PRODUCTION OF HYDROGEN BASED ON COMBINED TORREFACTION AND GASIFICATION PROCESS

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Dedicated to my parents, family, teachers and my friends.

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ABSTRACT

Fossil fuels are commonly used and highly dependent in worldwide market as the main energy sources despite of any other sources but this source is non-renewable energy. Hence, biomass is becoming the best effective alternative energy source to replace the fossil fuels in the future due to its abundances and availabilities. Different types of biomass; which are torrefied and non-torrefied biomass usually have a different impact on the production of high level of hydrogen gas which is useful in generating energy. Therefore, the objective of this study is to compare the performance of torrefied and non-torrefied biomass using gasification process. Both types of biomass considered in this study include *'meranti'* wood, *'cengal'* wood and *'kulim'* wood. In this work, fluidized bed is chosen as gasifier to produce the synthesis gas. The fluidized bed gasification model is developed in Aspen Plus as a simulator by considering the hydrodynamic and reaction rate kinetics simultaneously for both biomasses. For torrefied biomass, however, the process needs to undergo pretreatment process called as torrefaction in order to increase the energy level of the biomass. Based on this pretreatment, the torrefied biomass is then used as an input to this gasifier. It is expected that the torrified biomass will gives higher performance due to the higher level production of hydrogen gas compared to the non-torrefied biomass because torrified biomass undergoes torrefaction process before proceed to gasification process will have a lower moisture content and higher heating value.

ABSTRAK

Bahan api fosil sangat biasa digunakan dalam kehidupan seharian dan harganya sangat bergantung pada pasaran di seluruh dunia sebagai sumber tenaga utama jika dibandingkan dengan sumber tenaga lain tetapi walaubagaimanapun sumber ini merupakan sumber tenaga yang tidak boleh diperbaharui. Oleh itu, biomas menjadi sumber tenaga alternatif yang berkesan dan terbaik untuk menggantikan bahan api fosil pada masa akan datang biomass mudah didapati jika dibandingkan dengan bahan api fosil itu sendiri. Dua jenis biomas; yang *torrefied* dan yang bukan *torrefied* biasanya mempunyai kesan yang berbeza pada penghasilan gas hidrogen yang berguna dalam menjana tenaga. Oleh itu, objektif kajian ini adalah untuk membandingkan prestasi biomas *'torrefied'* dan bukan '*torrefied*' menggunakan proses pengegasan. Kedua-dua jenis biojisim ini dipertimbangkan dalam kajian ini termasuk, kayu cengal, kayu kulim dan kayu meranti. Dalam kajian ini, *'fluidized bed gasifier'* dipilih sebagai *gasifier* untuk menghasilkan gas sintesis. Model *'fluidized bed gasifier'* akan digunakan sebagai input di dalam Aspen Plus, iaitu simulator dengan mengambil kira kinetik hidrodinamik dan kadar tindak balas untuk di uji ke tas kedua-dua jenis biomas. Untuk biomass *'torrefied'*, bagaimanapun, proses ini perlu menjalani proses rawatan awal atau perlu menjalani eksperiment dipanggil sebagai *torrefaction* untuk meningkatkan tahap tenaga biomas itu sendiri. Berdasarkan eksperimen, biomas *torrefied* kemudiannya digunakan sebagai input kepada *gasifier* ini di dalam Aspen Plus. Hasil daripada proses penggegasan di dalam Aspen Plus, ia dijangka bahawa biomass jenis *torrified* memberikan prestasi yang lebih tinggi disebabkan oleh pengeluaran gas hydrogen yang jauh lebih tinggi daripada biomass jenis bukan *torrefied* kerana biomass *torrified* yang telah menjalani proses *torrefaction* sebelum meneruskan proses pengegasan akan mempunyai kandungan kelembapan yang lebih rendah dan nilai pemanasan yang lebih tinggi.

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LIST OF SYMBOLS

min	minute
ºC	degree Celcius
K	Kelvin
g	gram

LIST OF ABBREVIATIONS

CH_4	Methane
CHNS	Carbon-Hydrogen-Nitrogen-Sulphur
CO	Carbon monoxide
CO_2	Carbon dioxide
FC	Fixed carbon
GCC	Gult Cooperation Council
H_2	Hydrogen gas
HHV	High Heating Value
MC	Moisture content
OPEC	Organization of the Petroleum Exporting Countries
SO_2	Sulphur dioxide
TGA	Thermogravimetric Analyzer
VM	Volatile matter

CHAPTER 1

INTRODUCTION

1.1   Background of the Study

Currently, the main energy source of fossil fuels and coals are widely used for many applications in daily life such as electricity production, cosmetics, medicines and fuels for transportation. However, this source of energy is non-renewable and the level itself almost decreasing in alarming rate. Therefore, the most precise replacement for fossil fuels is biomass due to its abundances and availabilities in world. Biomass has been used as an energy source since man starts burning wood to keep warm or cook and nowadays biomass is still the world's largest source of renewable energy (Ratte *et. al*, 2011). In many ways, biomass is a new source of power. While wood has always served as a fuel source for fires and ovens and conventional heating methods, biomass energy advancements are a few steps beyond that. Now these biomass fuel products are harvested and mass-produced and used in everything from engines to power plants.

The search for new renewable energy sources is ongoing as the world's supplies of fossil fuels are used up. The energy source that shows the most promise is the use of biomass and biofuels at the present time. As technology improves, biomass is becoming a more attractive alternative to fossil fuels because it produces fewer emissions, contributes to local economies, mitigates global climate change, and can increase national security.

As a renewable energy, biomass is generally considered CO_2-neutral. This is particularly the case with regard to agricultural residues, which are periodically planted and harvested. During the growth, these plants have removed CO_2 from the atmosphere for photosynthesis, which is released again during combustion. Although the direct emissions of SO_2 and NO_x at generation stage are smaller than from fossil fuels because of the relatively low nitrogen and sulfur content of biomass, its environmental impact

cannot be ignored from the perspective of life cycle assessment. The main reason is that cultivation, harvesting, transportation, and pre-treatment of biomass are energy-consuming processes that are accompanied by significant emissions.

At present, there are three mature technologies of biomass power generation: direct-combustion, gasification, and co-firing. The environmental capacity and energy consumption at all stages of generation process could be understood comprehensively using life cycle assessment methodology, thereby considering adopting measures to conserve resources and protect the environment.

1.2   Motivation

Since fossil fuel is non-renewable energy, the biomass can be replaced as the best alternative way in generating energy due to its abundances and availabilities. Biomass come from living sources which life is always cyclical, so these products are potentially never run out as long as there is something living on earth and there are humans to turn that living things components and waste products into energy. In the United Kingdom, biomass fuels are made from recycled chicken droppings while in the United States and Russia, there are plentiful forests for lumber to be used in the production of biomass energy (Sommer *et. al*, n.d.).

Fossil fuels are responsible for producing the most carbon emissions compared to the biomass. Biomass does release carbon dioxide but captures carbon dioxide for its own growth during photosynthesis and the general view has been that carbon emitted into the atmosphere from biological materials is carbon neutral which part of a closed loop whereby plant regrowth simply recaptures the carbon emissions associated with the energy produced (Sedjo, 2013). Instead of that, biomass can produce clean energy so biomass energy is expected to play a major role in the substitution of renewable energy sources for fossil fuels over the next several decades.

Other than that, the cost of producing biomass for use as fuels and energy sources is very cheap compared to the cost of finding and extracting fossil fuels. The cost of finding is eliminated when one deliberately plants certain types of plants to be used in the production of biomass fuels. Extraction is generally no more expensive than harvesting crops for food. In some cases, growing and harvesting crops for biomass fuel use is cheaper than raising food crops. Even though the cost of rendering the biomass into usable fuels make it unfeasible on a small scale and more energy is needed to produce the fuel than is gained by using the fuel but, these numbers quickly reverse and biomass become a highly feasible and trusted renewable source of energy on a larger scale.

1.3   Problem Statement

The fossil fuel, which is non renewable energy are widely used as a main source of energy. An alternative way is needed to find the most suitable source to replace fossil fuels as future energy. Due to this situation, biomass is the best renewable, storable and transportable as the replacement for new energy source (Miao, 2012).

Carbon dioxide released by fossil fuels, is released into the atmosphere and is harmful to the environment. The emission of carbon emissions from this source of energy is needed to be control due to the side effect of greenhouse gases that tends the global warming. Furthermore, many of the environmental problems the world faces nowadays, including climate change, air pollution, oil spills, and acid rain which result from our dependence on fossil fuels. The burning of fossil fuels produces heat-trapping gases that are the main cause of the ongoing rise in global atmospheric temperatures. Despite a growing list of global warming indicators, underscored by the alarmingly rapid recession of Arctic sea ice, opportunistic oil companies continue to exploit the ever-increasing human need for energy consumption and are constantly on the lookout for untapped oil and gas sources. In addition to ecological disturbances from fossil fuel extraction, there are certain cultural consequences for communities around the Pacific Rim. These communities, many Indigenous, are threatened by the depletion of specific resources they depend upon for their livelihoods and culture (Pimental et. al, 2002).

The rising prices of fossil fuels indicate the unstable condition in its industry. Many producer countries are dependent on them for constant supply these fuels. Organization of the Petroleum Exporting Countries (OPEC) monitors the volume of oil consumed and then adjusts its own production to maintain its desired barrel price, results in worldwide price fluctuations (Morriss, n.d). For example, Low oil prices, pushed down further by OPEC's meeting, have impacted world economies, energy stocks, and several currencies. From the fate of the Russian rouble to Venezuelan deficits to American mutual funds full of Exxon or Chevron stock, OPEC's decision was the shot heard round the world for troubled commodities (Carlson, 2014). The economic crisis in South and East Asia in the late 1990s reduced demand for Gulf Cooperation Council (GCC) oil and together with this, the deflationary effects of the crisis on the world economy, put downward pressure on prices and reduced the amount of finance available in oil producing countries for further development of that sector. Since then, as the recovery has taken hold, prices have increased sharply (Paul Rivli, 2000).

1.4   Objectives

The main objective in this research is to compare the performance of torrefied and raw biomass using gasification process in fluidized bed gasifier.

1.5   Scopes of Study

The following are the scopes of this research:

1) Simulates using Aspen Plus for non-torrefied and torrefied biomass of *meranti* wood, *cengal* wood and *kulim* wood.
2) Analyzes the properties of torrefied of *meranti* wood, *cengal* wood and *kulim* wood from torrefaction experiment.
3) Compares the performance of different types of biomass (torrefied and non-torrefied).
4) Improves the performance based on sensitivity analysis.

CHAPTER 2

LITERATURE REVIEW

2.1   Definition of biomass and its carbon cycle

The word biomass consist of "bio" and "mass", originally used in the field of ecology simply referring to amount of animal and plant. But the meaning of the word was widened beyond ecological field and came to include the meaning "biological resource as energy sources" since it was vigorously proposed that alternative energy sources should be promoted (Yokoyama, 2008). From the perspective of energy resources, biomass had been considered as merely a kind of renewable resources but nowadays the world sees it as an independent category of new energy.

Biomass is carbon based and is composed of a mixture of organic molecules containing hydrogen, usually including atoms of oxygen, often nitrogen and also small quantities of other atoms, including alkali, alkaline earth and heavy metals. These metals are often found in functional molecules such as the porphyrins which include chlorophyll which contains magnesium. The carbon used to construct biomass is absorbed from the atmosphere as carbon dioxide (CO_2) by plant life, using energy from the sun. Plants may subsequently be eaten by animals and thus converted into animal biomass. However the primary absorption is performed by plants. If plant material is not eaten it is generally either broken down by micro-organisms or burned. Once it broken down it releases the carbon back to the atmosphere, mainly as either carbon dioxide (CO_2) or methane (CH_4), depending upon the conditions and processes involved. As the biomass burned, the carbon is returned to the atmosphere as CO_2.
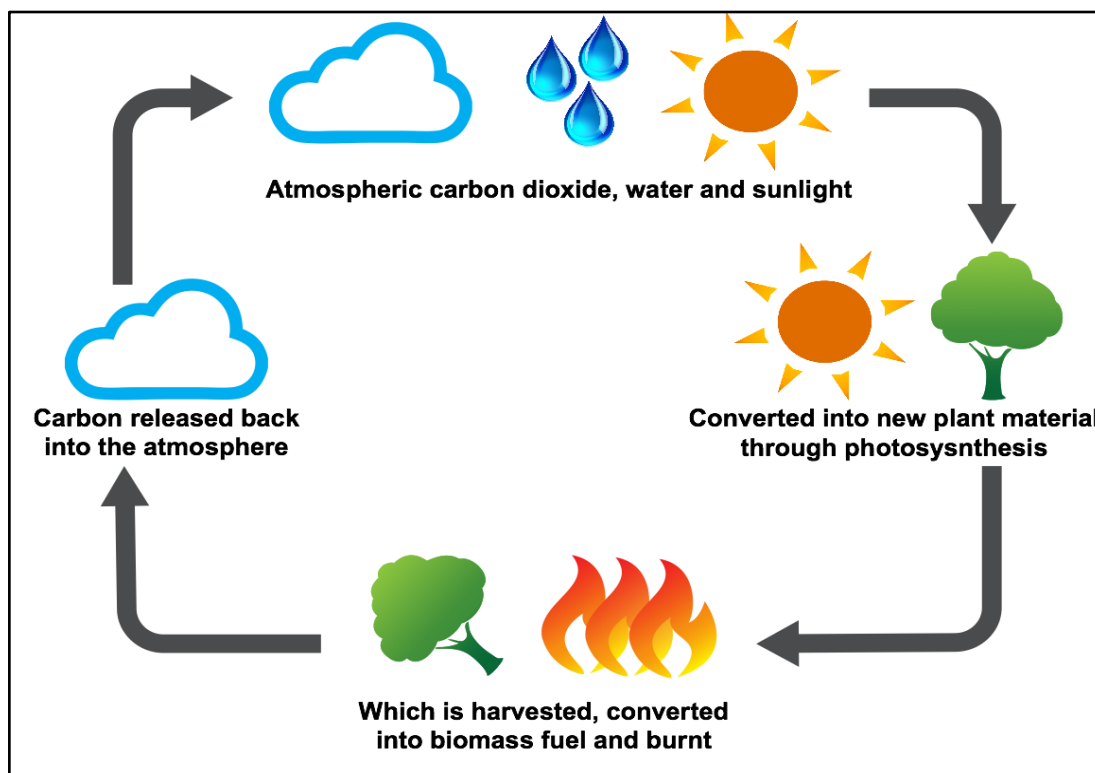
Figure 2-1: The Carbon cycle of biomass

From Figure 2-1, the carbon cycle begins as atmospheric carbon is absorbed by growing trees. In the presence of sunlight and water, this is then converted into the complex carbohydrates required for tree growth by the process of photosynthesis. When trees die, the process of decomposition begins. During this process of decay, heat energy, water, and carbon dioxide are released back into the atmosphere. The carbon released during decomposition is reabsorbed by the growth of new trees, resulting in zero net atmospheric carbon gain. The use of wood to produce biomass fuel works within this cycle. Dead trees can clutter forest floors for years due to the relatively slow rate of decomposition. But by harvesting these biomass resources, the heat energy they contain can be captured and used. The natural process of decay is accelerated when woody biomass is burned, and the heat energy given off can be used to heat your property. When biomass wood pellets are burnt, the carbon is returned to the atmosphere as part of the natural carbon cycle. New trees absorb the reintroduced carbon, and the cycle repeats.

2.2 Type of gasifier

In order to increase the hydrogen content of the producer gas, steam is used along with air in a gasifier (Sharma and Sheth, 2016). There are three types of gasifiers involved in gasification process which are fixed (moving) bed gasifier – divided into two which are updraft and downdraft, entrained-flow gasifier and fluidized-bed gasifier.
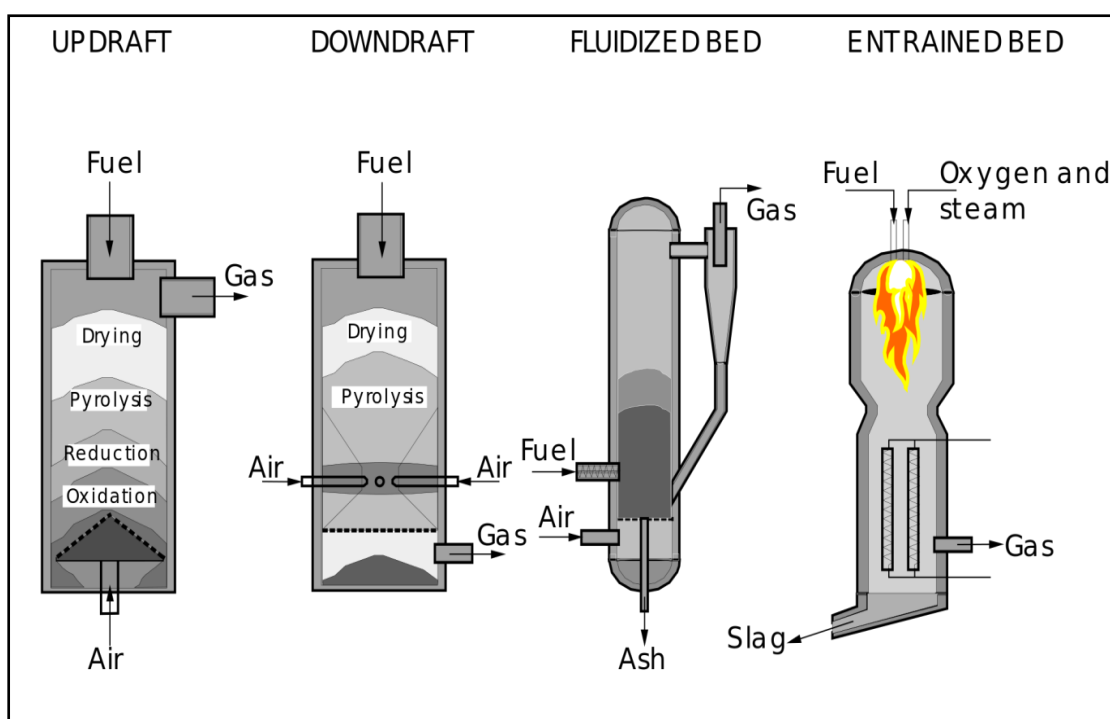

Figure 2-2: The structure of different types of gasifiers

Moving bed gasifiers known as countercurrent flow reactors in which the coal enters at the top of the reactor and air or oxygen enters at the bottom. As the coal slowly moves down through the reactor, it is gasified and the remaining ash drops out of the bottom of the reactor. Due to the countercurrent flow arrangement, the heat of reaction from the gasification reactions serves to pre-heat the coal before it enters the gasification reaction zone. Consequently, the temperature of the syngas exiting the gasifier is significantly lower than the temperature needed for complete conversion of the coal. The residence time of the coal within a moving bed gasifier may be on the order of hours. The properties of moving bed gasifier are low oxidant requirements, relatively high methane content in the produced gas, best production of hydrocarbon

liquids such as tars an oils, high "cold gas" thermal efficiency when the heating value of the hydrocarbon liquids are included, limited ability to handle fines and need to consider special requirements for handling caking coal.

Finely-ground coal is injected in co-current flow with the oxidant for entrained flow gasifier. The coal rapidly heats up and reacts with the oxidant. The residence time of an entrained flow gasifier is on the order of seconds or tens of seconds. Because of the short residence time, entrained flow gasifiers must operate at high temperatures to achieve high carbon conversion. Consequently, most entrained flow gasifiers use oxygen rather than air and operate above the slagging temperature of the coal. The entrained-flow gasifier has characteristics of high-temperature slagging operation, entrainment of some molten slag in the raw syngas, relatively large oxidant requirements, large amount of sensible heat in the raw syngas and ability to gasify all coal regardless of rank, caking characteristics or amount of fines.

A fluidized bed gasifier is a back-mixed or well-stirred reactor in which there is a consistent mixture of new coal particles mixed in with older, partially gasified and fully gasified particles. The mixing also fosters uniform temperatures throughout the bed. The flow of gas into the reactor (oxidant, steam, recycled syngas) must be sufficient to float the coal particles within the bed but not so high as to entrained them out of the bed. However, as the particles are gasified, they will become smaller and lighter and will be entrained out of the reactor. It is also important that the temperatures within the bed are less than the initial ash fusion temperature of the coal to avoid particle agglomeration. Typically a cyclone downstream of the gasifier will capture the larger particles that are entrained out and these particles are recycled back to the bed. Overall, the residence time of coal particles in a fluidized bed gasifier is shorter than that of a moving bed gasifier. The characteristics of fluidized-bed gasifier are extensive solids recycling, uniform and moderate temperature and moderate oxygen and steam requirements (Boyce, 2012).
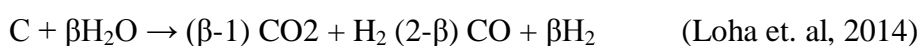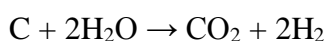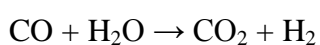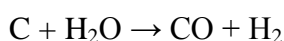
Table 2-1: Considerations about the current main types of gasifiers (Ruiz et.al, 2013)

GASIFIER	Downdraft	Updraft	Bubbling fluidized bed	Circulating fluidized bed	Entrained flow bed	Twin fluidized bed
Technology	Simple and proven. A simple reactor with relatively low investment cost		Plants with higher investment costs. Proven technology with coal		Complex construction	Complex construction
Fuel specifications	<51 mm	<51 mm	<6 mm	<6 mm	<0,15 mm	<6 mm
Maximum fuel moisture (%)	25	60	<55%	<55%	<15%	11–25
Gas LHV (MJ/Nm3)	4.5–5.0	5–6	3.7–8.4	4.5–13	4–6	5.6–6.3
Tar (g/Nm3)	0.015–3.0	30–150	3.7–61.9	4–20	0.01–4	0.2–2
Ash and particles in syngas	Low	High	High	High	Low	High
Reaction temperature	1090 °C		800–1000 °C		1990 °C	800–1000 °C
Ash melting point	>1250 °C	>1000 °C	>1000 °C		>1250 °C	>1000 °C
Syngas output temperature	700 °C	200–400 °C	800–1000 °C		>1260 °C	800–1000 °C
Admissible powers	Up to 1 MWe	Up to 10 MWe	1–20 MWe	2–100 MWe	5–100 MWe	2–50 MWe
Residence time	Particles are in bed until its discharge		Particles spend substantial time in bed.	Particles pass repeatedly through the circulation loop (few seconds)	Very short (few seconds)	Particles spend substantial time in bed.
Carbon conversion efficiency	High	High	High. Loss of carbon in ash.	High	High	High
Process flexibility	Very limited. Any change in process variables needs a new design		Flexible to loads less than design		Very limited. Size and energy content of the fuel	Flexible to loads less than design

GASIFIER	Downdraft	Updraft	Bubbling fluidized bed	Circulating fluidized bed	Entrained flow bed	Twin fluidized bed
					must be in a narrow range.	
Temperature profile	High gradients		Vertically almost constant. Little radial variation	Vertically almost constant	Temperatures above the ash melting temperature	Constants in each reactor
Hot gas efficiency	85–90%	90–95%	89%	89%	80%	90–95%

2.3   Gasification Process

Gasification is one of the most promising technology for utilizing renewable resources to produce fossil fuel alternatives (Đurišić-Mladenović et.al, 2016). Gasification is a process that converts organic or fossil fuels based carbonaceous materials into carbon monoxide, hydrogen and carbon dioxide. This is achieved by reacting the material at high temperatures (>700 °C), without combustion, with a controlled amount of oxygen and or steam. The resulting gas mixture is called syngas (from synthesis gas or synthetic gas) or producer gas and is itself a fuel. The power derived from gasification and combustion of the resultant gas is considered to be a source of renewable energy if the gasified compounds were obtained from biomass (Srivasta, 2008). The gasified compound exit through gasification process include the hydrogen gas (H_2), methane gas (CH_4), carbon monoxide gas (CO) and carbon dioxide (CO_2) (Guo et. al, 2014). The product specification for gasification process is the amount of syngas produced particularly hydrogen gas (Balaji et. al, 2014). The steam gasification reaction occurred are:

$$C + H_2O \rightarrow CO + H_2$$

$$CO + H_2O \rightarrow CO_2 + H_2$$

$$C + 2H_2O \rightarrow CO_2 + 2H_2$$

$$C + \beta H_2O \rightarrow (\beta\text{-}1)\, CO2 + H_2\, (2\text{-}\beta)\, CO + \beta H_2 \qquad \text{(Loha et. al, 2014)}$$

2.4   Torrefaction Process

Torrefaction process is a mild pyrolysis process at temperature between 200ºC to 350ºC. The raw biomass has high water content which varies according to its nature; wood for example, contains 45%-60% of water, is heated to evaporate the moisture at the initial stage (Arnsfeld et. al, 2014 and Ratte et. al, 2011) . It loses physically bonded water during the second stage and is heated up to 200ºC slowly. Then the main torrefaction process starts with low heating rates around 10K/min and the release of volatiles until the torrefaction temperature is reached. After torrefaction time, the product is quenched in order to conserve the solid yield. During the torrefaction time, the molecular bonded with water is released. In contradiction to charcoal, the torrefacting biomass does not undergo a molecular reorganization of its structure. Its ignition temperatures in air atmosphere are lower than those of charcoal due to the faster reaction kinetics of the original molecular structure. The specific heating values of the solid products are higher than of raw biomass due to the loss of water during torrefaction process (Arnsfeld et. al, 2014).
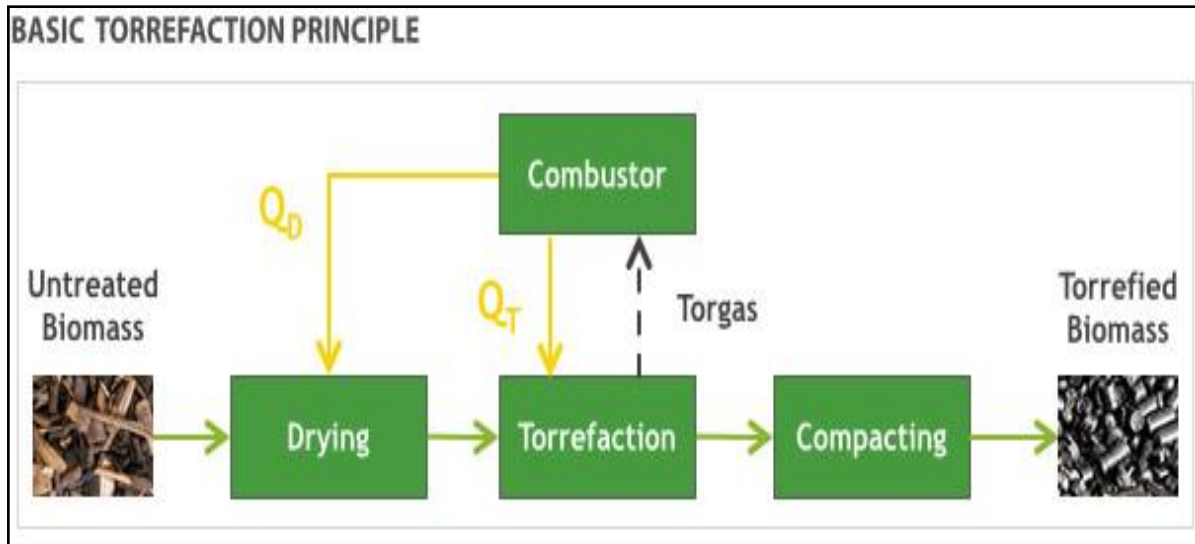

Figure 2-3: Basic principle of terrefaction process.

Other names for the torrefaction process are roasting, slow- and mild pyrolysis, wood cooking and high temperature drying. In recent history torrefaction has only been applied to various types of woody biomass, but already around 1930 the torrefaction process was studied in France. The amount of publications on torrefaction is relatively small but increasing in the last few years. Literature about torrefaction of diverse

biomass resources can be found, namely: maritime pine, chestnut, oak and eucalyptus, Caribbean pine, birch, pine, bagasse, bamboo, wood briquette, willow and beech, pedunculate oak, lauan wood, and oil palm wastes. Just below 200 ℃ thermal methods are used for wood preservation, while torrefaction is for energy purposes (Stelt *et al*, 2011).

Torrefaction is used as a pre-treatment step for biomass conversion techniques such as gasification and co-firing. The thermal treatment not only destructs the fibrous structure and tenacity of biomass, but is also known to increase the calorific value. Also after torrefaction the biomass has more hydrophobic characteristics that make storage of torrefied biomass more attractive above non-torrefied biomass, because of the rotting behavior. During the process of torrefaction the biomass partly devolatilizes leading to a decrease in mass, but the initial energy content of the torrefied biomass is mainly preserved in the solid product so the energy density of the biomass becomes higher than the original biomass which makes it more attractive for example is transportation.

2.5   Ultimate and proximate analysis

The ultimate and proximate analysis is commonly used in order to determine the coal's energy value. Ultimate and proximate analysis provides valuable information about chemical composition. Proximate analysis parameters include sulfur, moisture, volatile matter, ash, and fixed carbon. Ultimate analysis, which is more comprehensive, is dependent on quantitative analysis of various elements present in the coal sample, such as carbon, hydrogen, sulfur, oxygen, and nitrogen. The experimental determination of ultimate analysis data requires special instrumentation, while proximate analysis data can be obtained easily by using common equipments (Shen *et al*, 2010).

The ultimate analysis gives the composition of the biomass in wt% of carbon, hydrogen and oxygen (the major components) as well as sulphur and nitrogen (if any). The carbon determination includes that present in the organic coal substance and any originally present as mineral carbonate. The hydrogen determination includes that in the organic materials in coal and in all water associated with the coal. All nitrogen determined is assumed to part of the organic materials in coal.

Meanwhile, the proximate analysis gives moisture content, volatile content, consisting of gases and vapor driven off during pyrolysis (when heated to 950ºC), the fixed carbon and the ash, the inorganic residue remaining after combustion in the sample and high heating value (HHV) based on the complete combustion of the sample carbon dioxide and liquid water. Proximate analysis is the most often used analysis for characterizing coals in connection with their utilization. Table 2-2 shows one of the example of proximate and ultimate analysis data from biomass and coal.

Table 2-2: Physical and chemical properties of the thermally treated biomass and coal (Park et. al, 2012).

Sample	Proximate analysis (wt.%,adb)				Ultimate analysis (wt.%,db)					GCV (kcal kg^{-1},db)
	M	FC	VM	Ash	C	H	O	N	S	
Coal	6.67	54.50	27.25	11.58	74.12	4.22	6.93	1.91	0.41	6385
WC raw	3.81	7.42	88.72	0.05	46.73	6.46	46.35	0.41	ND	4895
TW250	3.19	15.51	81.19	0.11	52.22	5.18	41.94	0.55	ND	5314
TW275	2.90	23.46	73.47	0.17	57.33	4.95	37.17	0.37	ND	5387
TW300	2.81	29.36	67.58	0.25	59.03	4.78	35.59	0.34	ND	5387
CW325	3.29	51.57	44.71	0.43	68.43	4.31	26.47	0.35	ND	6956
CW350	3.40	52.56	43.57	0.47	69.20	4.28	25.71	0.32	ND	6998
CW375	4.03	61.36	31.13	0.48	71.39	3.80	24.00	0.31	ND	7140

adb, Air-dried basis; db, dry basis; M, Moisture; O$^{differ.}$, by difference [(FC + VM)db − (C + H + N + S)db]; ND, not detected; S_{BET}, specific surface area determined on N_2 isotherms by the BET method.

2.6   Aspen Plus

Aspen Plus is software of process modelling tool for steady-state simulation, design, performance monitoring, optimization and business planning for chemicals, specialty chemicals, petrochemicals and metallurgy industries (Schefflan, 2011). Figure 2-4 shows the basic gasification modeling in Aspen Plus software. This software performs solids modeling within the same platform, which offers the ability to rigorously model chemical processes that involve both fluids and solids. Aspen Plus V8

allows the ability to describe granular solids in detail and provides a comprehensive model library including for drying, granulation, crystallization, crushing, fluid-solid separation, classification, and more.

Other than that, Aspen Plus address the needs of both process engineering and particle science with built-in solids modeling software, supports rigorous descriptions of solids processing steps. It incorporates a comprehensive library of solids unit operations (such as dryers, granulators, crystallizers, fluidized beds, crushers, gas/solid and liquid/solid separators, classifiers, and conveying systems) and can allow for a detailed description of granular solids. With conceptual solids models, modeling solids processing equipment is more accessible to everyone.

The fluidized bed model allows users to consider chemical reactions and their impact on fluid mechanics. Get a better understanding of re-circulation rates and particle size distributions throughout the production process with the fluidized bed model. Minimize loss of fines, recycle rates, and improve yields and selectivity. It simply said that Aspen Plus can achieve higher throughputs.
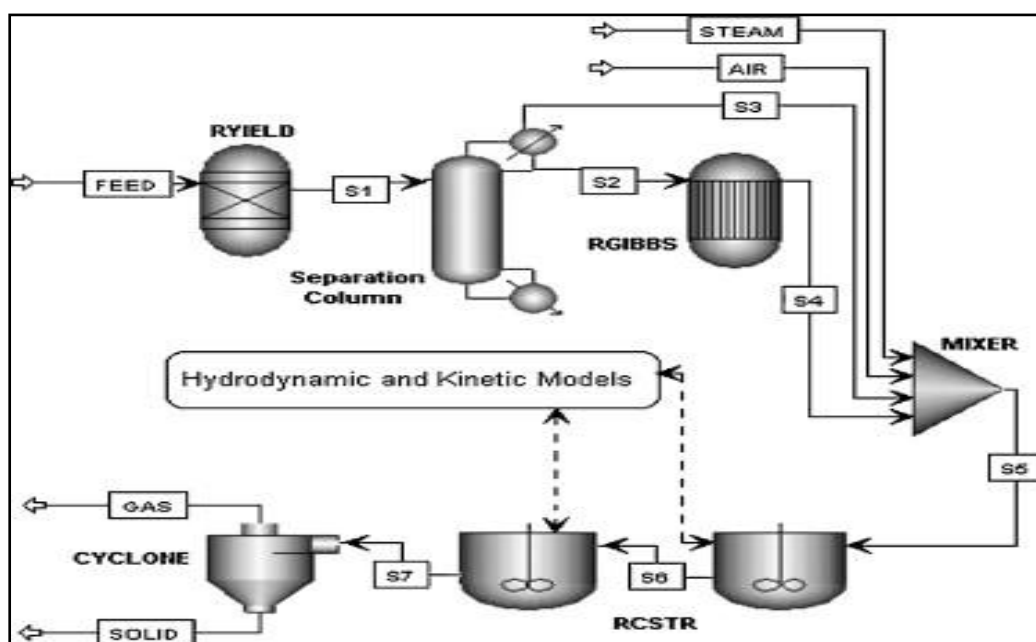

Figure 2-4: Simulation diagram in Aspen Plus for fluidized bed gasification (Puig-Arnavat et. al, 2010)

CHAPTER 3

METHODOLOGY

3.1 Experimental

3.1.1 Material and method

Three different types of wood which are common in Malaysia were chosen; *cengal* wood, *meranti* wood and *kulim* wood. The wood samples were, without bark and already chipped, sieved into particle size of 500 μm to 1mm. Before the real torrefaction takes place, the sieved biomass is heated to evaporate the moisture content at the initial stage which is drying process. At pre-drying process, the free water is evaporated from the biomass at constant temperature at 100 ℃. Meanwhile, the temperature of biomass is increased to 200 ℃ at post-dying and intermediate heating. During this stage, physically bound water is released while the resistance against mass and heat transfer within the biomass particles. Some mass loss can occur during this process as light fractions can evaporate. After the drying, the torrefaction takes place with low heating rate at around 10 K/min along with nitrogen gas flow. The weight of wood biomass used for torrefaction experiment is approximately in the range of 1.5g to 3.0g. The torrefaction will start when the temperature when the temperature will reach 200 ℃ and end when the process is again cooled dowm from the specific temperature to 200 ℃. The temperature torrefaction is defined as the maximum constant temperature. Most of the mass loss of the biomass occurs during torrefaction process. The biomass is torrified at three different temperatures to get samples from all temperature levels of the torrefaction process; 240 ℃, 270 ℃ and 300 ℃. After 30 minutes of torrefaction, the biomass is cooled below 200 ℃ to room temperature and will undergo further analysis in TGA (thermogravimetric analyzer) and CHNS elemental analyzer. The process of torrefaction experiment is shown in Figure 3-1.
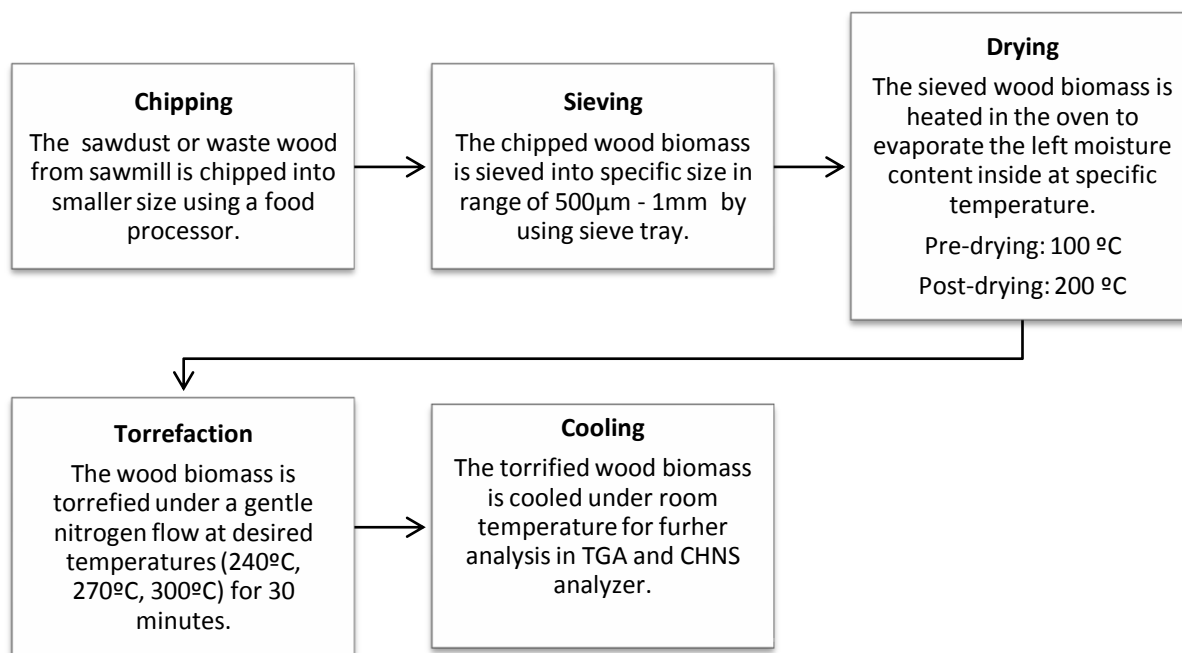
Figure 3-1: Process flow of torrefaction experiment.

3.1.2 Thermogravimetric Analyzer (TGA)

In this study, a thermogravimetric ignition technique was adopted for the ignition temperature measurement, as it is being widely used by many researchers. The thermogravimetric experiments were performed using a TGA instrument and the mass loss rate with temperature was recorded under nitrogen and air atmosphere. In both cases, the gas flow rate was maintained at 10 mL/min. and a typical sample mass of 1–5 mg was used with a heating rate of 10 °C/min. First, the samples were heated from room temperature to 105 °C and kept for 10 min. to remove the moisture contained in the samples, and then they were heated up again with the same heating rate to a temperature of 900 °C. The TGA experiments were performed at least twice to validate repeatability of the mass-temperature loss curves results, which was found to be quite satisfactory with (max. ±1%) standard deviation. From the result of proximate analysis in TGA, the moisture content (MC), volatile matter (VM), fixed carbon (FC), and ash content of a wood biomass samples can be obtained, which useful in determining of syngas production by key in its values in Aspen Plus software as the input. From the TGA result, the properties of wood biomass samples are presented in Table 4.1 and Table 4.2 in result and discussion section in chapter 4.

3.1.3 CHNS Analyzer

The ultimate analysis used elemental analyzer or CHNS analyzer to obtain the mass fraction weight of C, H, N, S element in the wood biomass samples. The amount of oxygen in ultimate analysis was calculated manually as the following Eq.:

$$\% [O] = 100\% - \%[Ash] - \%[C] - \%[H] - \%[N] - \%[S] \tag{1}$$

3.1.4 Higher Heating Value (HHV) of Biomass

HHV which is an important parameter in determining the energy yield of the solid product during torrefaction. For validation, the HHV of a few torrefied samples were also analyzed in a bomb calorimeter and compared with the calculated results of Eq. 2 and the deviation was found to be no more than 5%. The amount of HHV of was calculated as the following Eq:

$$HHV = 3.55C^2 - 232C - 2230H + 51.2C \times H + 131N + 20,600 \tag{2}$$

where C = carbon, H = hydrogen and N = nitrogen content and HHV is expressed in kJ/kg on a dry basis (wt%) (Gucho *et.al*, 2015).

3.1.5 Mass and Energy Yield

Energy yield per kg of dry originating biomass is defined as the amount of energy conserved in the solid part after the torrefaction process. It is an important parameter for the evaluation of the process and it is calculated from the mass yield of the solid torrefied product. The energy yield of the torrefied biomass is less compared to higher heating value of the original biomass, as some of the volatile matter which contributes to the energy content leaves the solid torrefied product. On industrial scale, the torrefaction gaseous by-product is combusted and the generated heat is reused in the process itself. In this way, the overall energy balance of the process can be enhanced. According to Bergman *et al*.(2005), the mass and energy yield of the torrefied biomass are defined by Equations (3) and (4), respectively.

$$Mass\ Yield\ (\%) = \frac{Mass\ of\ torrefied\ solid\ product\ (kg)}{Mass\ of\ originating\ biomass\ (kg)} \times 100\% \tag{3}$$

$$Energy\ Yield\ (\%) = \frac{Mass\ yield\ \times HHV\ torrefied\ solid\ product\ (\frac{kJ}{kg})}{HHV\ of\ originating\ biomass\ (\frac{kJ}{kg})} \times 100\% \tag{4}$$

3.2   Simulation by ASPEN PLUS software

Aspen Plus has been chosen as a simulator for the systematic framework of gasification process as shown in Figure 3-2. The gasification process is tested in simulation after the torrefaction experiment in the laboratory. The data analysis from the experiment is important because it will be transferred into the gasification process to determine the hydrogen production at the end of the stream.
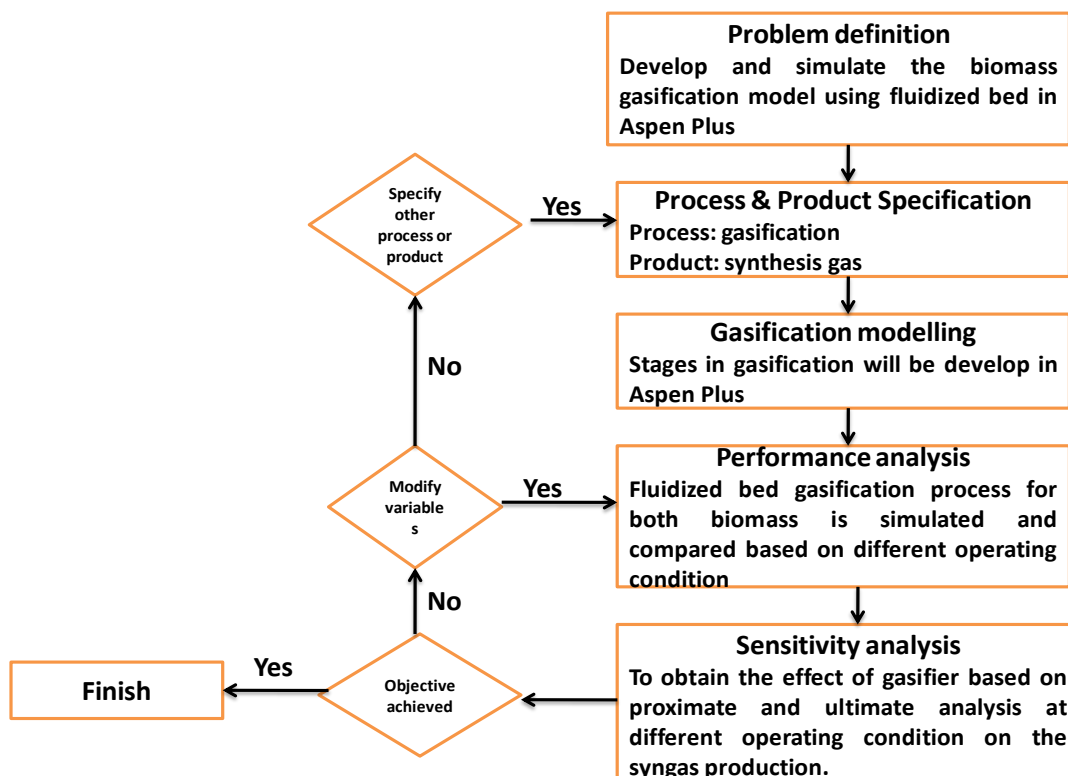

Figure 3-2: Systematic framework of gasification process.

3.2.1 Problem statement

The main objective is to develop and simulate the biomass gasification model using fluidized bed in Aspen Plus software. The gasification model is tested using torried and non-torrified biomass.

3.2.2 Process and product specification

The process used is gasification process in fluidized bed gasifier and the product desired is the amount of syngas produced particularly hydrogen gas. Few assumptions are considered for the gasification model (Nikoo and Mahinpey, 2008) which consists of:

- Steady state and isothermal process.
- All the gases are distributed uniformly and evenly during emulsion phase.
- The particles are in uniform size, spherical and the average diameter remains constant.
- Char only compose of carbon (organic solid) and ash (inorganic solid).
- Gasification of char started in bedand finishes in the freeboard.
- The gasification process is started from pyrolysis, followed by combustion and steam gasification.

3.2.3 Gasification modelling

Gasification process is classified into 4 stages are required for which consists of decomposition of feed, volatile reactions, char gasification and the separation of gas and solid that will be develop in Aspen Plus.

- Stage 1: Biomass will decompose and converted into its components once it enters the yield reactor.
- Stage 2: The separator will separate the products into volatile matter and solids. Separated volatile matter then is fed into the Gibbs reactor for combustion process.

- Stage 3: The first CSTR and the second CSTR are for combustion reaction and gasification reaction, respectively.
- Stage 4: The syngas product from both CSTR reactors is separated in the cyclone into gas and solid products under specific operating condition.

Table 3-1: Example of operating conditions for gasification process.

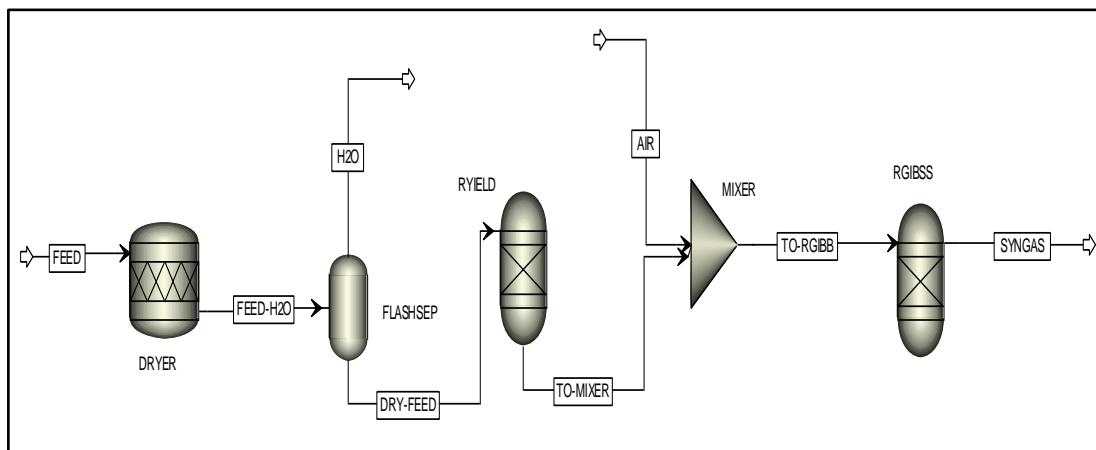| Fluidized bed gasifier	
Temperature (degree C)	700-900
Pressure (bar)	1.05
Air	
Temperature (	65
Flowrate (Nm^3/h)	0.5-0.7
Steam	
Temperature	145
Flowrate (kg/h)	0-1.8

Figure 3-3: Gasification modeling in fluidized bed reactor.

3.2.4 Performance analysis

The fluidized bed gasification process for both types of biomasses is simulated and compared based on different condition. In this study, the performance is tested at different temperatures, which are 800ºC, 900ºC and 1000ºC.

3.2.5   Sensitivity analysis

The effect of gasifier based on proximate and ultimate analysis will be determined at different operating condition on the syngas production. The biomass used in the sensitivity analysis is torried and non-torrified biomass of 'meranti wood', 'cengal' wood and pine wood. The parameter used for sensitivity analysis is temperature condition in fluidized bed gasifier.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Visual Observation

The physical appearance of raw biomass and after being torrified can be seen in Figure 4-1.

	Cengal wood	*Kulim* wood	*Meranti* wood
Raw T=27 ℃			
T = 240 ℃			
T = 270 ℃			

T = 300 ℃			

Figure 4-1: The raw biomass particles and their torrefied products at different torrefaction temperatures for 30 minutes.

Based on Figure 4-1, it is evident that, irrespective of the biomass type, the colour of the torrefied product changes from light brown to dark brown and to black as the temperature and time increases. The increased amount of volatiles release facilitated the removal of oxygenated and hydrogenated compounds leaving the solid torrefied product more concentrated in fixed carbon. In general, wood biomass with high fixed carbon tends to be more black in colour.

4.2   Proximate and ultimate analysis

Table 4-1 and Table 4-2 recorded the influence of the temperature on CHNOS elements, the volatile matter, moisture content and fixed carbon of the torrefied and non-torrefied wood biomass product.

Table 4-1: Proximate analysis of torrefied and non-torrefied wood biomass.

Type of biomass	Temperature level	MC (wt.%)	VM (wt.%)	FC (wt.%)	Ash (wt.%)	HHV (MJ/kg)
Cengal wood	27 ℃ (Raw)	4.2	77.8	17.8	0.92	19.19
Cengal wood	240 ℃	3.9	75.1	19.97	1.03	20.90
Cengal wood	270 ℃	3.2	68.7	26.9	1.21	21.84
Cengal wood	300 ℃	2.7	65.9	30.2	1.23	23.25
Kulim wood	27 ℃ (Raw)	6.9	75.1	16.98	1.02	18.93
Kulim wood	240 ℃	4.5	73.3	21.05	1.15	20.43
Kulim wood	270 ℃	4.3	70.6	23.8	1.29	21.37
Kulim wood	300 ℃	3.5	67.8	27.4	1.34	22.16

Meranti wood	27 ℃ (Raw)	8.7	74.9	15.29	1.09	19.07
Meranti wood	240 ℃	7.1	70.4	21.4	1.11	19.14
Meranti wood	270 ℃	5.6	68.5	24.72	1.18	19.99
Meranti wood	300 ℃	4.5	64.9	29.3	1.33	20.49

Table 4-2: Ultimate analysis of torrefied and non-torrefied wood biomass.

Type of biomass	Temperature level	C (wt.%)	H (wt.%)	N (wt.%)	S (wt.%)	O (wt%)
Cengal wood	27 ℃ (Raw)	47.26	7.472	1.62	0.026	42.702
Cengal wood	240 ℃	50.80	7.19	1.99	0.023	40.567
Cengal wood	270 ℃	52.91	6.923	1.96	0.018	36.979
Cengal wood	300 ℃	55.82	6.836	1.88	0.013	34.221
Kulim wood	27 ℃ (Raw)	46.75	7.537	1.42	0.011	43.262
Kulim wood	240 ℃	50.28	6.997	0.87	0.017	40.736
Kulim wood	270 ℃	52.42	6.772	0.78	0.014	38.724
Kulim wood	300 ℃	54.26	6.586	0.68	0.016	37.118
Meranti wood	27 ℃ (Raw)	46.87	7.528	2.05	0.012	42.45
Meranti wood	240 ℃	47.4	6.682	1.86	0.009	42.94
Meranti wood	270 ℃	49.61	6.386	1.43	0.005	41.39
Meranti wood	300 ℃	51.54	5.601	0.95	0.004	40.58

As the temperature increase, the moisture content (MC) and volatile matter (VM) of the torrefied biomass are more apparent. Both moisture content and volatile content of cengal wood biomass represented the highest volatile content. Meanwhile, the fixed carbon (FC) and ash content are decreased with increasing of torrefaction temperature as shown in Table 4-1. The higher heating values of the torrefied and non-torrefied biomass samples also are presented in Table 4-1.

From the results obtained as shown in Table 4-2, the carbon content is increased, meanwhile the element of hydrogen, oxygen, nitrogen and sulphur are decreased after torrefaction. The oxygen content of torrefied *cengal* wood obtained the lowest value compared to torrefied *kulim* wood and *meranti* wood at temperature of 300 ºC. Based on the ultimate analyses, the torrefaction process intensifies the carbon and reduces the oxygen content of the torrefied sample as the process conditions get more severe at high temperature.

There will be difference in value of fixed carbon in Table 4-1 and carbon in Table 4-2 as some carbon lost in hydrocarbons in volatile matter. Fixed carbon is arrived by substracting the volatile matter. Ultimate analysis determines the total carbon content present in volatile matter.

4.3   High Heating Value, Mass and Energy Yield

The important property of biomass fuel, the value of HHV (MJ/kg) is also reported in Table 4.1 for torrefied and non-torrefied wood biomass samples. It is proved that the values of HHV of torrefied biomass obtained from the experiment have increased against the temperature as shown in Figure 4.2(a). From the graph bar plotted in Figure 4-2, the HHV of the torrefied *cengal* wood, for instance, increased to 23.25 MJ/kg, which is comparable to torrefied *kulim* wood and *meranti* wood that have HHV value of 22.16 MJ/kg and 20.49 MJ/kg, respectively at higher torrefaction temperature of 300 °C.
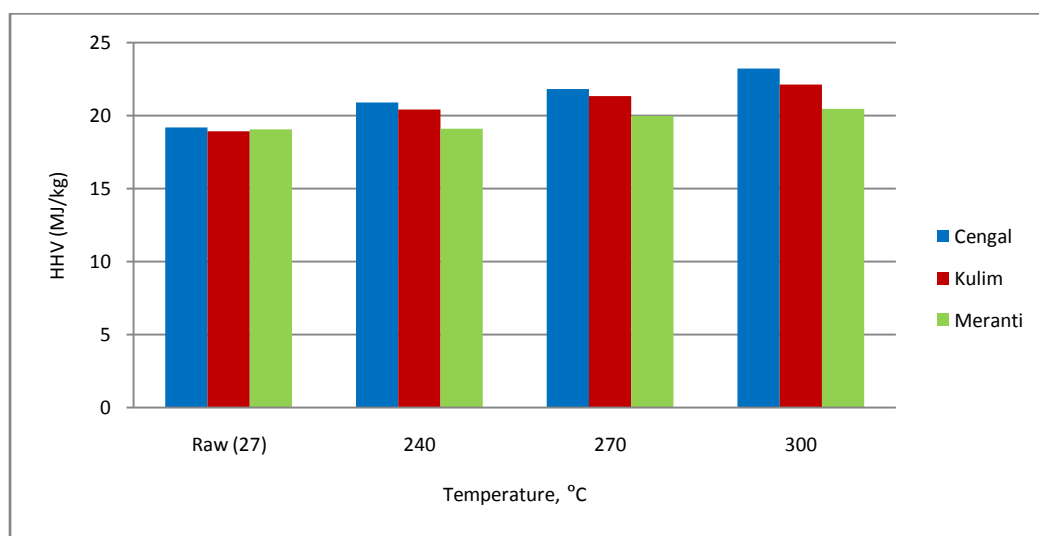

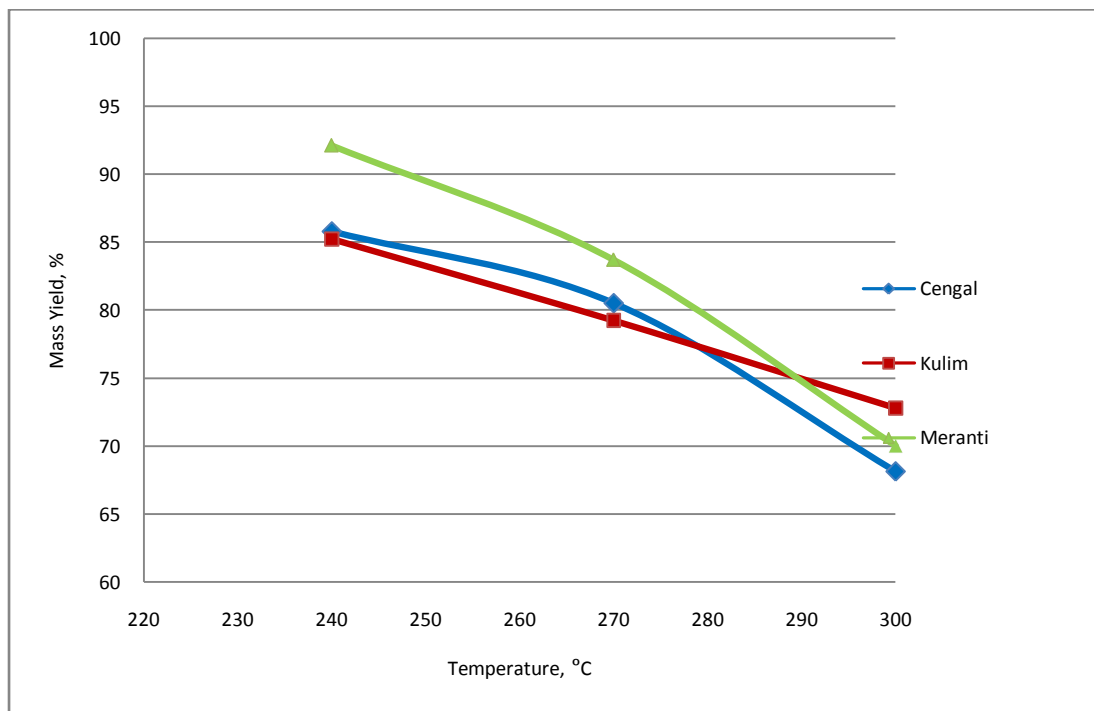Figure 4-2: HHV for torrefied wood biomass.

Figure 4-3: Mass yield percentage of torrefied biomass at different torrefaction temperatures.

Figure 4-3 shows the influence of the torrefaction temperature on the mass yield of the torrefied product. As expected, an increase in temperature decreased the mass yield of the solid product. The decrease in mass yield is more significant for a higher torrefaction temperature (>270 °C). The mass and energy yield will be considered to evaluate the torrefaction process of biomass. The heating rate to the final temperature has an influence on the final mass yield. However, the influence is only noticeable and having slightly different for temperatures above 270 °C. For torrefaction temperatures above 270°C, the exothermic chemical reaction commences. As the process becomes exothermic, released heat increases the volatile releasing rate in turn the mass loss.

From the graph, *meranti* wood recorded the highest difference mass yield percentage from 240º to 300ºC torrefaction temperature for about 22.11% compared to *cengal* wood and *kulim* wood which are 17.63% and 12.25%, respectively. Both *cengal* wood and *kulim* wood almost had same difference mass yield. However, between all these three types of wood, *kulim* wood gave the highest mass yield percentage which is 72.80% compare to *cengal* wood (68.17%) and *meranti* wood (70.03%) at 300 ºC torrefaction.
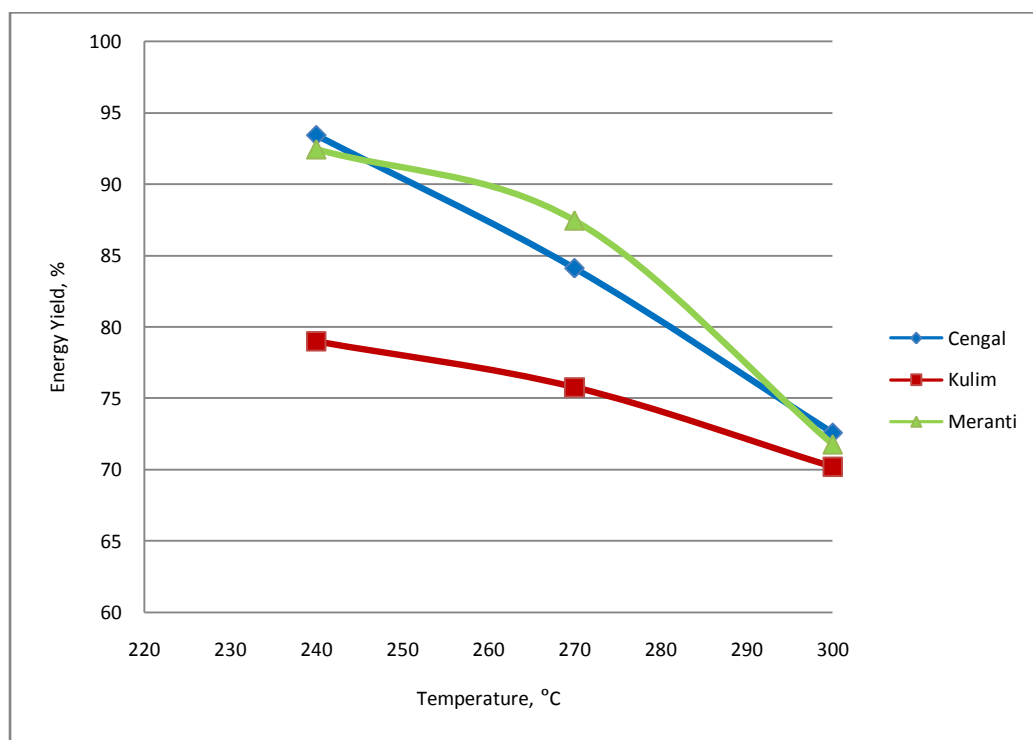
Figure 4-4: Energy yield percentage of torrefied wood biomass at different torrefaction temperatures.

The energy yield decreases with increasing temperature at 30 minutes of torrefaction as shown in Figure 4-4. Similar trends were also observed for the mass yield because the change in energy yield is more significant for a torrefaction temperature above 270 °C. From the graph plotted, *kulim* wood gave the highest energy yield percentage (75.49%) compared to *meranti* wood (71.78%) and *cengal* wood (72.57%) at 300ºC torrefaction temperature. However, the difference of percentage energy yield is only significant between these types of wood biomass.

In all the cases studied, the mass reduction is greater than the energy reduction because of the loss of water and carbon dioxide, which do not contribute to the final energy content of the torrefied product.

4.4   Performance of hydrogen gas

Figure 4-5 shows the influence of torrefaction temperature on the hydrogen gas production. The operating condition of fluidized bed gasifier was tested at 900 ℃ by simulation in Aspen Plus.
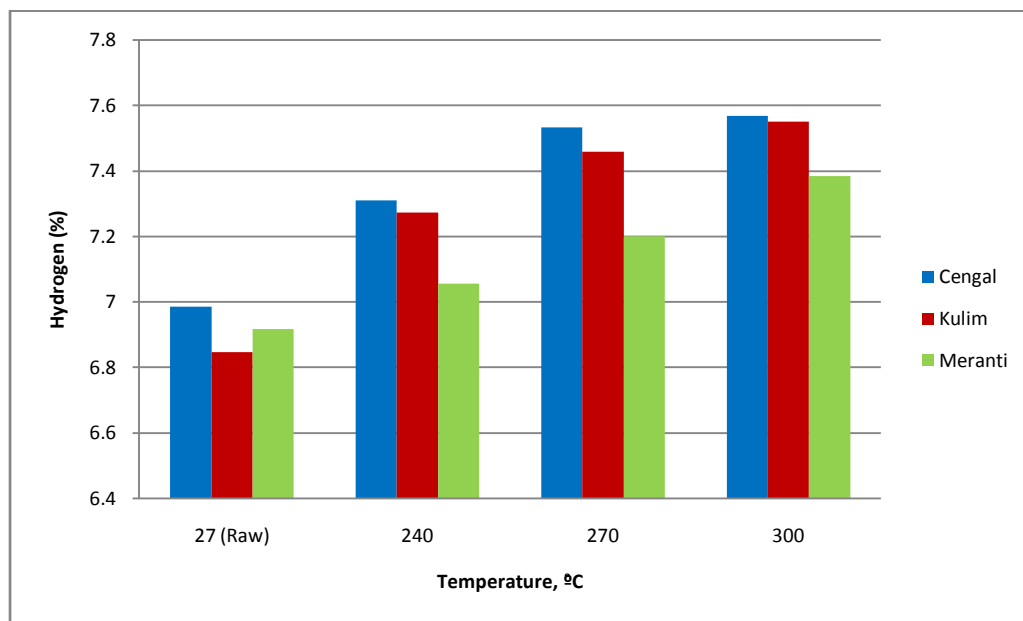
Figure 4-5: Performance of hydrogen gas between different types of wood biomass.

Based on this figure, the highest torrefaction temperature which is at 300 ℃ gave higher production of hydrogen gas. *Cengal* wood recorded as the best type of wood biomass in producing hydrogen gas which is comparable to *kulim* wood and *meranti* wood. There is only significant difference of hygrogen gas production of *cengal* wood at torrefaction temperature between 270 ℃ and 300℃. *Cengal* wood biomass produced hydrogen at 270 and 300 torrefaction From the graph bar plotted, *cengal* wood gave the highest performance in term of syngas produced especially hydrogen gas (7.57%) compared to torrefied *kulim* wood (7.55%) and *meranti* wood (7.38%).

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1   Conclusion

Torrefied biomass is viewed as an attractive feedstock for combustion and gasification, mainly due to its advantages over raw biomass in storage, handling and transportation. The torrefied biomass induced the production of hydrogen gas in term of energy generated compared to raw biomass. From three types of wooden biomass tested in this study, *cengal* wood gave the highest performance in term of syngas produced especially hydrogen gas compared to torrefied *kulim* wood and *meranti* wood. Based on the experiment, the best torrefaction temperature is at 300ºC. The torrefied biomass induced the production of hydrogen gas in term of energy generated compared to raw biomass. From three types of wooden biomass tested in this study, *cengal* wood gave the highest performance in term of syngas produced especially hydrogen gas (7.57%) compared to torrefied *kulim* wood (7.55%) and *meranti* wood (7.38%).

5.2   Recommendation

Since the production of hydrogen gas from all three types of wood biomass (*cengal*, *kulim*, *meranti*) for generating power is in the small amount (less than 10%), other types of wood is recommend to increase the hydrogen gas production itself. For example, acacia wood can be tested as biomass sample under torrefaction and gasification process to determine the hydrogen gas produced for generating power's purpose due to its availability and abundance are higher compared to these three types of wood in Malaysia. Acacia trees grow wild in country since it can adapt with Malaysia's climate which can be categorized as equatorial, being hot and humid throughout the year.

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APPENDIX

Part A: Data extracted from torrefaction experiment

experiment torrefaction
by: Nuralyaa binti Ibrahim

size of biomass used: 500 μm - 1mm
time of torrefaction each samples : 30 minutes

| R = reactor |
| B = biomass sample |
| G = glasswools |

Sample	temp, C	run 1						mass loss, g	
		initial mass, g				final mass, g			
		R	B	G	R+B+G	B	R+B+G	B	R+B+G
kulim	240	140.9572	1.1189	0.1723	142.2395	0.9538	142.0521	0.1651	0.1874
	270	140.9472	1.1065	0.1958	142.2445	0.8768	142.0078	0.2297	0.2367
	300	140.9543	1.1285	0.1969	142.2772	0.8215	141.9612	0.307	0.316
cengal	240	140.6484	2.0294	0.3064	142.9783	1.7412	142.6704	0.2882	0.3079
	270	140.7724	2.1771	0.2363	143.1801	1.7532	142.7418	0.4239	0.4383
	300	140.6399	2.2506	0.2698	143.1543	1.5342	142.4549	0.7164	0.6994
meranti	240	140.9455	1.1245	0.1531	142.2236	1.0361	142.0842	0.0884	0.1394
	270	140.9506	1.1936	0.1906	142.3348	0.9993	142.1066	0.1943	0.2282
	300	140.863	1.1959	0.1893	142.2324	0.8375	141.9041	0.3584	0.3283

mass yield, %

	240	270	300
cengal	85.79876	80.52914	68.16849
kulim	85.24444	79.24085	72.79575
meranti	92.13873	83.72151	70.03094

energy yield, %

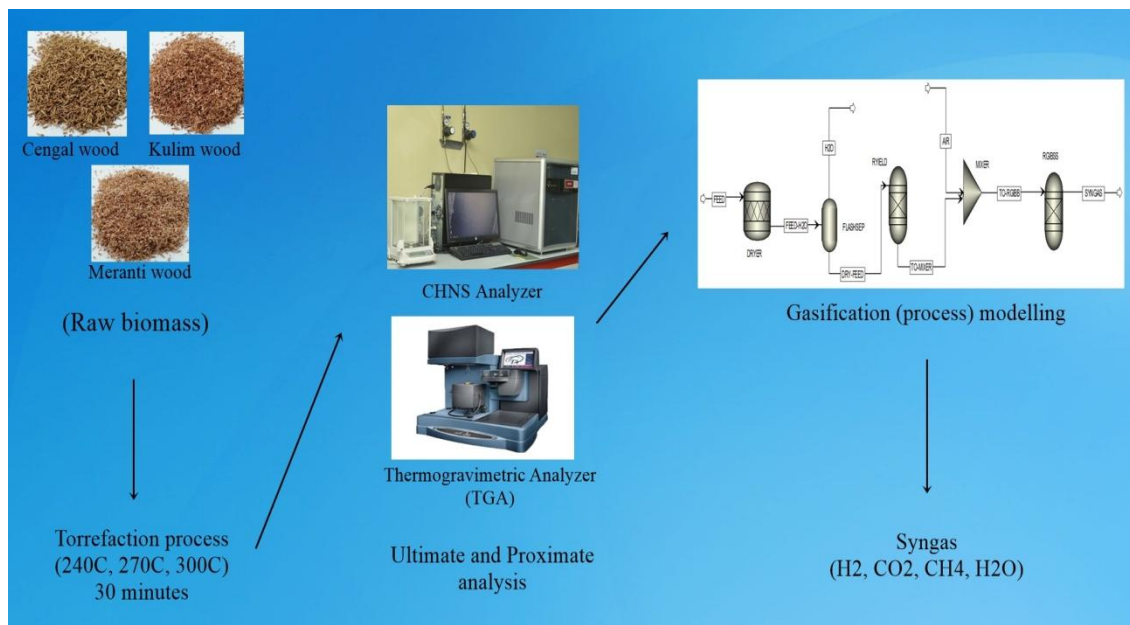|	240	270	300
cengal	93.44419	84.15103	72.56948
kulim	91.99915	82.88678	75.48684
meranti	92.47694	87.43955	71.78259

Figure A-1: Methodology for both torrefaction and gasification process of wood biomass.

Part B: Instrument used for analysis.

Figure A-2: CHNOS Analyzer. (*Source: UMP Central Laboratory*)

Figure A-3: Thermogravimetric Analyzer (*Source: UMP CARIFF*)