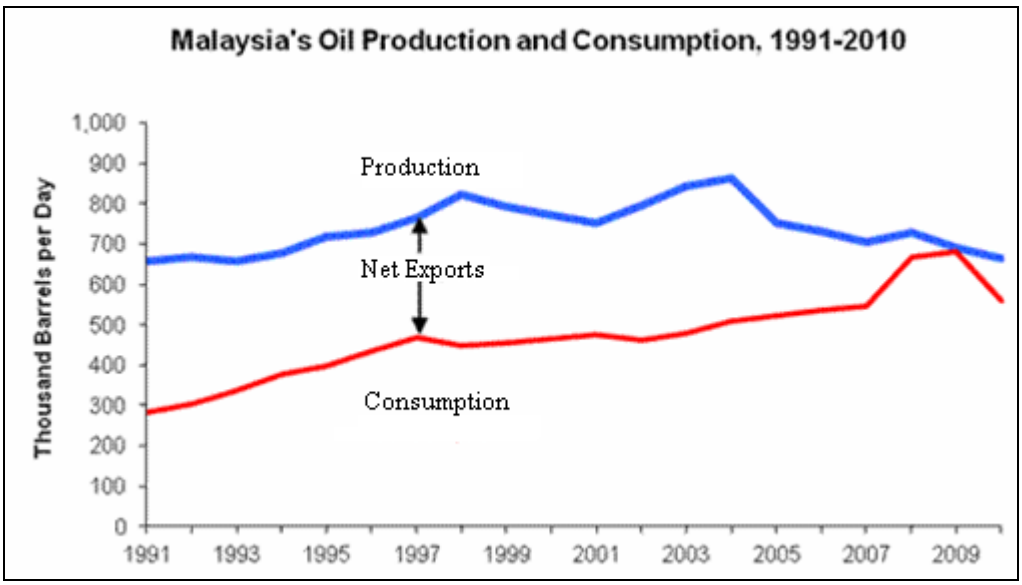


Figure 1.1- Malaysia oil reserves (Top), Malaysia oil production and consumption (below) (EIA, 2011)

Growing demand for crude oil or petroleum (element in fossil fuel) in Malaysia which is set to become a developed nation by 2020 will result in higher energy cost and greater dependence on imported oil given the current crude oil capacity (Bari *et al*. 2011). This can have a potential negative impact on the nation's economic growth as rising commodity prices are closely tied to

inflation rates. That is why many developed as well as developing countries, Malaysia included, have tried to utilize alternative and renewable sources of energy.

Renewable energy offers opportunity to lower fossil fuel consumption. Energy derived from solar, wind, wave, tidal, hydroelectric, geothermal and biomass sources are considered renewable. Because most forms of renewable energy are derived either directly or indirectly from the sun, there is abundant supply of renewable energy available, unlike fossil fuels. The use of renewable energy also provides environmental, economic and political benefits, and they are also not subjected to depletion in time. One of the fuels for renewable energy is biomass.

1.2 Biomass

Biomass is the most abundant resources and available in all parts of Malaysia. It has the potential to be one of the best options for providing on demand renewable fuel that can be utilized in various energy conversion technologies and also has the advantage of being carbon sink without contributing to the net production of carbon dioxide, CO_2. Biomass is defined as organic materials existing in all plants and animals such as forest and mill residues, wood wastes, agricultural crops and wastes, animal wastes and municipal solid wastes.

Malaysia abundant biomass wastes come from its wood, oil palm and agro-industries. Malaysia produces more than 70 million tones per year of biomass wastes worth RM 23 million annually. Over the next 20 years, palm oil industry is expected to expand by 40%, hence the value of residues from wood and oil palm is estimated to be about RM 500 million by 2020. Of these residues, wood waste, shells, empty fruit bunch and fibres provide the greatest potential for commercial operations (Malaysia Energy Centre, 2008). For instance, in year 2011 Malaysia used Envergent Rapid Thermal Processing (RTP) technology to perform the engineering design to convert palm biomass to renewable heat and electricity. This RTP facility will complete in early 2013, and it will be Malaysia first plant to use RTP for the production of a clean-burning liquid biofuel derived from biomass for the purpose to generate renewable electricity and heat (PR Newswire, 2011).

In biomass technologies, there are two main processes for converting biomass sources into useful forms of energy: thermochemical and biochemical/biological chemistry (McKendry, 2002b). In thermochemical conversion, there are four processes: combustion, pyrolysis, gasification and liquefaction. In this research study only the thermochemical in particular gasification process will be focused. Figure 1.2 shows the thermochemical process, intermediate energy carriers and final energy products resulting from the thermochemical conversion.

Biomass gasification is a technology to produce low to medium energy fuel gas (Klass, 1998) for internal combustion engines or coupled to turbines to generate power. Gasification is defined as the thermochemical conversion of carbonaceous feedstocks such as biomass and coal into producer gas or synthesis gas (syngas - composed primarily of hydrogen and carbon monoxide) using air, oxygen or steam to react with the solid fuel at high temperature, typically in the range of 800-900^{O}C. Only inert ashes by products are produced at the end of the process. According to Rezaiyan (2005) and McKendry (2002b), the producer gas produced has heating value (*HV*) around 4-6 MJ/*N*m^{3}.

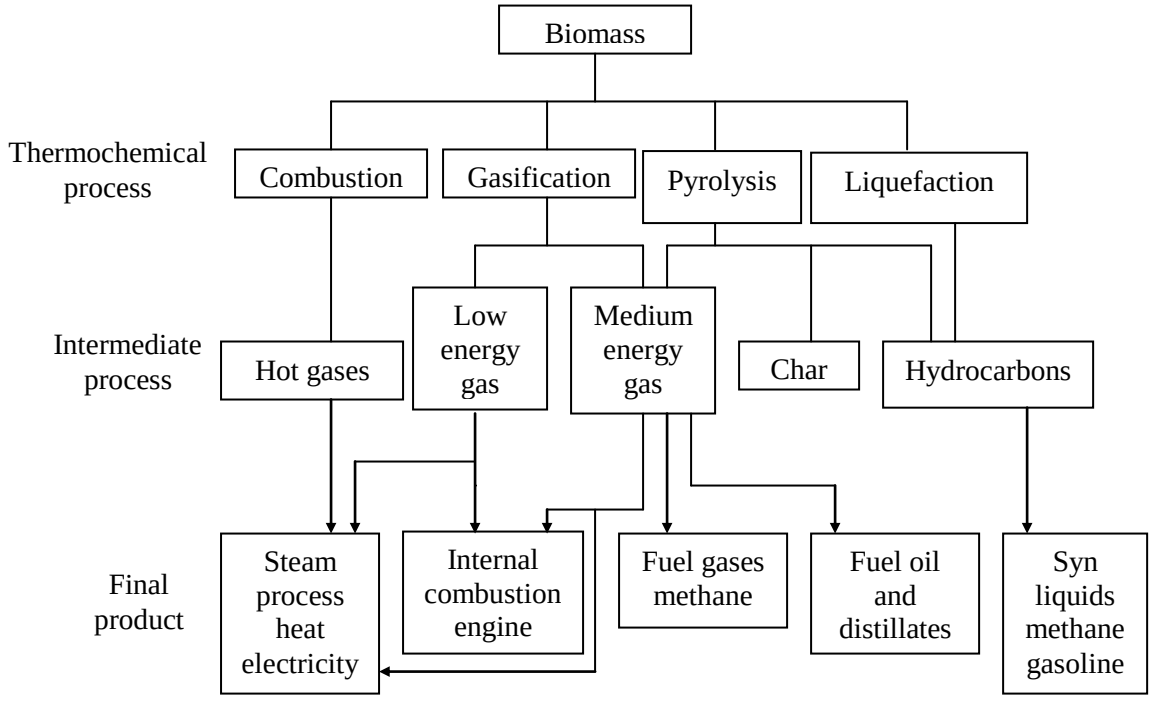


Figure 1.2: The thermochemical processes and products.

(McKendry, 2002b)

1.3 Carbon Dioxide Capture

Carbon dioxide (CO_2), which is containing in producer gas-air mixture is only 9-15% carbon dioxide, and others are hydrogen (15-20%), carbon monoxide (10-15%), methane (3-5%) and 40-50% of nitrogen by volume (Sen, 2005 and Reed, 1988). CO_2 content in the producer gas reduces its heating values as CO_2 acts as a diluent. Removing CO_2 from the producer gas will inadvertently increase its heating value. It will also improve the percentage of its hydrogen and all combustible gas contents.

Recently, there are a few methods have been proposed for CO_2 capture. The methods for capturing CO_2 from flue gases are membrane separation (Dindore *et al*., 2004), cryogenic fractionation (Khoo *et al*., 2006), and solvent absorption (Al-Juaied *et al*., 2006), either physical or chemical sorption on solid surfaces (Gupta *et al*., 2002). However, these two methods such as membrane separation and cryogenic fractionation have not favored for CO_2 separation. Example membrane separation systems, even though are highly efficient and have been employed for the separation of CO_2, but due to their complexity, high energy cost, and limited performance, membrane systems are not entirely well suited. As well as cryogenic fractionation systems, it also required high energy requirements.

Solvent absorption, on the other hand is well recognized. It is using various solvents, for instance, Selexol (Kohl *et al*. 1997) as a physical solvent or mono-ethanol amine (MEA) (Filburn *et al*., 2005) as a chemical solvent. However, according to Rao *et al.* (2002) severe energy penalties and the high

cost of the system are significant disadvantages of the method especially on the use of amine. The low concentration of CO_2 in the flue gases at atmospheric pressure and low temperature (40 - 150^oC) required for the absorption and solvent recovery process leading to the high cost of system.

The only one of the most promising method is based on the reversible absorption of CO_2 on specific metal oxides at high temperature. CO_2 capture using sorbents based on the oxides of calcium (Manovic *et al.*, 2009), potassium (Lee *et al*., 2006), lithium (Fauth *et al*., 2005), sodium (Knuutila *et al*., 2009) and magnesium (Lee *et al*., 2008) have been reported. The most have attention owing among these is calcium oxide (CaO) based sorbents because of their wide availability, low cost, higher absorption capacity and high selectivity for CO_2.

According to Chen *et al*. (2009), Alvarez *et al.* (2007), Lu *et al.* (2008) and Akiti *et al.* (2002), precursors such limestone (also known as Calcium Carbonate, $CaCO_3$), dolomite, calcium acetate and calcium sulphate hemihydrate can be processed to derive CaO. Among these, the most common CaO precursors are limestone. This is because of the availability and low cost of limestone as mention. According to Zulasmin (2007), Malaysia country is blessed with abundant reserve of limestone resources. Extensive limestone resources are located in the states of Perak, Pahang, Kelantan, Kedah and Negeri Sembilan. It was estimated over 10 billion tonnes of limestone resources throughout the country. Example in the state of Perak, there is a 0.0405 km^2 limestone quarry and it estimated limestone reserve of 4

million tones. With current monthly usage of 5000 tonnes per month, the quarry can provide raw limestone for the next 66 years (Zantat 2009). Talking about the price of raw limestone, it is sold for only RM60 a tonne compared to processed and value-added limestone that can fetch around RM390 a tonne, according to Elan (2011).

The reaction of solid CaO with CO_2 can be shown as in Equation 1.1 called absorption, and this is a spontaneous exothermic process at ambient conditions. At elevated temperatures, the reversed endothermic reaction called desorption (Equation 1.2) occurs.

$$CaO(s) + C0_2(g) \text{ --- } CaC0_3(s) \quad (1.1)$$

$$CaC0_3(s) \text{ --- } CaO(s) + C0_2(g) \quad (1.2)$$

In theory the reactions in Equations 1.1 and 1.2 are fully reversible; thus, they can be appropriated to capture CO_2 from producer gas and upon desorption, CO_2 will be released. The cycle of desorption and absorption is repeated over and over. For such a process, two situations and temperatures are employed. Desorption is performed at higher temperature above 800^oC and absorption at temperature below 800^oC. The absorption–desorption reaction will be discussed in detail in Section literature.

1.4 Problem Statements

The need for doing research on biomass sources is due to the fossil fuels is non-renewable. They are limited in supply and will one day be depleted. By 2050, the depletion of fossil fuels is expected to be worse and alternative fuels have to be discovered (Takuyuki Yoshioka *et al*., 2005). Hence the search for alternative energy sources for sustaining energy requirement by human beings must be done because the depletion of fossil fuels is occurring at a faster rate due to increasing gap between demand and production of fossil fuels. This problem is the foremost reason why renewable sources of energy must be studied.

The fossil fuels combustion emits carbon dioxide (CO_2), a green house gas will contributes to global warning. Magnus *et al*. (2006) wrote that CO_2 concentration in the atmosphere is 30% higher than it was before the industrialization era and the annual emissions are still increasing today. To overcome this problem, renewable energy sources such as biomass can be used as fuel. Burning biomass contributes no net CO_2 to the atmosphere because replanting biomass will absorbs the CO_2.

The unused biomass wastes are abundant whilst managing them is difficult and expensive without any added value. According to Wan Asma *et al*. (2010), unused biomass wastes are collected more than 15 million tonnes per year. The wastes can be used as fuel to generate energy with added value.

Producer gas has small hydrogen concentration, approximately 6-20% by volume when using a downdraft biomass gasifier (Munoz *et al*., 2000; Sridhar *et al*., 2001; Zainal *et al*., 2002; Dogru *et al*., 2002; Adnan *et al*., 2002 and Uma *et al*., 2004). To increase the percentage of the hydrogen, a new method has to be studied and developed. For instance, the producer gas from biomass gasification system contains high amount of CO_2, 15-20% (Sridhar *et al*. 2001 and Dogru *et al.* 2002) thereby lower its heating value as CO_2 acts as a diluent. Removing CO_2 from the producer gas will improve its heating value. The heating value of producer gas will be expected to increase from 4 MJ/*N*m^3 to about 6 MJ/*N*m^3. This will not just improve its heating value but inadvertently improve the amount of combustible gases such as hydrogen, carbon monoxide and methane.

With these problems statement, there are two contributions can be raised up. The first is the improvement of compressed producer gas-air from downdraft gasifier. As mentioned in literature section, there appears no systematic study on the useage of CaO-sand mixture for CO_2 absorption process by CaO in downdraft gasifier application. The second contribution is the study of CO_2 absorption in the CO_2 BFBAR. The concept of the kinetic reaction between CaO and CO_2 are focused. Therefore, there is still room for researchers to study the concept, development and application of CO2 absorption by CaO to producer gas from the downdraft gasification process for future use, particularly in improving the quality of producer gas.

1.5 Research Objectives

The main objective of the work presented in this thesis is to enhance the quality of the producer gas using CaO as absorbent reagent mixed with sand. The CO_2 gas contained in the producer gas is absorbed via a reactor known as bubbling fluidized bed CO_2 absorption. The sub-objectives are:

- To simulate the hydrodynamic characteristic of a bubbling fluidized bed CO_2 absorption reactor using computer software, FLUENT.
- To design and develop a bubbling fluidized bed CO_2 absorption reactor.
- To conduct cold study to characterize the hydrodynamic behavior of the system using CaO – sand mixtures.
- To characterize the bubbling fluidized bed CO_2 absorption reactor using absorption – desorption process with the electric ceramic heater band.
- To determine the CO_2 absorption reaction rate of CaO using Thermogravimetri analysis (TGA).
- To test and analyze the bubbling fluidized bed CO_2 absorption reactor with simulated producer gas (SPG) and actual compressed producer gas (CPG) from downdraft gasifier.

1.5 Research Scope

The scope of the research can be divided into four stages as follows:

1. Study on calcium oxide as absorbent of CO_2 gas, focusing on absorption – desorption processes.

2. Study on the 3-D computational fluid dynamics software such as FLUENT to simulate the hydrodynamic phenomena of CaO in the bubbling fluidized bed CO_2 absorption reactor (CO_2 BFBAR).

3. Study on the development of the bubbling fluidized bed CO_2 absorption reactor based on the principles of bubbling fluidization.

4. Study on the biomass material such as furniture wood and the process of gasification, focusing on the small downdraft gasifier.

1.7 Organization Chapters

Chapter 1 contains the overview and the direction of the study. It highlights the background of the research in which the scenario of fuel in Malaysia is discussed, followed by an introduction on biomass and CO_2 gas capture. Problems statement explaining the reasons why this research is done and the contributions are also clearly stated. The objectives and scopes of the research are listed to provide preliminary knowledge about the direction of this project.

Chapter 2 contains relevant theories, concepts and literature review of past researches to strengthen the framework of the research. Various elements associated with biomass are described. The detail of absorption process of carbon dioxide and the simulation method obtained from other researchers are also listed and discussed.

Chapter 3 represents the design and development of a bubbling fluidized bed CO_2 absorption reactor (CO_2 BFBAR). The dimensions of the CO_2 BFBAR are shown and thoroughly described.

The experimental setup, procedures and experimental studies are detailed in Chapter 4. The description of the apparatus and their usage are also included.

The results and discussion of the experiments conducted are presented in Chapter 5. All results obtained are arranged in tables, graphs and figures, and carefully laid out for easy reference. Operations of the moisture content test, bomb calorimeter test, 3-D FLUENT simulation, statistical analysis, gasifier experiment, cold model, thermogravimetric analysis (TGA) and hot model experiment for the CO_2 absorption are also stated and discussed.

Finally in Chapter 6, summarizes and concludes the findings of the study are made and recommendations suggested for future work are listed to complete the thesis.

CHAPTER TWO

LITERATURE REVIEW

2.1 Introduction

In this chapter, a literature review has been done on the types of gasifier and chemical conversion of biomass material via the gasification process into gaseous fuel. This includes an elaboration on the combustible components of the gas, as well as the work of other researchers on the process of increasing the percentage of hydrogen, the absorption process of CO_2 and simulation method.

2.2 Biomass Gasification

Biomass is a non-fossil, energy-containing form of carbon and includes all land and water based vegetation. It is the only indigenous renewable energy resource that is capable of displacing large amounts of solid, liquid and gaseous fossil fuels. Biomass can be used for generating electric power and industrial processes. One of the methods used to convert biomass into useful form of energy is called gasification, producing combustible gas, which can be used in internal combustion engines. The gases typically consist of hydrogen (H_2), carbon monoxide (CO), methane (CH_4), carbon dioxide (CO_2) and nitrogen (N_2) commonly named as “producer gas”. According to Zhang *et al*. (2004), biomass when burnt or gasified will produce dirty raw gas mixture or producer gas composed of hydrogen, carbon monoxide, carbon dioxide, water, methane and various light hydrocarbons along with undesirable dust

(ash and char), tar, ammonia, alkali (mostly potassium) and some other trace contaminants.

2.2.1 Types of Gasifier

The gasification process occurs in a reactor called gasifier. There are basically two types of gasifiers: fixed bed and fluidized bed. The fixed bed gasifier has been the traditional process used for gasification and operates at high temperature around 1000^{o}C. It can be classified as downdraft, updraft and cross flow, depending on the direction of the flow (McKendry, 2002c).

2.2.2 Fixed Bed Gasifier

A downdraft gasifier, where the biomass feed and air move in the same direction or co-current. The product gases leave the gasifier after passing through the hot zone, enabling partial cracking of the tar formed during gasification and leaving a producer gas with low tar content. The overall energy efficiency of the downdraft gasifier is low because the gases leave the gasifier unit at a high temperature of 900-1000^{o}C and due to the high heat content carried over by the hot gas (McKendry, 2002c). Figure 2.1 shows a diagram of a downdraft gasifier.

In an updraft gasifier, biomass feed is introduced at the top and the air flows from the bottom of the unit via a grate in a counter current flow. The solid char forms higher up the gasifier above the combustion zone. An updraft gasifier is simple in design and can handle biomass with high moisture

content up to 50% by weight (McKendry, 2002c). Figure 2.2 shows a diagram of an updraft gasifier.

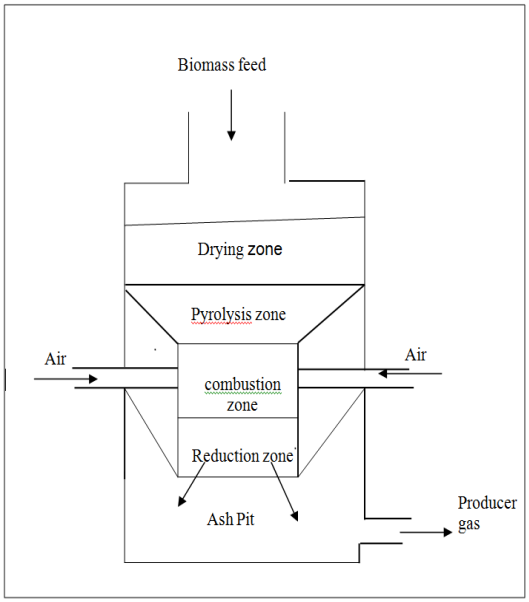


Figure 2.1: Diagram of a downdraft gasifier

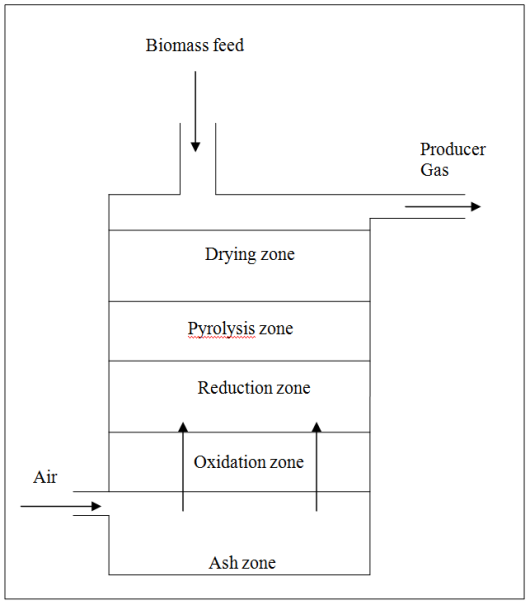


Figure 2.2: Diagram of an updraft gasifier

The gasifier where the air is introduced from the side while the feed biomass moves downwards is called cross flow gasifier. The producer gas is drawn from the opposite side of the unit at the same level of the air introduced. A hot gasification zone forms around the entrance of the air, with the pyrolysis and drying zones being formed higher up in the vessel. Ash is removed at the bottom and the temperature of the gas leaving the unit is 800-900^{o}C. This gives low overall energy efficiency for the process and a gas with high tar content (McKendry, 2002c). Figure 2.3 shows the diagram of a cross flow gasifier.

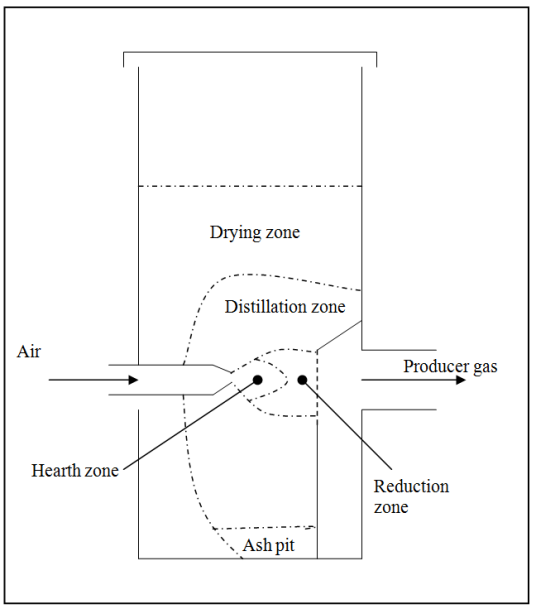


Figure 2.3: Diagram of a cross flow gasifier

2.2.3 Fluidized Bed Gasifier

The fluidized bed gasifier has been used extensively for coal gasification for many years (Garcia-Ibanez *et al*., 2001). It is favored by many gasifier designers for using smaller feedstock sizes. In the gasification zone of the fluidized bed, a uniform temperature distribution is achieved. There are

two main types of fluidized bed gasifier that can be classified as: bubbling fluidized bed and circulating fluidized bed (Fouilland *et al*., 2010 and McKendry, 2002c).

A bubbling fluidized bed gasifier (BFB) consists of a vessel with a grate at the bottom through which air is introduced. Above the grate is the fluidized bed of fine-grained materials into which the prepared biomass feed is introduced. The BFB utilizes minimum fluidization velocity of sand and fine-grained bed material to achieve fluidization state. The sand acts as a heat transfer medium. The ash entrained out of the gasifier is collected in a cyclone separator. For biomass material with high moisture content and low heating value, BFB is recommended (Ciferno and Marano, 2002). A detailed concept of the BFB will be discussed in Section 3.2 and the diagram of the bubbling fluidized bed is shown in Figure 2.4 (a).

A circulating fluidized bed is suitable for waste fuels with a high percentage of non-combustibles (heating value 5 - 35 MJ/kg). It operates at a temperature around 800-900^{0}C (McKendry, 2002c). Crushed coal along with sorbent (limestone) is fed to the lower furnace where it is kept suspended and burnt in an upward flow of combustion air. The sorbent is fed to facilitate capture of sulphur from the coal in the bed itself resulting in consequent low sulphur emission. Due to high gas velocities, the fuel ash and unburnt fuel are carried out of the combustor with the flue gases. These are then collected by a recycling cyclone separator and returned to the lower part of the gasifier. Figure 2.4 (b) shows a diagram of a circulating fluidized bed.

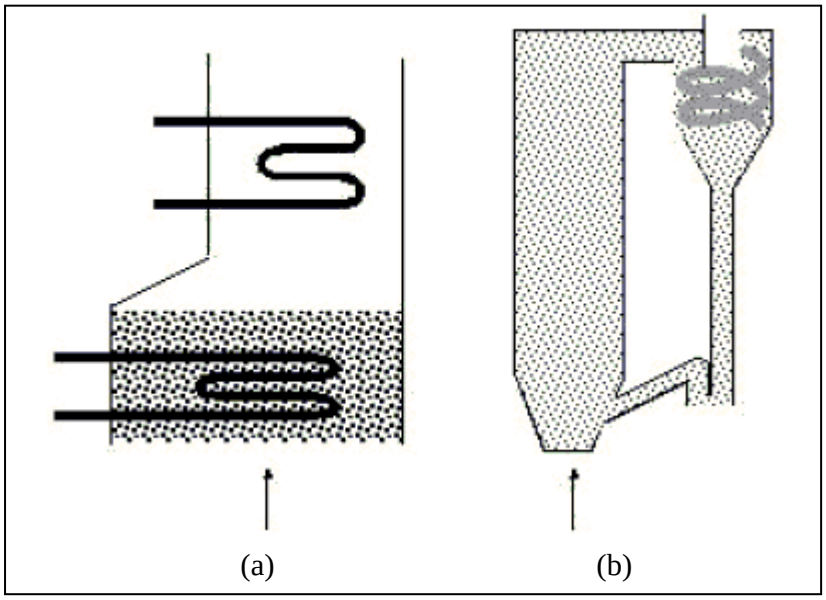


Figure 2.4: Diagram of a) bubbling bed. b) circulating bed

2.2.4 Wood Gasification

Wood is an example of biomass source. It is composed of cellulose, hemicellulose, and lignins (Lee 1996). It is defined as lignocellulosic material. The structures of hemicellulose and lignins are very complicated, which contribute to the complexity of the thermochemical conversion reactions. Consider wood with a chemistry formula $CH_{1.44}\ O_{0.66.}$ and the ideal gasification reaction with oxygen can be written as follows:

$$\mathbf{CH_{1.44}\ O_{0.66} + 0.17O_2 \rightarrow CO + 0.72H_2} \qquad (2.1)$$

This ideal gasification reaction is an endothermic process, which means the reaction in the system absorbs energy from the surroundings in the form of heat. Oxygen is partially supplied to gasify the wood and produce carbon monoxide and hydrogen. According to Reed (1988), the surplus oxygen reacts with about a third of the CO and H_2 ideally produced. The Equation 2.2 is the theoretical global gasification reaction with CO_2 and H_2O as additional products.

$$\mathbf{CH_{1.44}\,O_{0.66} + 0.45O_2 \rightarrow 0.7CO + 0.3CO_2 + 0.55H_2 + 0.2H_2O} \qquad (2.2)$$

Biomass materials, wood will undergo three stages of mass loss in the gasification process. The stages are drying, pyrolysis and gasification. Inherent moisture in the biomass is removed in the drying stage. The temperature rises and the biomass particles begin to decompose and release volatiles. When this happens it is called the pyrolysis stage. In the last stage, where there is insufficient oxygen supplied, partial oxidation takes place. Under this condition, partial oxidation of the residues and volatiles occurs. This results in the generation of partially oxidized products and combustible gases such as hydrogen, carbon monoxide and methane. These stages occur rapidly.

The amount of water depends on wood species and most species can absorb about 30% water (U.S. Departments of Agriculture, 2007). According to McKendry (2001), moisture content of wood more than 30% makes ignition

difficult and will reduce heating value of the producer gas due to the need to evaporate additional moisture before combustion occurs. So to improve heating value of the producer gas, moisture content of the wood must be kept below 30%.

2.2.5 Producer Gas Composition from a Downdraft Gasifier

The producer gas consists of combustible and incombustible gases. Theoretically, the composition of producer gas-air mixture is 15-20% hydrogen, 10-15% carbon monoxide, 9-15% carbon dioxide, 3-5% methane and 40-50% nitrogen by volume (Sen, 2005 and Reed, 1988).

Gunderson and Darren (2000) reported that hydrogen could be obtained from biomass and many other compounds such as water and fossil fuels. To obtain hydrogen from biomass, pyrolysis or gasification must be applied, which typically produces a gas containing 20% hydrogen by volume, which can be further steam-reformed to make higher quality gas.

Munoz *et al*. (2000) stated that producer gas contained 14% hydrogen, 22% carbon monoxide, 13% carbon dioxide, 3% methane and rest, nitrogen that had been determined from wood residues using a downdraft fixed bed gasifier. In their research, they used two fuels, producer gas and gasoline to measure the performance of a spark ignition engine. The results showed that there was so much loss of power in using producer gas compared with gasoline. The hydrocarbon and carbon monoxide were also reduced but carbon dioxide increased when using producer gas.

Sridhar *et al*. (2001) also stated that the producer gas can be fuelled into a spark ignition engine converted from a diesel engine. They used open top downdraft gasifier system to convert biomass (causurina species wood) into producer gas. The composition of the producer gas was 19% hydrogen, 19% carbon monoxide, 12% carbon dioxide, 2% methane, 2% water and rest, nitrogen. They found that the performance of the engine at higher compression engine had been smooth and the cylinder pressure-crank angle trace had shown smooth pressure variations during the entire combustion process without any sign of abnormal pressure raise.

Zainal *et al*. (2002) reported that the gas composition found in their experiment on a downdraft biomass gasifier using furniture wood and wood chips was 14.05% hydrogen, 24.04% carbon monoxide, 14.66% carbon dioxide, 2.02% methane, 1.69% oxygen and 43.62% nitrogen.

Dogru *et al*. (2002) also investigated gasification potential of hazelnut shells using a downdraft gasifier. They obtained a gas composition of 11.11%-14.77% hydrogen, 8.56%-18.56% carbon monoxide, 9.52%-16.33% carbon dioxide, 1.4%-2.47% methane and 53.33-59.67% nitrogen.

Adnan *et al*. (2002) studied hydrogen production from sewage sludge by applying downdraft gasification technique. They conducted the experiment using a pilot scale throated downdraft gasifier and concluded that the combustible gases from sewage sludge had a high percentage of hydrogen

(8.98% to 11.4%) within 20.09% – 23.83% of combustible gases. This amount of gases is enough to run internal combustion engine.

Uma *et al*. (2004) reported that biomass gasification was one such process where producer gas could be obtained and used for power generation purposes. Biomass gasifier-based systems capable of producing power from a few kilowatts up to several hundred kilowatts had been successfully developed. They investigated emission characteristics of a diesel engine using diesel alone and producer gas which were used to run the diesel engine at different load conditions. The producer gas was produced by a downdraft gasifier system. The gas composition containing 14% hydrogen, 19% carbon monoxide, 10% carbon dioxide, 1.9% methane, and the remaining is nitrogen. Carbon monoxide emissions from the producer gas were higher than that from diesel alone at all operated load conditions, but nitrogen oxide and sulphur dioxide emissions decreased.

Based on studies reviewed, the maximum combustible gas composition obtained from producer gas produced through biomass gasification stated by the researchers has only below 24% by volume. Table 2.1 shows a summary of the producer gas composition obtained by other researchers in their experiments using downdraft gasifier.

Table 2.1: Gas composition in the producer gas reported by other researchers using downdraft gasifier.

	Biomass	% Vol.				
	Material	H_2	CO	$C0_2$	CH_4	N_2
Munoz et al. (2000)	wood residues	14	22	13	3	48
Sridhar et al. (2001)	causurina wood	19	19	12	2	48
Zainal et al. (2002)	furniture wood and wood chips	15	24	15	2	44
Dogru et al.(2002)	hazelnut shells	15	19	16	2	48
Uma et al. (2004)	saw dust of wood	14	19	10	2	55

To increase the percentage of combustible composition in the producer gas, some methods have to be studied. The hard work of other researchers to increase combustible composition in the producer gas is elaborated in Section 2.3.

2.2.6 Producer Gas Energy Content

Producer gas which is has the energy content greater than 4 MJ/*N*m^3 can be applied to run a diesel engine (Kumar, 2010). The energy content of producer gas can be calculated from the energy content of the components using low and high heating values for each gas as shown in Equation 2.3.

$$HV_{pg} = \sum (\%vol.)_g (HV)_g \tag{2.3}$$