The Production of Synthetic Diesel from Biomass

Krongkaew Laohalidanond*

Coking, Briquetting and Thermal Waste Treatment Group, Faculty of Georesources and Materials Engineering, RWTH Aachen University, Germany

**Corresponding author. E-mail: laohalidanond@kobra.rwth-aachen.de*

ABSTRACT

This paper discusses the situation of biomass potential in Thailand and the alternative methods for the production of synthetic liquid transportation fuel from biomass to replace the conventional biofuels derived from esterification and fermentation processes which have some disadvantages, e.g., they cannot be used directly in the automobile without any blending with conventional fossil fuel.

The integration of biomass gasification and the Fischer-Tropsch synthesis (BG-FT) and the catalytic depolymerization processes (CDP) will be considered as an alternative way to overcome the disadvantages of both conventional biofuels. From the theoretical calculations of this work, the maximum synthetic diesel fuel from BG-FT with amount of 260.54 g/kg biomass for oxygen gasification and 373.24 g/kg biomass for steam gasification can be obtained, when palm oil fiber is used as feedstock. The maximum synthetic diesel fuel from CDP is 299.36 g/kg biomass when palm oil shell is used as feedstock.

In this work, the calculation is based only on the stoichiometric equations for BG-FT and on energy efficiency from former experimental works for CDP. In the future work, the model of BG-FT will be carried out by using Aspen Plus® program, whereas the CDP will be set up in laboratory for investigation of the effects of different conditions.

Key words: Synthetic Diesel, Biomass Gasification (BG), Fischer-Tropsch Synthesis (FT), Catalytic Depolymerisation Process (CDP)

INTRODUCTION

Nowadays, the industrial and transportation sector in Thailand is rapidly developing and growing continuously, therefore, its energy demand increases similarly. However, Thailand has insufficient fossil fuel resources to meet its own needs. Consequently, Thailand's energy supply depends on imported crude oil from Middle East countries. As today, according to the very high price of crude oil, Thailand faces the problem of oil crisis, therefore the government and also private organizations try to find the alternative ways to solve this problem. The use of biomass is one of the best alternative solution because biomass is still the cheapest fuel available in Thailand and it will be the major alternative fuel source in the future (Papong et al., van Thujl et al., 2003; Ministry of Energy, 2004). In addition to solving the problem of oil crisis, the utilization of biomass is CO_2-neutral which means the biomass fuel emission has no effect on the CO_2-cycle because only assimilated CO_2 is emitted. This is important because CO_2 is the primary factor in the global warming process.

Biomass is an organic material from various sources which is a natural source of energy and can be used to produce energy. Biomass can be ranged from agricultural products to waste streams as follow (Papong et al., 2001).

- Agricultural crop such as sugarcane, cassava, corn, etc., that are sources of carbohydrates, starch and sugar, including energy crops, can be used for the production of vegetable oil to energy fuel.
- Agricultural residues such as rice straw from rice fields, cassava rhizome from tapioca fields, corncobs from cornfields, etc.
- Wood and wood residues such as fast-growing trees, wood wastes from wood mill, waste from pulp and paper mill, etc.
- Waste streams such as rice husk from rice mills, molasses and bagasse from sugar refineries, residue from palm oil extraction plants, municipal solid waste, etc.

In this study, the biomass can be divided into two categories: the agricultural-based biomass and the wood-based biomass.

Agricultural-based biomass

Although there are many agricultural products available in Thailand, only 4 main agricultural products are taken into account. These are sugarcane, rice, cassava and palm oil. The annual production of these products from crop year 00/01 to 05/06 has been reported by the Office of Agricultural Economics of Thailand and is illustrated in Table 1 (Office of Agricultural Economics, 2005).

Table 1. Annual Production of Agricultural Products (kton).

	00/01	01/02	02/03	03/04	04/05	05/06
Sugarcane	49,563	60,013	74,263	70,101	67,900	63,621
Rice	25,844	26,523	26,057	26,841	24,977	26,493
Cassava	19,064	18,396	16,868	19,718	16,977	18,246
Palm oil	3,256	4,089	4,001	4,903	5,192	5,513

Biomass residues of these products are bagasse and trash from sugarcane, rice straw and rice husk from rice, cassava rhizome and stalk from cassava and fibre, empty fruit bunch and shell from palm oil. The potential of these residues can be estimated from Crop-to-Residue Ratio (CRR), Surplus Availability Factor (SAF) and Lower Heating Value (LHV) of each biomass which are shown in Table 2 (Papong et al., 2001; Ministry of Energy, 2004; Energy for Environment, 2005).

The biomass potential for 4 main agricultural products, based on crop year 05/06, has been shown in Table 2 and is accounted for 41.22 million tons and a total energy potential of $51{,}232 \times 10^6$ GJ. In addition to these biomass residues from

4 main agricultural products, there are also biomass residues from other sources, e.g., corncob, coconut, etc.

Table 2. CRR, SAF, LHV and biomass potential of 4 main agricultural products in 05/06.

Source	Residues	CRR	SAF	LHV (kJ/kg)	Potential (kton/year)	Potential (10^6GJ/year)
Sugarcane	Bagasse	0.29	0.01	7,368	184.5	1.36
	Trash	0.30	0.98	15,479	18,704.57	289.53
Rice	Rice husk	0.23	0.45	14,204	2,742.03	38.95
	Rice straw	0.45	0.78	12,330	9,299.04	114.66
Cassava	Cassava rhizome	0.46	0.98	5,494	8,225.30	45.19
	Stalk	0.09	0.70	13,380	1,149.50	15.38
Palm oil	Fiber	0.15	0.13	11,800	107.50	1.27
	EFB	0.25	0.58	7,240	799.39	5.79
	Shell	0.05	0.04	16,900	11.03	0.19

Wood-based biomass

There are a lot of forest areas and wood products in Thailand, however, only 3 types of wood have been considered as fuel wood in this study. These are rubber wood, eucalyptus and palm oil tree (Papong et al., 2001). The annual production of rubber, palm oil and eucalyptus is shown in Table 3 (Thaiutsa et al., 1997; Papong et al., 2001; King Mongkut's Institute of Technology Thonburi, 2004).

Table 3. Annual Production of Woody Products (kton).

Wood-based Biomass	Production (kton)
Rubber wood in year 2004	6,867
Eucalyptus in year 2000	9,299
Palm oil tree in crop year 05/06	5,513

The woody biomass residue can be separated into barks from forest and saw mill from wood processing. The potential of woody biomass can also be calculated from CRR, SAF and LHV, the same as for agricultural-based biomass. Table 4 shows the CRR, SAF and LHV (Junginger et al., 2001; Papong et al., 2001; King Mongkut's Institute of Technology Thonburi, 2004) and also the woody biomass potential in Thailand.

The potential of woody biomass in Thailand amounts to 23.13 million tons per year with the energy potential of 195×10^6 GJ/year.

Table 4. CRR, SAF, LHV and biomass potential of woody biomass.

Source	Residues	CRR	SAF	LHV (kJ/kg)	Potential (kton/year)	Potential (10^6GJ/year)
Rubber wood	Saw mill	0.20	1	13,962	1,373.44	19.18
	Bark	0.40	1		2,746.89	38.35
Eucalyptus	Saw mill	-	0	6,300	0	0
	Bark	1	0.50		4,649.25	29.29
Palm oil tree	Frond	2.604	1	7,540	14,355.86	108.24

There are many routes to produce liquid transportation fuel from biomass, e.g., extraction, fermentation, pyrolysis and other thermo-chemical conversion (van Thuijl et al., 2003), but the production of transportation liquid fuel in Thailand is based currently on transesterification process to produce biodiesel from oil-containing biomass and fermentation process to produce ethanol from bagasse (Ministry of Energy, 2004). The main disadvantages of these conventional processes are:

- Both ethanol and biodiesel cannot be directly used in the automobile. They have to be blended with the commercial gasoline and diesel, respectively, with the ratio of 85:15 blends of gasoline and ethanol (Ministry of Energy, 2004) or not more than 5% biodiesel in conventional diesel (van Thuijl et al., 2003).
- Both renewable transportation fuels can only be derived from the oil- and starch-containing biomass. Lignocellulosic biomass cannot be used as feedstock to produce biofuel via these processes with high efficiency (Specht et al., 2005)
- They also cannot be distributed using existing infrastructure (van Thuijl, 2003).

In this study, the objective is to investigate the possible processes to produce synthetic diesel that can be directly used in the automobile and existing infrastructure

MATERIALS AND METHODS

Biomass feedstock

All of the agricultural biomass residue in Thailand from Table 2 and the woody biomass from Table 4, including corncob, were used as the raw material for the theoretical calculation of liquid transportation fuel production. The biomass characteristics are reported by EFE (Energy for Environment, 2005) and have been listed in Table 5.

Table 5. Proximate and elemental analyses of biomass feedstock.

	Bagasse	Cane trash	Rice husk	Rice straw	Cassava rhizome	Corncob
Proximate analysis (wt% as received)						
Moisture	50.73	9.20	8.20	10.00	59.40	7.00
Volatile	41.98	67.80	58.90	60.70	31.00	70.40
Fixed C	5.86	16.90	19.70	18.90	8.10	21.20
Ash	1.43	6.10	13.20	10.39	1.50	1.40
Elemental analysis (wt% as received)						
C	21.33	41.60	39.10	38.17	18.76	43.70
H	3.06	5.08	4.59	5.02	2.48	5.21
N	0.12	0.40	0.18	0.58	0.32	0.19
O	23.29	37.42	34.70	35.28	17.50	42.50

	Palm oil fiber	Palm oil EFB	Palm oil shell	Rubber wood tree	Eucalyptus tree	Palm oil tree
Proximate analysis (wt% as received)						
Moisture	31.84	58.60	12.00	45.00	50.00	48.40
Volatile	48.61	30.44	68.20	45.70	35.00	38.70
Fixed C	13.20	8.04	16.30	7.71	11.95	11.70
Ash	6.35	2.92	3.50	1.59	3.05	1.20
Elemental analysis (wt% as received)						
C	31.35	15.11	44.44	15.56	13.25	23.89
H	4.57	1.51	5.01	3.19	2.65	3.04
N	0.02	2.57	0.28	0.14	0.19	0.56
O	25.63	19.13	34.70	24.48	20.85	22.91

The production of transportation fuel

To overcome the disadvantages of both conventional biofuels, the integration of Fischer-Tropsch synthesis and Biomass Gasification (BG-FT) and the Catalytic Depolymerisation Process (CDP) are considered in this work. Both are the thermo-chemical conversion of biomass to liquid transportation fuel.

The **BG-FT** can be separated into 3 sub-processes; biomass gasification, gas cleaning and Fischer-Tropsch process (Tijmensen et al., 2000) as shown in Figure 1.

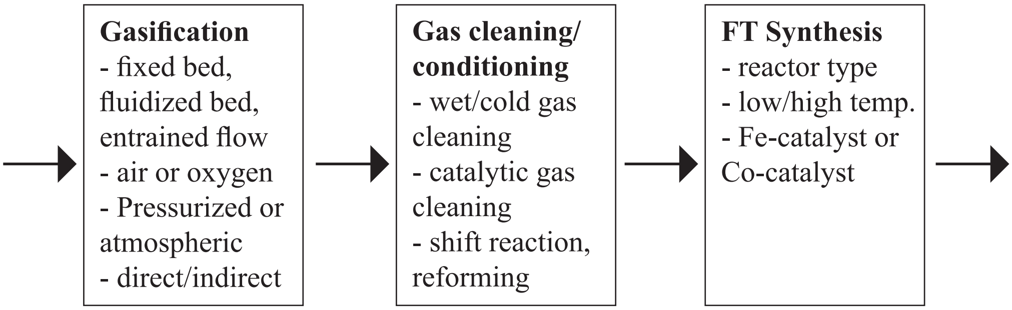

Figure 1. A schematic presentation of converting biomass to FT-liquids.

At first, the biomass material is converted into the synthesis gas which comprises mainly of hydrogen and carbon monoxide at the elevated temperature of between 700-1500°C (van Thuijl et al., 2003), depending on the type of gasifier and the characteristics of biomass feedstock. This sub-process is called "biomass gasification process". The synthesis gas must be free from tar, particulate and other catalyst-poisoning substances, therefore, this synthesis gas has to be cleaned and conditioned until it is suitable to be used as feedstock for Fischer-Tropsch reaction. This step is named as "gas cleaning and conditioning process". After the synthesis gas has been cleaned, it can be used as feedstock to produce synthetic diesel in the Fischer-Tropsch process. The FT reaction is a process to convert synthesis gas to a wide range of long-chain hydrocarbon products, from LFG, gasoline, naphtha, diesel and wax (Tijmensen et al., 2000), with the aid of iron- or cobalt-based catalysts under high temperature and high pressure. The produced long-chain hydrocarbon is finally distilled, hydro-cracked or upgraded before being used as liquid transportation fuel.

The maximum amount of liquid fuel from the BG-FT process has been investigated from the stoichiometric chemical equations of both biomass gasification process and Fischer-Tropsch process and the composition of biomass feedstock (Vogel et al., 2003). The stoichiometric chemical equations of both processes are described as follows:

Biomass gasification with oxygen as gasification agent

$$C_aH_bO_c + xO_2 \rightarrow yCO + zH_2 \qquad \text{(Eq. 1)}$$

Biomass gasification with water vapour as gasification agent

$$C_aH_bO_c + xH_2O \rightarrow yCO + zH_2 \qquad \text{(Eq. 2)}$$

Simple Fischer-Tropsch reaction with n = 1

$$CO + 2H_2 \rightarrow CH_2 + H_2O \qquad \text{(Eq. 3)}$$

The **CDP** is the alternative route to produce liquid transportation fuel from biomass. This process is principally based on direct liquefaction of biomass (Willner, 2005; Willner et al., 2004; Willner, 2005). In this process, the long-chain hydrocarbons or organic materials have been cracked into light crude oil with the aid of ion-exchanged catalysts under a temperature of less than 500?C and atmospheric pressure (Willner, 2005). The example of depolymerisation reaction of organic feedstock into liquid transportation fuel is illustrated in Figure 2.

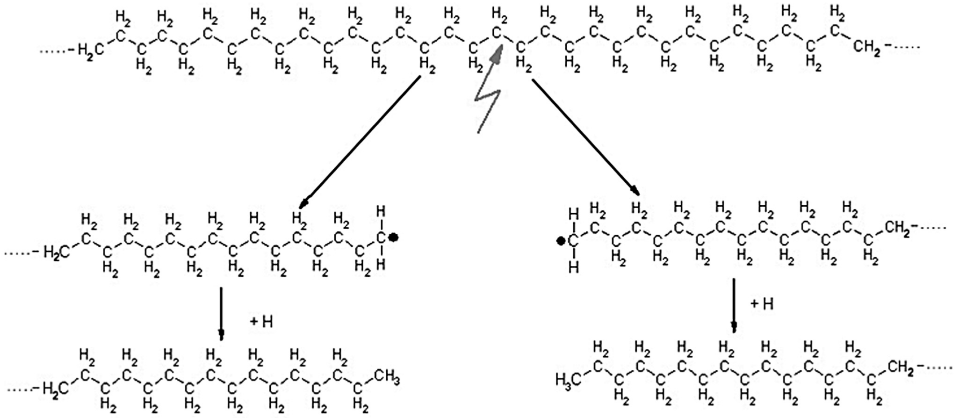

Figure 2. Depolymerisation reaction of organic feedstock into diesel.

Because there are no exact chemical equations for the catalytic depolymerisation process, the results of this process can be obtained only from the experimental work. There are many studies for investigation of liquid fuel production from CDP available (Willner, 2005; Willner, 2005; Willner et al., 2004). The results of the experimental work can be summarized as follows.

The experiment was carried out at the reaction temperature of about 370°C at atmospheric pressure on the laboratory scales with a biomass capacity of 1 kg/h. At first, the biomass has been tested to study the mass balance of the process and the result is (Willner, 2005):

100 %wt biomass → 43 %wt oil + 22 %wt water + 15 %wt coal + 20 %wt gas

For the energy balance consideration, 5% of energy consumption (EC) has been taken into account (Willner et al., 2004).

100 %e biomass + 5 %e EC→ 76 %e oil + 20 %e coal + 7 %e gas + 2 %e energy loss

From both mass and energy balances, it can be concluded that the oil yield from the CDP is more than 40% by weight. and the energy efficiency is also more than 70%.

These following assumptions will be used for the estimation of the theoretical amount of diesel produced from CDP:

- Energy efficiency of this process is 70% (based on higher heating value). This value is the minimum value from (Willner, 2005).

- Higher heating value of produced synthetic diesel fuel is 42,714 kJ/kg (Willner, 2005).
- Density of produced synthetic diesel fuel is 865 kg/m3 (Kaltschmitt and Hartmann, 2001).
- Higher heating value of biomass from Thailand is obtained from (Energy for Enviroment, 2005).

RESULTS AND DISCUSSION

According to the biomass analysis and the Equations 1 to 3 in the last section, the results of the maximum (theoretical) liquid fuel production via BG-FT, based on 1 kg of biomass, have been shown in Table 6, in this case when both oxygen and water vapor are used as a gasification medium.

Table 6. The maximum amount of liquid fuel production via BG-FT from Thai biomass.

Type of biomass	Chemical formula	Max. fuel production with O_2 as agent (g/kg biomass)	Max. fuel production with H_2O as agent (g/kg biomass)
Sugarcane trash	$C_{4.1}H_6O_{2.8}$	210.00	301.00
Bagasse	$C_{3.7}H_{6.4}O_3$	226.80	274.26
Rice husk	$C_{4.1}H_{5.8}O_{2.8}$	203.42	294.56
Rice straw	$C_4H_{6.3}O_{2.8}$	222.60	301.56
Palm oil fibre	$C_{4.2}H_{7.4}O_{2.6}$	260.54	373.24
Empty fruit bunch	$C_{3.4}H_{4.1}O_{3.3}$	146.86	154.14
Palm oil shell	$C_{4.4}H_{5.9}O_{2.6}$	205.94	331.52
Cassava rhizome	$C_4H_{6.4}O_{2.8}$	225.82	310.52
Rubber wood	$C_4H_6O_{2.4}$	227.36	348.46
Eucalyptus	$C_{4.1}H_{5.7}O_{2.8}$	200.20	280.84
Palm oil tree	$C_4H_{6.1}O_{2.9}$	212.52	289.10

From Table 6, it can be indicated that:

- Biomass which consists of the higher hydrogen content can generate the more amount of liquid transportation fuel. This is due to the hydrogen content in biomass contributes to promote the more hydrogen in synthesis gas, compared to biomass with less hydrogen content.
- The gasification system with water vapor as a gasification agent can produce more amount of liquid fuel than with oxygen as gasification agent because hydrogen in water vapor will increase the amount of hydrogen in synthesis gas.

This means that the amount of produced liquid fuel depends strongly on the synthesis gas component. The ratio of hydrogen to carbon monoxide molecule in synthesis gas has to be 2:1, as shown in Eq. 3. Besides, the composition of biomass, the use of gasification agent, the type of gasifier and operation temperature and pressure have also the effects on synthesis gas composition (Kaltschmitt and Hartmann, 2001). The special gasifiers for production of synthesis gas are presented together with the synthesis gas composition in Table 7.

Table 7. Special gasifiers for synthesis gas production (Henrich and Dinjus, 2003; Rudloff, 2005; Schingnitz and Volkmann, 2004; Sonntag et al., 2004; Vodegel et.al, 2005, Rauch, 2004).

	Carbo V	Entrained flow gasifier	CFB (CUTEC)	Blue Tower Gasifier	FICFB (TU Wien)
Total system	Low temp. gasifier and entrained flow gasifier	Pyrolysis and entrained flow gasifier	Circulation Fluidized Bed gasifier (CFB)	Pyrolysis and reformimg	Fast internal gasification
1st stage	Low temp. gasifier at 400-600°C	Flash pyrolysis 500°C	-	Pyrolysis 550-600°C	-
Gasifier	Autotherm entrained flow gasifier 2 zones: Combustion 1300-1500°C, Gasification 800-900°C P_{atm}	Autotherm entrained flow gasifier ~1300°C and >50 bar	Autotherm CFB with silica sand as bed, >900°C, P_{atm}	Allotherm reformimg with ceramic as heat carrier, 950°C, P_{atm}	CFB gasification with 2 zones: Combustion with air 970°C, Gasification 900°C
Gasification Agent	O_2/air	O_2	O_2/H_2O	H_2O	H_2O
Gas cleaning	Bag filter, wet scrubber, SO_2-removal	Wet scrubber, cooling, SO_2-removal	Hot gas filter with ceramic, wet scrubber, carbon adsorber	unknown	Filter, wet scrubber, ZnO adsorber, removal of S und CI unknown
Gas conditioning	WGS, CO_2-removal	WGS, CO_2-removal	WGS, CO_2-removal	unknown	unknown
Synthesis gas composition (%vol.) after gas cleaning					
H_2	40.2 (22.1)	27	26.04	53	38-40
CO	39.2 (21.8)	50	29.91	12	22-26
CO_2	20.4 (11.4)	14	33.69	25	20-22
CH_4	0.1 (0)	< 0.1	8.8	6	9-11
N_2	0.1 (44.7)	6.3	0.17	2	1.2-2

The number in the brackets for Carbo V gasifier is the result when air is used as gasification agent.

However, it must be kept in mind that this maximum liquid fuel is calculated under the stoichiometric equations and without any loss condition. Actually, in the real process or real plant, there are many losses, for example, energy loss and friction loss during the gasification process, gas cleaning process, FT-synthesis and also upgrading process. The energy efficiency of BG-FT depends on many processes, ranging from gasification process until upgrading process. From the work of Boerrigter et.al. (Boerrigter et al., 2004), the overall energy efficiency of synthesis gas conversion to final diesel yield (η_{GtL}) has been estimated to be about 71%, based on lower heating value of biomass while the overall energy efficiency of biomass conversion to final diesel yield of 42-50% for pressurized systems and 33-40% for atmospheric systems have been reported (Specht et al., 2005). That means the amount of diesel yield from BG-FT is always less than the result from calculation.

According to the assumption for calculation of the liquid transportation fuel from Thai biomass via CDP and the higher heating value of biomass, the results of liquid fuel production, based on 1 kg of biomass via CDP, have been demonstrated in Table 8.

Table 8. Amount of diesel product from Thai biomass via CDP.

Type of biomass	HHV of biomass (kJ/kg)	Diesel production (g/kg biomass)	Diesel production (l/kg biomass)
Sugarcane trash	16,794	275.22	0.32
Bagasse	9,243	151.47	0.18
Rice husk	15,400	252.38	0.29
Rice straw	13,650	233.70	0.26
Palm oil fibre	13,548	222.03	0.26
Empty fruit bunch	8,969	146.98	0.17
Palm oil shell	18,267	299.36	0.35
Cassava rhizome	7,451	122.11	0.14
Rubber wood	10,365	169.86	0.20
Eucalyptus	8,514	139.53	0.16
Palm oil tree	9,354	153.29	0.18

After the theoretical calculations from both biomass gasification with integrated Fischer-Tropsch synthesis (BG-FT) and catalytic depolymerisation process (CDP), it can be concluded that the CDP can provide more diesel output due to its higher energy efficiency (more than 70%), whereas the energy efficiency of BG-FT is only 33-50%. Besides, the higher heating value, all the operation parameters and the catalysts will affect the amount and quality of liquid fuel produced from CDP. The effects of these parameters will be investigated in the future. In addition to the higher diesel output, the cost of CDP is also less than the BG-FT which consists of a 2-step conversion process: biomass gasification and FT-synthesis, therefore, only CDP will directly be tested on the laboratory scale, whereas the suitable operating condition of BG-FT process will firstly be investigated with Aspen programme.

The advantages of FT-diesel and CDP-diesel are that they are of high quality and clean transportation fuels with very low sulphur content and aromatic compounds. The specification of FT-diesel and diesel derived from CDP is illustrated in Table 9, in comparison with petroleum-derived diesel (van Thuijl et al., 2003; Willner, 2005). In addition, the FT-Diesel and diesel derived from CDP can be directly used in the automobile and its infrastructure without any adaptation.

Table 9. Specification of FT-diesel and CDP-diesel in comparison to conventional diesel.

Fuel specification	FT-Diesel	CDP-Diesel	Conventional diesel
Chemical formula	Paraffin		$C_{12}H_{26}$
Molecular weight (kg/kmol)			170-200
Cetane number	> 74	63	50
Density (kg/l) at 15°C	0.78	0.865	0.84
Lower Heating Value (MJ/kg) at 15°C	44.0	42.7	42.7
Lower Heating Value (MJ/l) at 15°C	34.3		35.7
Stoichiometric air/fuel ratio (kg air/kg fuel)			14.53
Oxygen content (%wt)	~ 0		0-0.6
Kinematic viscosity (mm^2/s) at 20°C	3.57	10.6	4
Flash point (°C)	72	77	77

OUTLOOK

The next step for this work is to carry out both the calculation of gas composition and maximum amount of synthetic fuel from BG-FT process under many process conditions by using Aspen® programme and the laboratory investigations of the CDP with the variation of operation parameters, e.g., temperature, retention time, amount of catalyst, moisture content of biomass and finally, types of biomass feedstock from Thailand. Figure 3 shows the experimental set-up of CDP, which consists of small glass reactor and cooler (Willner, 2004). The selected feedstock in the first phase is biomass residues from the palm oil industries, including palm oil shell, empty fruit bunch and palm oil fibre. Because palm oil is an oil-containing agricultural product, it tends to have more oil product than other sources of agricultural residues. After tests with palm oil residues, the corncob residue is also preferable feedstock material according to diesel output from calculations. However, the municipal plastic waste also seems to be a very interesting feedstock for this process due to its high organic contents and consequently, more product oil should be obtained.

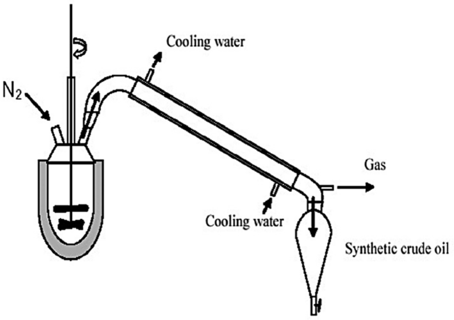

Figure 3. CDP experimental set-up.

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