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공학박사학위논문

**Modeling and Optimal Design of Biomass
Torrefaction Process**

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Abstract

Modeling and Optimal Design of Biomass Torrefaction Process

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Although the potential of biomass is immense as fuel, raw biomass has poor properties such as low energy density, hydrophilicity and high grinding energy requirement. Torrefaction is thermal pretreatment technology to enhance the biomass property. It is called as a mild pyrolysis occurring in the temperature range of 200–300°C and at atmospheric pressure in the absence of oxygen. Biomass torrefaction has been known as a feasible method to convert the biomass feedstock into attractive solid fuel which is utilized for combustion and gasification. Moreover, the torrefied biomass has high energy density, hydrophobic property, high grindability and a lower oxygen to carbon ratio. However, the economical infeasibility of torrefaction process is major difficulty in commercial production of torrefied biomass. In order to overcome the difficulty in economics, structural improvement of process design and optimization of design variables are required. Torrefaction reactor model is needed for the development of reactor and process

designs for biomass torrefaction. In the thesis, a one-dimensional reactor model was developed based on the kinetic model describing volatiles components and solid evolution and the thermochemical model proposed by Bates and Ghoniem [1, 2] considering heat and mass balance. The developed reactor model used the temperature and flow rate of the recycled gas, which can be used as the practical manipulated variables instead of the torrefaction temperature, under operating conditions based on the process scheme proposed by Bergman et al. [3]. The temperature profiles of the gas and solid phase were generated, depending on the practical thermal conditions, using developed model. Moreover, the effects of each selected operating variables on the parameters of the torrefaction process and effect of whole operating variables with particular energy yield were analyzed. Through the results of sensitivity analysis, it is represented that the residence time insignificantly influenced to energy yield when the flow rate of recycled gas is low. Moreover, higher temperature of recycled gas with lower flow rate of recycled gas and residence time produces the attractive properties, including HHV and grindability, of torrefied biomass when the energy yield is specified. Using the developed model, the optimization of operating variables in the basic process design was carried out. For the formulation of optimization problem, the assessment method to evaluate the torrefaction process was analyzed and chosen. In order to develop the objective function of optimization problem, the economic evaluation model was made based on reasonable assumptions. It includes the capital cost of main facilities and operating cost of natural gas and electricity. To enhance the basic process design, the drawbacks in the base case reactor was analyzed and found several opportunities to improve the process efficiency. Based on the opportunities to improve the process, three process alternatives was proposed. The

operating variables of process alternatives were optimized and compared to propose the optimal process design of biomass torrefaction.

Keywords: Biomass, Torrefaction, Reactor modeling, Operating conditions, Gas recycling, Parametric study, Process alternatives, Process optimization

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CHAPTER 1 : Introduction

1.1. Research motivation

The threats of energy system have been maintained for exhaustion of fossil fuels and regional ubiquity. The view that higher oil price will be maintained are increasing and uncertainties of Middle East oil production is concentrated are growing. After 2 times oil crisis, interest in sustainable resources has been increased. Obvious candidate to replace oil have not emerged. As the Kyoto Protocol enters into force, governments and businesses endeavor procurement of technologies for sustainable resources to reduce the greenhouse gas. Figure 1-1 presents the distribution of energy consumption in developed countries and developing countries [4]. Developed countries are highly dependent on oil. With world energy demand projected to rise by about 40% from now to 2020, it is possible that natural gas, which supplies almost 25% of the world's energy demand today, overtakes oil as the most important energy source. This trend is also supported by environmental concerns such as global warming which have resulted in calls for increased use of natural gas. Looking beyond the era of natural gas, coal may become increasingly used, but this requires CO₂ sequestration. Eventually, fossil fuel reserves will dwindle. Biomass resource like trash, grain and agricultural by-product commonly seen in our surroundings are emerging as viable solution. Biomass is the general term for organic materials which the organisms have in the natural world. Biomass

is the typical resource the humankind has historically used for food, energy, construction materials and supplies. However, the value of biomass has not been evaluated because of usability and practicality of fossil fuels. Recently, the situation is changing fast. It is known that the biomass has a potential for sustainable energy and alternatives to products based on petroleum chemistry. Biomass has been unique ground resource to replace the underground resources like oil and coal in the world. Biomass, currently the fourth largest energy source in the world, could therefore in principle become the main energy source. Several scenarios for the future predict a strong increase in the use of biomass between 2025 and 2050.

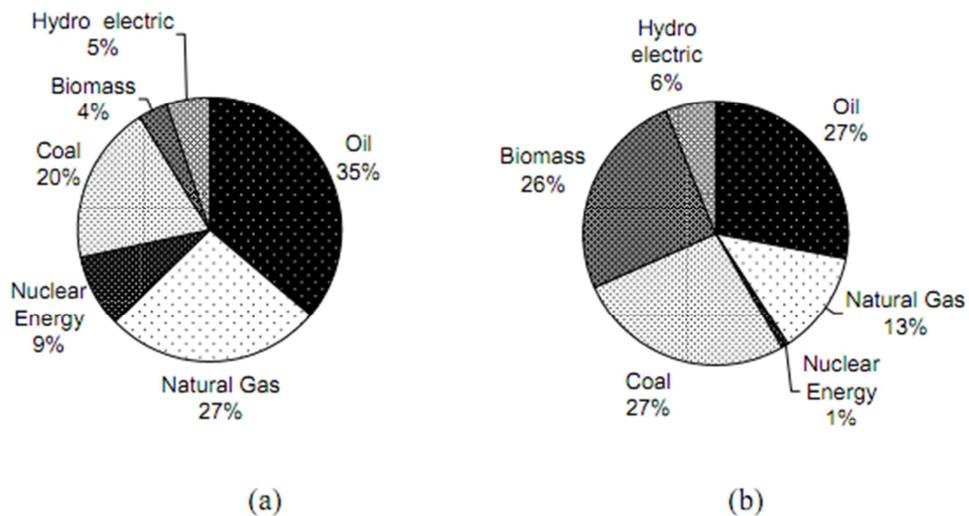


Figure 1-1 Primary energy consumption in a) developed countries, b) developing countries [4]

Biomass torrefaction has been recognized as a technically feasible method of converting raw biomass into a solid that is suitable for commercial and residential

combustion and gasification applications, given that it has high energy density, is hydrophobic, compactable, and grindable, and has a lower oxygen-to-carbon (O/C) ratio. In the development status of torrefaction technology, torrefaction reactor technologies are proven for other applications. Many technology developers (>50) are related due to strong market pull. Several torrefaction demo plants in Europe, have to prove they are able to continuously produce large volumes of high quality torrefied product. As a next step multiple commercial sized (100 kton/yr) torrefaction plants will be erected for supply to Europe, especially in regions where biomass is widely available and relatively cheap. High energy efficiency is crucial in view of overall cost and sustainability in which overall energy efficiency is strongly dependent on heat integration design [5]. However, at this stage of development only results from pilot plants are available. It will be a challenge for developers to develop a full commercial torrefaction plant, which incorporates the necessary design and process modification for good commercial performance [6].

1.2. Research objective

The objective of this thesis is to solve the technical challenge of development status for biomass torrefaction. For this purpose, the optimal design of biomass torrefaction process was proposed based on a simulation study using process model developed in this study for the moving-bed type reactor. Through the modeling, optimization and design results, the meaningful achievement was obtained in each

chapter. Using developed reactor model, the temperature profile in reactor was analyzed and effects of operating conditions on the process parameter were known. To propose the optimal design of biomass torrefaction process, optimization of design variables was conducted based on existing design and process alternatives obtained from analysis of base case were compared. Finally, optimal process design for biomass torrefaction was developed.

1.3. Outline of thesis

Each chapter of this thesis considers a modeling, effect of operating conditions, process optimization and design alternatives for biomass torrefaction. Chapter 2 addresses reactor and process modeling for biomass torrefaction which includes various modeling basis. Using developed model, parametric study is described in Chapter 3. Effects of operating conditions on process parameters and optimization of base case design are addressed for biomass torrefaction process. Chapter 4 addresses design alternatives based on analysis results of base case design and propose the optimal design. Lastly, in Chapter 5, we present the thesis conclusion and recommendation for future works.

CHAPTER 2 : Modeling of biomass torrefaction process

2.1. Introduction

In order to scale up the torrefaction process for an industrial plant, a torrefaction process design is required. As shown in Figure 2-1, a general torrefaction process scheme, the raw biomass is dried in a predrier to reduce the moisture content before torrefaction [3]. The dried biomass is then heated by recycled gas and converted to torrefied biomass in the reactor. After torrefaction, the torrefied biomass is subjected to a size reduction process and a solid shape-forming process such as densification, depending on the application. In the reactor, the volatiles released during torrefaction has a heating value and can be utilized for combustion. This heat energy from combustion of the volatiles is used to heat the torrefaction reactor in the recycling loop in the process. A blower is used to recover the pressure drop of the recycled gas through the reactor for the recycling loop. This heat recovery component of the torrefaction process was proposed by the Energy Research Center of the Netherlands [3]. Because this heat recovery concept is expected to improve the energy efficiency of the torrefaction process and to reduce the price of torrefied biomass, many recent studies on torrefaction in general and the torrefaction process in particular have been based on this concept [7].

It is essential to develop a torrefaction reactor model for process designs based on

this heat recovery concept, because the composition and flow rate of the volatiles and the heat release from the torrefaction reactor determine the designs of all units in the process. Furthermore, the properties and quality of the torrefied product are determined using the torrefaction reactor model.

Studies of biomass torrefaction have mainly focused on experimental research into the effects of various operating parameters such as the temperature, residence time, feed stock, moisture contents and particle size [8, 9]. Few studies on modeling of the torrefaction reaction kinetics and reactor have been performed. Research on mathematical modeling of torrefaction has mainly focused on developing a model to predict the evolution of only the solid product. For example, a kinetic model of solid evolution was developed based on willow torrefaction experiments, using a two-step, first-order torrefaction mechanism [10]. Some authors have also developed data-driven models to calculate the solid yield, based on operating parameters such as the torrefaction temperature, residence time, and initial moisture content, using mathematical regression techniques and experimental data [7, 11]. However, in the development of the process design, not only a kinetic model is required to calculate the solid yield, but a model is also required to estimate the volatiles composition during torrefaction. Bates and Ghoniem developed a model to predict the volatiles and solid compositions during torrefaction and validated it with experimental data. A model of the reaction thermochemistry is required to estimate the torrefaction enthalpy so that the accurate energy balance of the reactor can be taken into consideration. Bates and Ghoniem suggested such a thermochemical

model combined with their kinetic model [2]. In another study, the torrefaction enthalpy was estimated using a heat balance model and experimental data [12]. Recently, Emanuela Peduzzi et al. proposed the model to estimate the mass and energy balance of the torrefaction unit [13]. Yash Joshi et al. developed the tool to simulate a steady state model of the torrefaction process by linking unit operation blocks of drying and torrefaction along with auxiliary process equipment [14].

Most torrefaction studies have regarded the reactor temperature and residence time as manipulated variables in assuming isothermal condition during torrefaction. Various experimental studies on torrefaction have used isothermal conditions, with an electric heater or furnace in the reactor [8, 9], so the torrefaction status was determined based on the reactor temperature and residence time as the operating variables. Although these two variables have been considered to be crucial manipulated variables in experimental studies, excessive flow rate of heat carrier gas should be required to maintain the isothermal condition during torrefaction in practical process; the reactor temperature is not a manipulated variable in practical processes. As Bergman's process scheme in Figure 2-1 shows, the temperature and flow rate of the recycled gas determine the temperature profile of the torrefaction reactor. The temperature profile is not able to maintain isothermal when the flow rate of the recycled gas is not excessive. Thus, the flow rate and temperature of the recycled gas should be replaced by the reactor temperature as the manipulated variable of the torrefaction reactor in a simulation study on torrefaction process. However, studies on torrefaction process using simulation have used assumption of

isothermal condition of torrefaction reactor [2, 13, 14] .

In this chapter, we developed a reactor model combining the existing kinetic model and thermochemical model of the solid yield and volatiles composition with heat transfer model for pyrolysis of biomass particle, to calculate the temperature profile in the reactor based on the mass and energy balances, considering energy balance between the heat carrier gas and biomass in the reactor. The required heat energy and information on the volatiles and product quality were evaluated using this model, considering practical operating conditions. Developed reactor model was integrated with the process model to predict the effect on process parameters.

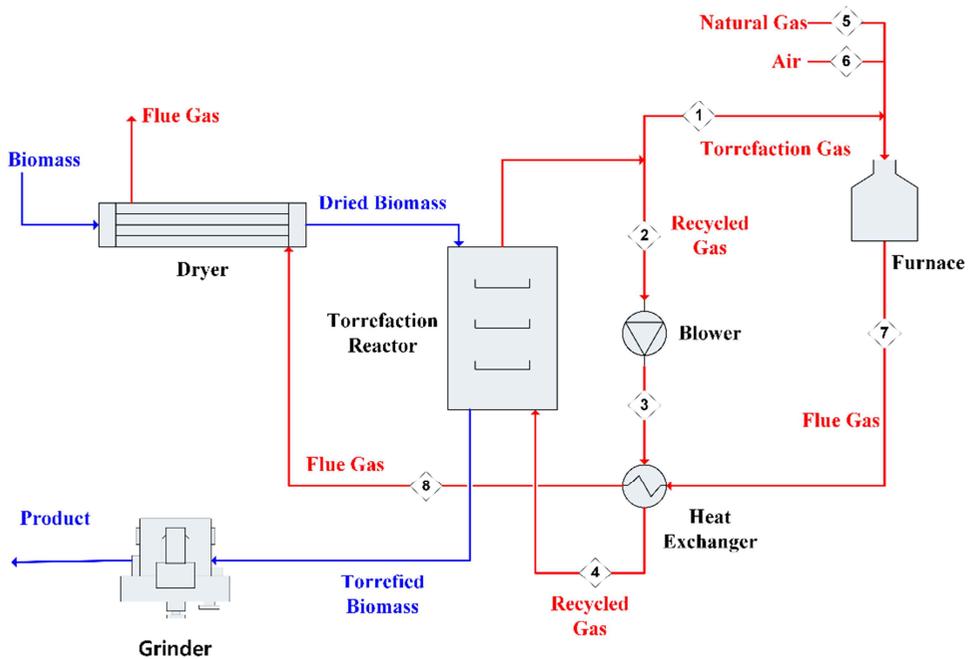


Figure 2-1 Scheme of torrefaction process [3]

2.2. Modeling

2.2.1. Conditions of reactor model

The base case model of torrefaction reactor was developed based on the conditions shown in Table 2-1. The modeling conditions of the base case in this chapter, except for the feedstock species, are based on the conditions used in the case study, in which Bergman et al. carried out a case study for process design and economic evaluation of the torrefaction process [3]. A moving-bed-type reactor was chosen and the production rate was determined for one moving-bed reactor. Moving bed reactor is originated from biomass gasification technology. Raw solid biomass particles enter from the top of the reactor. The particles undergo drying and torrefaction and exit at the bottom of the reactor. Neutral (oxygen free) hot gases enter the bottom of the column and travel upward. The loaded gases exit the top of the reactor. A condenser extracts water vapor and other condensable substances from the gas. Dry gas is combusted in a burner to generate hot gases for recirculation through the reactor. Excess gas is filtered before releasing it to the environment. Figure 2-2 shows the preliminary configuration of the reactor [15]. Biomass particles enter a hopper at the top of the reactor. The material is then passed through an airlock as it fills up the entire length of the column. The bottom of the reactor is a hopper leading to an airlock and a screw conveyor. The function of the screw conveyor is to cool the hot torrefied material to a temperature below its

ignition temperature. Raw solid biomass particles enter from the top of the reactor
Neutral (oxygen free) hot gases enter the bottom of the column and travel upward.
The loaded gases exit the top of the reactor

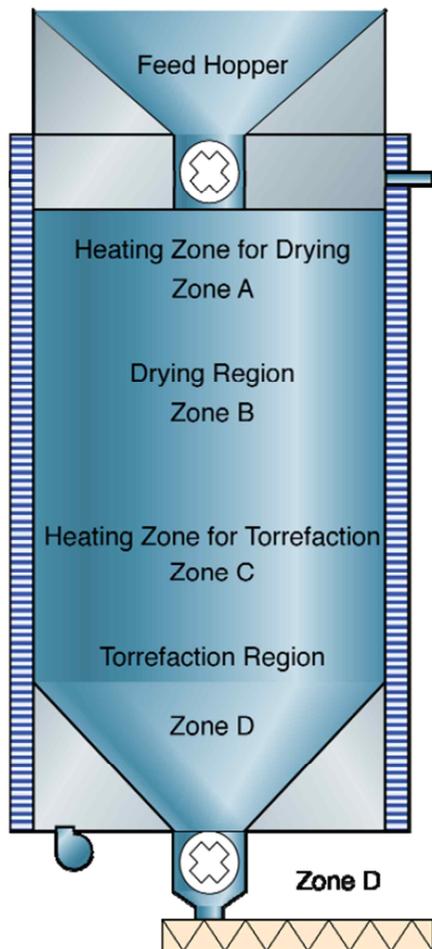


Figure 2-2 scheme of moving bed reactor [15]

Willow was used as the feedstock in this chapter, because the applied kinetic model is based on experiments on willow torrefaction, as described in next paragraph. Bergman et al. selected the reactor temperature and residence time as the

manipulated reactor variables [3]. As explained in previous section, the reactor temperature had to be replaced by the temperature and flow rate of the recycled gas as the manipulated variables in a practical model of the torrefaction reactor. Thus, in this chapter, we used the reactor temperature in Bergman’s case study for the temperature of the recycled gas. The recycled gas flow rate was calculated to achieve the energy yield target using the reactor model, as described in next section.

Table 2-1 Input data for reactor model for base case

Input data	Value	Reference
Feedstock	Willow chips	Bates (2012)
Feedstock size	$10 \times 30 \times 50 \text{ mm}^3$	Bergman (2005)
Moisture content of feedstock	50 %	Bergman (2005)
Energy yield target	96.1 %	Bergman (2005)
Production rate	1.45 kg/s	Bergman (2005)
Moisture content of input to reactor	15 %	Bergman (2005)
Thermal conductivity of biomass	$0.13+0.0003(T-273)$ W/mK	R.K. Jalan (1999)
Estimated heat transfer coefficient	200 W/m ² K	Bergman (2005)
Reactor type	Moving bed	Bergman (2005)
Reactor size	Diameter 1.3 m,	Bergman (2005)

	Length 6.7 m	
Feedstock composition	C: 0.472, H: 0.061, O: 0.451, N: 0.003, Ash: 0.013	Bates (2012)
Residence time	1260 sec (21 min)	Bergman (2005)
Temperature of recycled gas	280 °C	Bergman (2005)
Flow rate of recycled gas	1.436 kg/s	Calculated

2.2.2. Modeling approach

■ Reactor model

In order to develop the reactor model for moving bed type, one-dimensional plug-flow reactor was assumed. If the sophisticated modeling technique, such as computational flow dynamics (CFD) simulation, was implemented for this work, it was difficult to optimize the operation variables of process design due to computation time. Also, the CFD simulation is too heavy to develop the process design. For design and optimization of torrefaction process, simple model for torrefaction reactor was used.

■ Process model

The isentropic blower was used in process simulation and the isentropic and mechanical efficiency were assumed 0.7 and 0.8. The discharge pressure of blower was calculated from the pressure drop model described in section 2.2.7. The minimum temperature approach of the heat exchanger of 10 K was used. In the furnace unit of process simulation, torrefied gas and additional natural gas are combusted with air in stoichiometric ratio. The flow rate of natural gas is calculated to satisfy 2 specifications, in which one is the temperature of recycled gas and the other is adiabatic flame temperature of 1000 °C. When the volatiles is used as a fuel, the adiabatic flame temperature should be above 1000 °C for a stable combustion process [3]. The flowrate of natural gas was determined to specify the temperature of recycled gas and adiabatic flame temperature of the furnace.

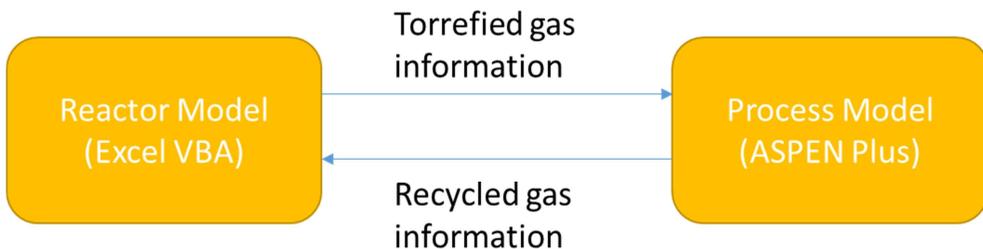
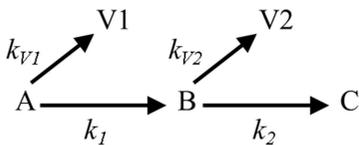


Figure 2-3 Structure of process and reactor model

2.2.3. Solid and volatile evolution kinetics

Bates and Ghoniem suggested a mass-loss kinetic model of the biomass torrefaction process [1]. This model was based on an existing model and validated by experimental data for willow torrefaction by Prins [10]. The Bates and Ghoniem model enables accurate prediction of the solid mass balance during torrefaction under kinetically limited conditions in the absence of transport limitations, and comprises three steps. The first step is analysis of the data on the kinetic mechanism of solid mass loss and volatile composition in torrefaction to determine the appropriate model, which was adapted from Prins [10]. The proposed mechanism, consisting of a two-stage, first-order reaction, showed good agreement with the experimental data. This mechanism is given as follows:



where A is the raw biomass, B is the solid intermediate reaction product, C is the residual solid product, and V1 and V2 represent volatiles. The first-stage reaction is much faster than the second-stage reaction. At any moment in time, the mass of the solid product is described by the sum of the masses of A, B, and C, and the total mass of the volatile is provided by the sum of V1 and V2. This mechanism was

originally proposed by Di Blasi and Lanzetta to describe pure hemicellulose decomposition [16]. The kinetics of this process are described as follows:

$$r_A = \frac{dm_A}{dt} = -(k_1 + k_{V1}) \times m_A \quad (1)$$

$$r_B = \frac{dm_B}{dt} = (k_1) \times m_A - (k_2 + k_{V2}) \times m_B \quad (2)$$

$$r_C = \frac{dm_C}{dt} = (k_2) \times m_B \quad (3)$$

$$r_{V1} = \frac{dm_{V1}}{dt} = (k_{V1}) \times m_A \quad (4)$$

$$r_{V2} = \frac{dm_{V2}}{dt} = (k_{V2}) \times m_B \quad (5)$$

where r_X is reaction rate of pseudo-component (X=A, B, C, V1, V2) in kg s^{-1} , m_X is mass of pseudo-component X in kg, and k_1 , k_2 , k_{V1} , k_{V2} represent Arrhenius kinetic parameters in the mechanism.

The second step is the development of a simplified volatile composition model, based on experiments determining the solid mass-loss kinetics and volatile composition. Prins reported the cumulative yield of nine chemical species generated during willow torrefaction under different process conditions [10]. The produced volatiles (V1 and V2) were modeled as a mixture of nine chemical components. The compositions of V1 and V2 differ: the first-stage reaction primarily yields hemicellulose decomposition products, whereas the volatiles (V2) produced in the second-stage reaction are cellulose decomposition products.

The third step is estimation of the compositions of the solid intermediates and products using the mass conservation conditions. The elemental composition of A is a known model input determined by performing analysis of the product. The solid mass yield Y_S and mass loss Y_{loss} are obtained by integrating Eq. (1) through (5), which were proposed by Bates and Ghoniem, to obtain

$$Y_S = \frac{m_A + m_B + m_C}{m_0} \quad (6)$$

$$Y_{loss} = \frac{m_{V1} + m_{V2}}{m_0} = 1 - Y_S \quad (7)$$

where m_0 is the initial dry mass of the solid. The instantaneous fractional yield of the solid phase can be determined by the formation rate of the product divided by the decomposition rate of the reactant [2].

Table 2-2 calculated volatiles composition and solid contents

Chemical component	Composition (%)	
	V1	V2
Acetic acid	14.8	16.1
Water	48.1	7.6
Formic acid	5.3	5.1
Methanol	4.2	30.1
Lactic acid	1.3	31.3
Furfural	1.1	0

Hydroxyl acetone	0.6	9.7
Carbon dioxide	20.4	0
Carbon monoxide	4.2	0.1
Sum	100	100
<hr/>		
Carbon	18	36
Hydrogen	7	9
Oxygen	75	55
Nitrogen	0	0
Ash	0	0
Sum	100	100
<hr/>		

2.2.4. Thermochemical properties

An accurate estimate of the reaction enthalpy is important for process design and control of the reactor. It is therefore necessary to quantify the thermal energy input required during torrefaction to assess the overall process efficiency. In particular, the feasibility of autothermal torrefaction, originally described by Bergman et al.,[3] in which the volatile products released during torrefaction are combusted to provide heat for the process itself, depends on accurate estimations of their composition and heating value. Bates and Ghoniem described a model for the energy balance and thermochemistry of torrefaction, linked to the previous kinetic model discussed above [2]. This thermochemical model can be used to estimate temperature-

dependent properties such as enthalpy, energy yield, and heat release, based on the detailed composition results obtained from the kinetic model. The total enthalpy at temperature T is given by sum of the formation enthalpy and sensible enthalpy:

$$H_i(T) = H_{f,i}^0 + \int_{T_0}^T c_{p,i}(T) dT \quad (8)$$

where $H_{f,i}^0$ is the standard heat of formation of species i in J kg^{-1} , $c_{p,i}$ is the specific heat capacity of component i in $\text{J kg}^{-1} \text{K}^{-1}$, T is the reaction temperature in Kelvin, and T_0 is the standard temperature (298.15 K). The energy yield is the sum of the solid product and volatile product energy yields. The solid product energy yield is defined as

$$\eta_s = Y_s \left(\frac{HHV_f}{HHV_0} \right) \quad (9)$$

where HHV_f is the heating value of the torrefied solid product in J kg^{-1} and HHV_0 is the initial heating value of the solid feed in J kg^{-1} . The energy yield of the total volatiles is defined as

$$\eta_{V_{tot}} = Y_{loss} \left(\frac{HHV_{V_{tot}}}{HHV_0} \right) \quad (10)$$

where $HHV_{V_{tot}}$ is the average heating value of all volatiles in J kg^{-1} .

The heat release rate for the overall reaction and that for each stage are calculated from the thermochemical properties and reaction rates. The rate of heat release at temperature T is given as

$$\frac{dq_r}{dt} + \sum_{X=1}^5 \frac{d(H_X m_X)}{dt} = 0 \quad (11)$$

where q_r is the cumulative heat release in J, HX ($X = 1, 2, 3, 4, 5$) is the enthalpy of component X at temperature T , and m_X ($X = 1, 2, 3, 4, 5$) represents the mass of component X . The X values 1–5 refer to the pseudo-components A, B, C, V1, and V2, respectively.

2.2.5. Solid drying rate

The dryer in the diagram in Figure 2-1 is used to reduce the moisture content of the biomass from 50% to 15%. The remaining moisture is removed during torrefaction to obtain a final moisture content of 1–3%. The water included in the biomass feedstock in the torrefaction reactor is mainly evaporated in the initial torrefaction stage. The torrefaction reactor model must therefore include a drying rate model for the moisture content of the biomass feedstock. During drying of the solid biomass, it is assumed that the heat energy is used only to evaporate the moisture content of the biomass and not to increase the temperature of the solid. The drying rate is thus expressed as

$$r_d = \frac{hA(T_G - T_S)}{\lambda_w} \quad (12)$$

where r_d represents the drying rate in kg s^{-1} , h is the heat transfer coefficient in $\text{W m}^{-2} \text{K}^{-1}$, A is heat transfer area in m^2 , T_S and T_G represent the temperatures of the solid and the gas, respectively, and λ_w is the enthalpy of water evaporation in J kg^{-1} .

2.2.6. Heat transfer model

The reasonable heat transfer model is needed to predict temperature profile inside torrefaction reactor. During the torrefaction of biomass chips, heat is transferred by conduction inside the solid chip and convection between solid and gas phase. R.K. Jalan and V.K. Srivastava represented the heat transfer model to describe the pyrolysis of a single biomass cylindrical pellet [17]. They assumed that heat transfer inside the solid is by conduction and occurs only in the radial direction. The generalized form of the heat transfer equation is represented in Eq. (13) and boundary conditions of that equation are represented in Eq. (14), (15).

$$k_s \left[\frac{1}{r} \frac{dT_s}{dr} + \frac{d^2 T_s}{dr^2} \right] + (-H_r) \frac{d(-\rho_s)}{dt} = \frac{d(C_{p,s} \rho_s T_s)}{dt} \quad (13)$$

Boundary conditions

$$r = 0, \frac{dT_s}{dr} = 0 \quad (14)$$

$$r = R, (-k_s \frac{dT_s}{dr}) = h(T_G - T_s) \quad (15)$$

The k_s represents conductivity of biomass chip in $W m^{-1} K^{-1}$, r is radius of cylindrical pellet in m, H_r is the enthalpy of torrefaction in $J kg^{-1}$, $C_{p,s}$ is heat capacity of biomass in $J kg^{-1} K^{-1}$ and ρ_s is density of biomass $kg m^{-3}$.

2.2.7. Heat and mass balance

A moving-bed reactor was chosen for torrefaction in this chapter; the details are given in Table 2-1. The geometry of the TORSPYD® developed by Thermya, which is a commercial moving-bed reactor, is shown in Figure 2-3 [18]. The column contains packed wood chip particles undergoing torrefaction and a convective gas surrounding the particle bed. The biomass continuously moves through the reactor, exchanging heat and volatiles with the surrounding gas phase. The gas flow is in the opposite direction to that of the biomass; this is called counter-current flow. The moving-bed reactor is compact and has a high heat transfer coefficient. These features mean that the residence time is small under the optimized conditions. However, because the reactor has a high fill percentage, the pressure drop in the moving-bed reactor is more severe than in other types of reactor. The operating cost of the blower needs to be considered in optimizing the process design when a moving-bed reactor is chosen [3].

The torrefaction reactor is described by a one-dimensional reactor model. To simplify this model, it is assumed that the biomass has uniformly sized particles, that all particles move with a constant velocity in the reactor, and that the recycled gas flows with perfect mixing. The torrefaction reactor model was developed on the basis of several balance equations. The mass balances of the solid and gas phases are described in the kinetic model for all components during torrefaction. The heat balances of the solid and gas phases are derived by combining the thermochemical model and the drying rate model with the heat transfer properties. The balance equations used are given below.

Mass balance of solid phase:

$$0 = -u_s(z) \frac{\partial m_{S,i}}{\partial z} + r_i \quad (16)$$

Mass balance of gas phase:

$$0 = -u_G(z) \frac{\partial m_{G,i}}{\partial z} - r_i \quad (17)$$

Heat balance of solid phase:

$$0 = -u_s(z) \frac{\partial T_S}{\partial z} \sum m_{S,i} c_{p,S,i} + hA(T_G - T_S) + \sum r_i(-H_r) + r_d \lambda_w \quad (15)$$

Heat balance of gas phase:

$$0 = -u_G(z) \frac{\partial T_G}{\partial z} \sum m_{G,i} c_{p,G,i} - hA(T_G - T_S) \quad (18)$$

where u is the velocity in m s^{-1} , z is the axial coordinate in m , and subscript G is gas phase and S is solid phase. The differential equations were solved using Excel VBA.

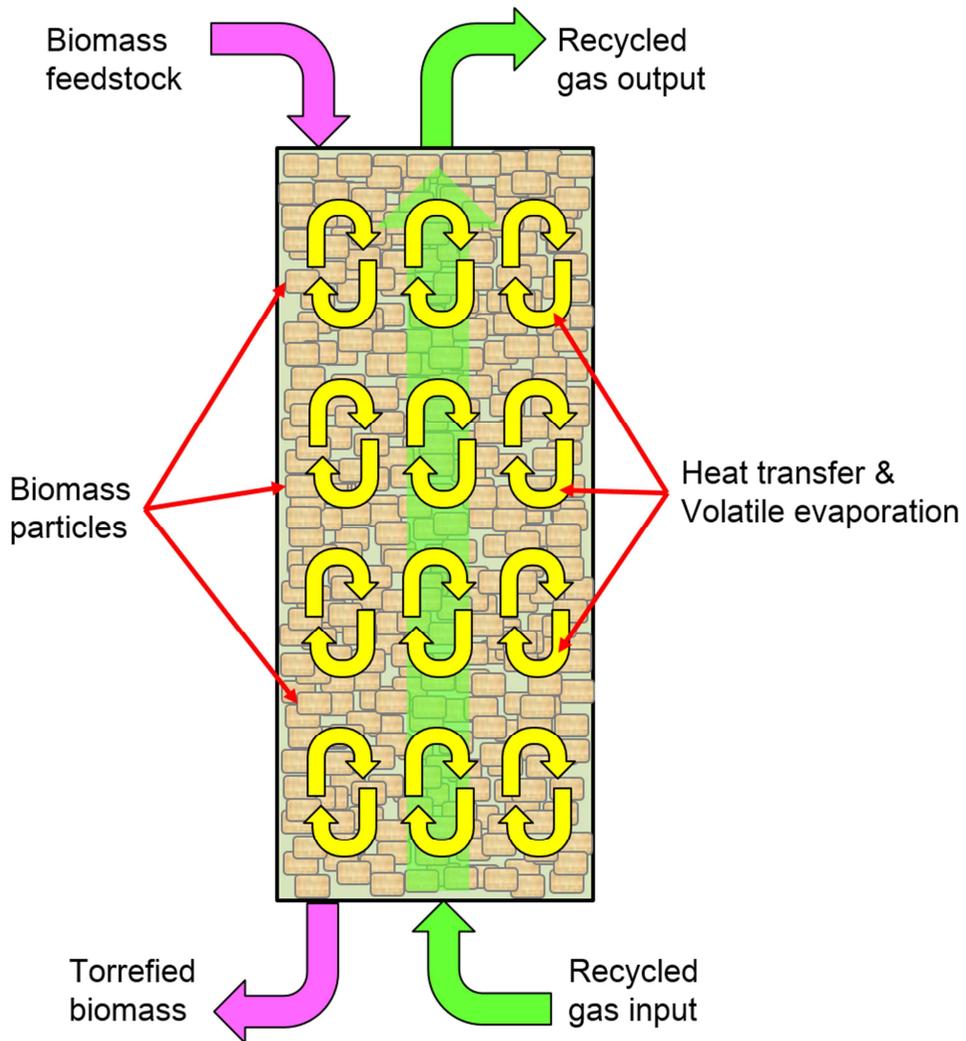


Figure 2-4 Scheme of torrefaction reactor

2.2.8. Pressure drop

The pressure drop in a moving-bed reactor is severe, as explained in previous section. The calculated pressure drop is required to estimate the blower duty in the biomass process scheme. In this chapter, the pressure drop was calculated using the

Ergun equation. The void fraction was calculated using the data in Bergman's case study, and the effective diameter (d) was predicted using an empirical equation from the work by Li and Ma [19]. The viscosity and density of the recycled gas were obtained using Aspen plus, which is a commercial chemical process simulation tool, using the Peng–Robinson physical property. The mass velocity varied with the flow rate of the recycled gas and the residence time.

$$-\frac{\Delta p}{z} = 150 \frac{(1-\varepsilon)^2 \mu}{d^2 \varepsilon^3} J + 1.75 \frac{(1-\varepsilon) \rho}{d \varepsilon^3} J^2 \quad (20)$$

where Δp is the pressure drop in kPa, ε is the porosity of the reactor, μ is the viscosity of gas phase is N s m^{-2} , d is the effective diameter in m, J is the superficial gas velocity, $\text{kg s}^{-1} \text{m}^{-2}$, ρ is the density of gas phase in kg m^{-3} .

2.2.9. Validation of reactor model

Since the developed reactor model is based on assumption that the reactor is heated-up by a heat carrier gas without electric heater for application to commercial plant scale, experimental data obtained from reactor with only heat carrier gas are required for model validation. However, experimental data for torrefaction with only heat carrier gas hasn't been reported until now. As previously explained, the developed reactor model is based on differential equations for fixed-bed reactor model integrating with various models for particular behavior such as heat and mass transfer. In the particular models, solid and volatiles evolution model,

thermochemistry model and heat transfer model inside the reactor mostly determine the accuracy of the properties of solid and volatiles when comparing with real plant data. Evolution model of volatiles used in developed reactor model was validated by comparing with experimental data by M. Pach et al. [20] for the cumulative yield of non-condensable and condensable volatiles versus solid mass loss in the Bates's work [1]. The volatiles evolution model satisfied the distribution of condensable and non-condensable product of experimental data. The decomposition model of solid product was validated comparing with experimental data from Prins [10], Arias et al. [21] and Medic et al. [8] of ultimate analysis of torrefied biomass [1]. The thermochemistry model was validated by comparing with measured exothermicity of pure hemicellulose from experiments by Khezami et al. [22] and Yang et al. [23] in the Bates's study [2]. The heat transfer model used in this chapter was validated with experimental results for biomass pyrolysis obtained by Pyle and Zaror [24]. The model prediction showed excellent agreement with the experimental data in the R. K. Jalan's work [17].

Table 2-3 comparison results to the experimental data [17]

r/r_0	Temp. Exp.	Model (11min)	Pyle- Zaror	Temp. Exp.	Model (6min)	Pyle- Zaror	Temp. Exp.	Model (4min)	Pyle- Zaror
0	643	612	605	563	535	520	483	469	473

0.3	643	614	605	583	542	530	493	480	485
0.5	643	617	605	590	554	540	510	499	495
0.7	643	622	600	600	570	555	525	524	520
0.9	643	620	595	610	590	570	580	556	550

2.3. Results and discussion

The reactor model developed in this chapter can predict the temperature profiles of the gas and solid phases in the reactor. One feature of the reactor model is that it can be applied to practical operating conditions, including the recycled gas temperature and flow rate, and residence time, to calculate the temperature profile. This allows us to estimate not only the product quality but also to obtain information on the factors that influence the process units, such as the composition and flow rate of the volatiles and the energy required by the torrefaction reactor. Such predictions are required for the design of industrial processes, because industrial torrefaction plants use recycled gas as a heat carrier instead of using an electric heater.

The temperature profile and solid yield with biomass residence time for the base case conditions were obtained using the developed reactor model. The results are shown in Figure 2-5. To satisfy the energy yield target of 96.1%, the required flow rate of the recycled gas was found to be 1.434 kg/s, using the developed model. Because the residence time on the x-axis can convert to the reactor length, with the assumption of constant biomass velocity, the direction of x-axis was regarded as a

direction for solid flow with reactor length (from 0 m to 6.7 m). However, residence time was used for the x-axis in this chapter because it is commonly used as a manipulated variable of torrefaction status. The direction for biomass flow is with increasing of residence time and the direction for recycled gas flow is with decreasing of residence time, as shown in Figure 2-5. The blue curve represents the temperature of the recycled gas and the red curve represents the temperature of the solid. Because the biomass feedstock is dried in the initial part of the reactor, the solid temperature is constant in the initial part and then increases with residence time. The temperature difference between the solid and the gas is not large because of the high heat transfer coefficient in a moving-bed reactor. Because the flow rate of the recycled gas is not large enough, torrefaction occurs under non-isothermal conditions. The solid temperature gradually increases toward the end of the reactor. The energy yield (black curve) reaches the target of 96.1%.

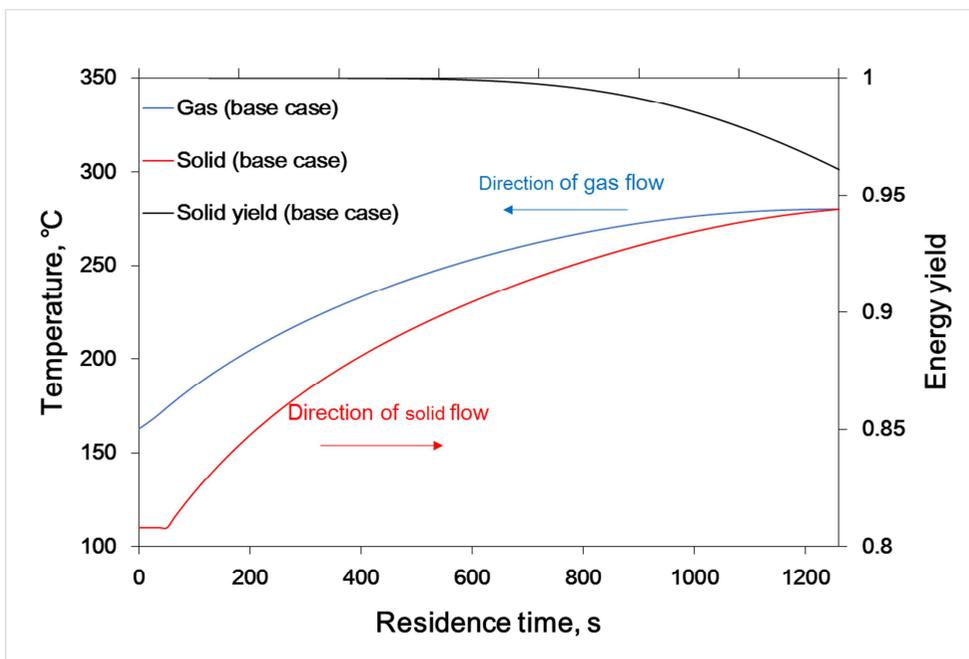


Figure 2-5 temperature and energy yield profile of base case

Figure 2-6 shows composition profiles of solid product during torrefaction. Torrefaction degree is progressed with residence time of solid in reactor. Although oxygen component, which has no heating value, is steadily reduced, carbon and hydrogen composition is mostly retained.

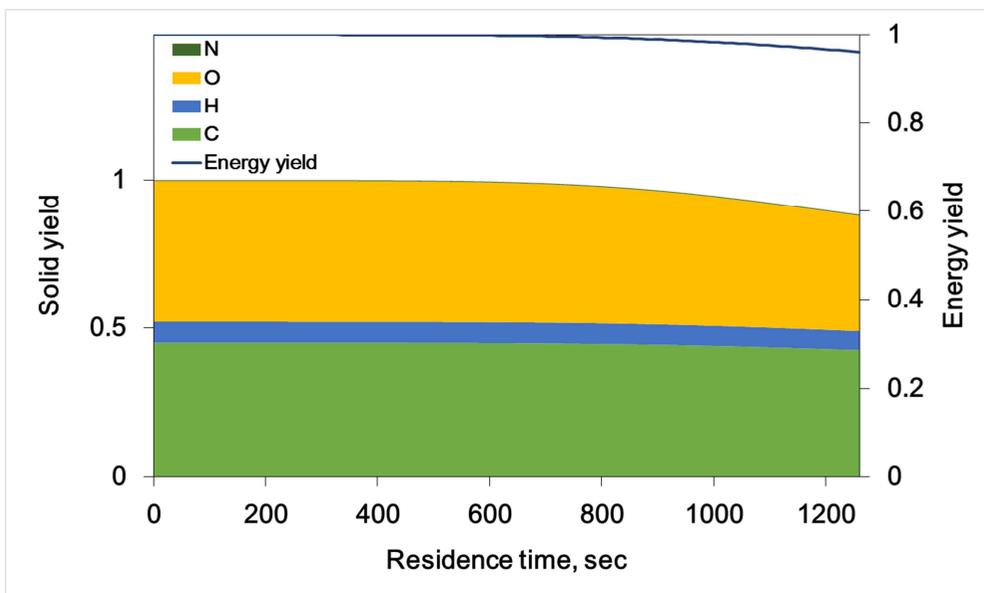


Figure 2-6 simulation results of solid yield composition

Figure 2-7 represents composition of pseudo components of solid product during the torrefaction. In this torrefaction conditions (final energy yield is 0.961), mild torrefaction is progressed. Char of C is not presented in this conditions. The reaction to convert into B mainly occurs. At the end of torrefaction, composition of B is mostly half of total mass flow rate.

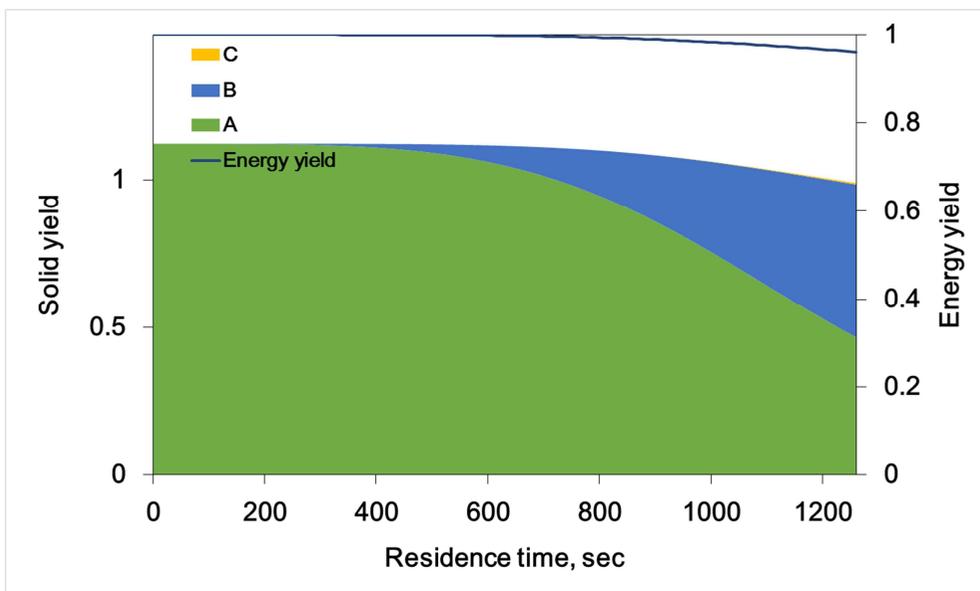


Figure 2-7 simulation results of solid pseudo component

Figure 2-8 shows generation rate of volatiles with residence time during the torrefaction. Because the temperature of solid at bottom of reactor is larger than that at top of reactor, generation rate of all components are more fast at the bottom of the reactor. The water, carbon dioxide and carbon monoxide are mainly generated during torrefaction.

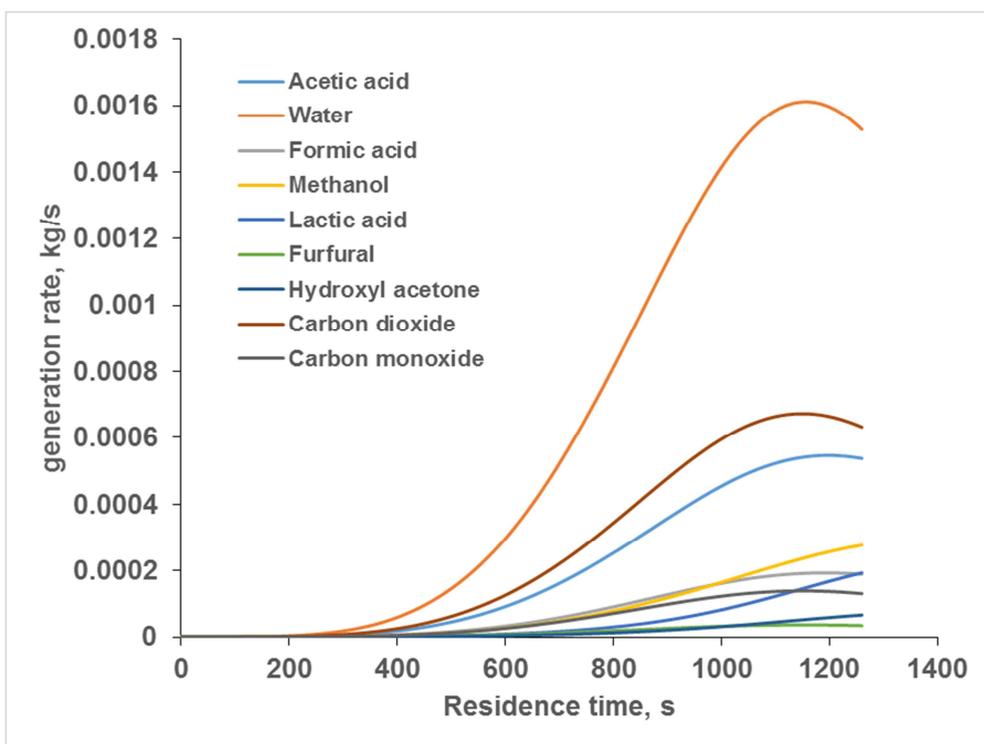


Figure 2-8 generation rate of volatiles components

Figure 2-9 shows the flow rate of gas stream in the reactor during torrefaction. Flow direction of gas stream is opposed to solid flow. Since water contents drying occurs at top of the reactor, water flow rate is drastically increased in early part of reactor. Because the flow rate of recycled gas is enough larger than generation rate of volatiles, the composition of gas stream is maintained.

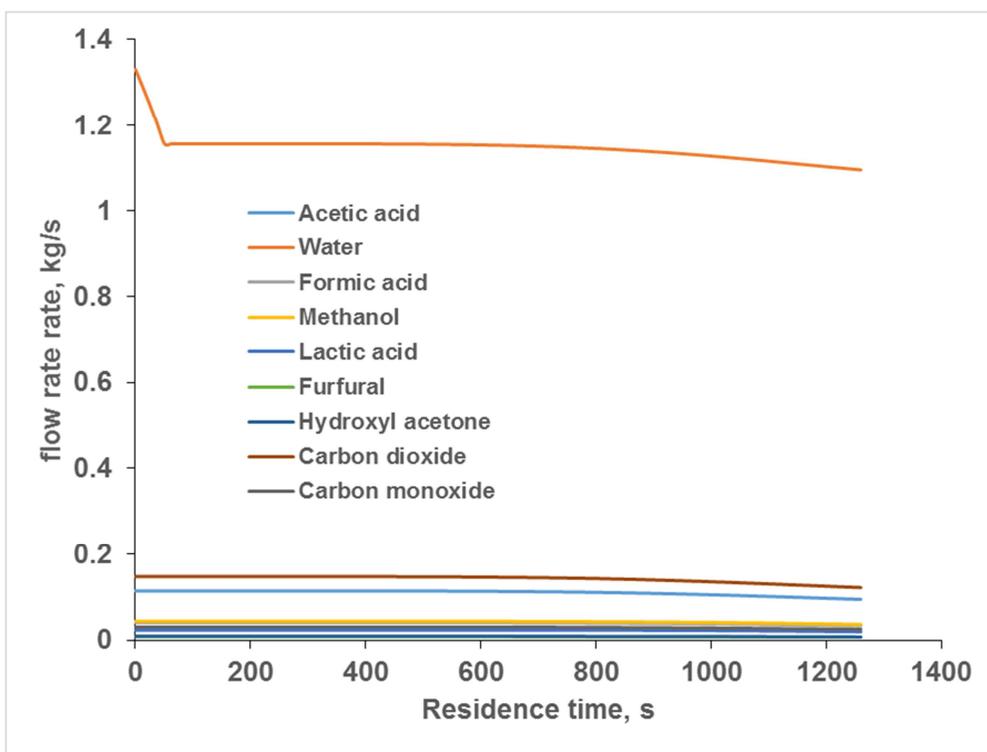


Figure 2-9 flow rate of volatiles components

Table 2-4 shows the gas stream information of torrefaction process model which was computed from the results of reactor model. The stream number in Table 2-4 coincide the number in Figure 2-1.

Table 2-4 Gas stream information of torrefaction process model for base case

	1	2	3	4	5	6	7	8
Temperature, °C	162.7	162.7	200.4	280.0	25.0	25.0	948.1	766.5
Pressure, bar	1.0	1.0	1.4	1.4	1.0	1.0	1.0	1.0
Flow rate, kg/s	0.310	1.436	1.436	1.436	0.004	0.377	0.691	0.691

Composition**(mass basis)**

N2					0.790	0.432	0.432
O2					0.210	0.024	0.024
CH4					1.000		
Acetic acid	0.065	0.065	0.065	0.065			
Water	0.762	0.762	0.762	0.762		0.394	0.394
Formic acid	0.023	0.023	0.023	0.023			
Methanol	0.025	0.025	0.025	0.025			
Lactic acid	0.013	0.013	0.013	0.013			
Furfural	0.005	0.005	0.005	0.005			
Hydroxy	0.005	0.005	0.005	0.005			
acetone							
CO2	0.084	0.084	0.084	0.084		0.150	0.150
CO	0.017	0.017	0.017	0.017			

In this chapter, a one-dimensional reactor model that considered the practical thermal conditions for biomass torrefaction was developed, based on an existing kinetic model and the thermochemistry of the reaction. The reactor conditions in the base case were generated using the developed model based on Bergman's case study. The reactor model developed in this chapter can predict the temperature profiles of the gas and solid phases in the reactor. One feature of the reactor model is that it can be applied to practical operating conditions, including the recycled gas temperature and flow rate, and residence time, to calculate the temperature profile. This allows

us to estimate not only the product quality but also to obtain information on the factors that influence the process units, such as the composition and flow rate of the volatile gas and the energy required by the torrefaction reactor. Such predictions are required for the design of industrial processes, because industrial torrefaction plants use recycled gas as a heat carrier instead of using an electric heater. Although the reactor model is based on willow experiments performed by Prins [10] , it will be useful for the design of general torrefaction processes.

CHAPTER 3 : Parametric study of operating conditions in the biomass torrefaction

3.1. Introduction

In order to optimize the process design and operating variables, influence of manipulated variables in the process on parameters which can determine the cost of whole process should be investigated. Studies of biomass torrefaction have mainly focused on experimental research into the effects of various operating parameters such as the temperature, residence time, feed stock, moisture contents and particle size [8, 9]. Furthermore, most studies on torrefaction have determined the reactor operating conditions such as temperature and residence time based only on product quality [3, 25, 26]. The reactor operating conditions affect the whole process design as well as the product quality. The utility and capital costs of the entire process are

determined by the operating conditions. The torrefaction reactor operating conditions need to be determined considering the effects on the process design. Thus, it is necessary to develop the process model of biomass torrefaction and analyze the effect of operating conditions on process parameters.

In this chapter, the influence of torrefaction residence time, flow rate and temperature of recycled gas on the properties of biomass as well as process parameters was investigated. Quantitative and qualitative data from the sensitivity analysis of operating conditions were used to investigate parameters in the torrefaction process. Developed process model is utilized to obtain the data of heat and mass flow in the process. Results from this work will provide knowledge which is difficult to be obtained in experimental study.

3.2. Sensitivity analysis of operating variables

3.2.1. Manipulated variables

The torrefaction progresses more intensely with increasing temperature and flow rate of the recycled gas and the residence time. However, a higher recycled gas temperature and flow rate and a longer residence time increase the operating and capital costs and affect the process efficiency. The recycled gas temperature is increased by increasing the natural gas (NG) flow rate. A higher recycled gas flow rate causes a higher pressure drop and increases the blower duty. A long residence

time needs the torrefaction reactor to be lengthened, so the equipment is more expensive. The effect of operating variables of the torrefaction reactor therefore needs to be analyzed considering parameters of the torrefaction process. Thus, three manipulated variables of the torrefaction reactor were chosen for sensitivity analysis which was conducted using torrefaction process model. Table 3-1 shows the comparison of experimental and practical operation variables.

Table 3-1 comparison of experimental and practical operation variables

	Experimental operation variable	Practical operation variable
1	Reactor temperature	Temperature of recycled gas
2	Solid residence time	Flow rate of recycled gas
3		Solid residence time

3.2.2. Process parameters

The parameters, which are affected by manipulated variables determined previous section, are chosen to measure the effects on the total cost. Table 3-2 shows the qualitative relations between operating conditions and process parameters. Product properties are influenced from all operating conditions. Also, natural gas consumption and heating value of recycled gas are influenced from all operating conditions. Pressure drop of gas stream in reactor and blower duty to recover the pressure drop are influenced from flow rate of recycled gas and residence time. The

reactor length is only influenced from residence time.

Table 3-2 influence of operating conditions on properties of product and process parameters

	Temperature of recycled gas	Flow rate of recycled gas	Residence time
Temperature	O	O	O
profile of solid			
Energy yield	O	O	O
Solid yield	O	O	O
HHV of solid	O	O	O
Pressure drop	X	O	O
Blower duty	X	O	O
NG consumption	O	O	O
HHV of recycled gas	O	O	O
Reactor length	X	X	O

3.2.3. Feed conditions

For analysis of effect for feed stock conditions, the moisture contents and temperature of solid feedstock were selected as variables. Moisture contents of biomass was varied with ambient temperature, humidity and exposure time.

Because, at the top of reactor moisture of solid is reduced, it is able to influence the temperature profile of reactor and process parameters. Outlet temperature of pre-dryer is specified as 110°C in the Bergman's case study. However heat energy of solid is reduced for heat loss of exposure to ambient. Hence the temperature of feed stock is decreased and able to require more heat energy in the reactor.

3.3. Results and discussion

3.3.1. Effect of temperature of the recycled gas on the process parameters

The recycled gas temperature, which is the first of the manipulated variables, affects the driving force of heat transfer between the solid and the recycled gas in the reactor. The consumption of natural gas is influenced by the temperature of the recycled gas. Figure 3-1 shows the results of the sensitivity analysis of temperature profile and energy yield profile with varying the temperature of the recycled gas. The solid lines represent temperature of biomass during torrefaction, and the broken line represents energy yield of biomass with residence time in reactor. The blue line represents the temperature and energy yield profiles for the base case. The temperature profiles of solid start from same temperature, and increase and reach to different temperature depending on the temperature of recycled gas. The energy yield decreases with increasing the recycled gas temperature, since the reaction rate

increases with the solid temperature as represented in most literatures for torrefaction experiments. After torrefaction, the final energy and solid yield and HHV of torrefied biomass are shown in Figure 3-2. The energy and solid yield decrease with increase in the temperature of recycled gas due to intensity of torrefaction progress, and the HHV of torrefied biomass increase with the intensity of torrefaction progress. The pressure drop of recycled gas in reactor slightly increase in Figure 3-3. The pressure drop is influenced by the velocity of gas stream and reactor length. Since the flow rate of torrefied gas, which moves from solid to gas phase, increase with the temperature of recycled gas, the velocity of gas flow increase with the pressure drop and blower duty. Figure 3-4 shows the HHV of recycled gas and NG consumption with varying the flow rate of the recycled gas. The torrefied gas mainly consist of water on account of the moisture contents in raw biomass. So the contents to be used as fuel in the torrefied gas increase with progress of torrefaction. The HHV of recycled gas increases with the temperature of recycled gas. Since the NG is used to make up for the torrefied gas for combustion, the NG consumption decrease with increase in the HHV of recycled gas.

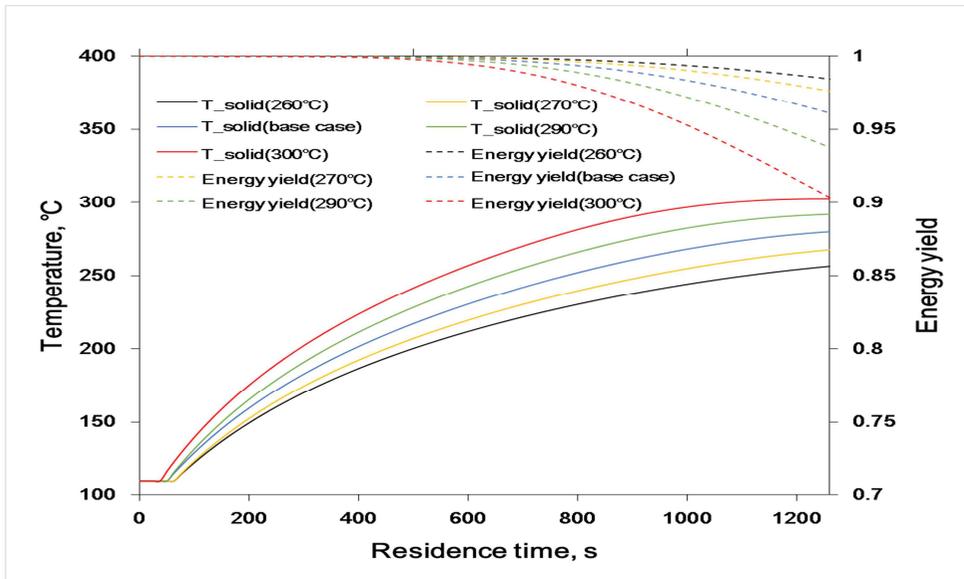


Figure 3-1 Sensitivity analysis of temperatures and energy yield profile with varying the flow rate of the recycled gas.

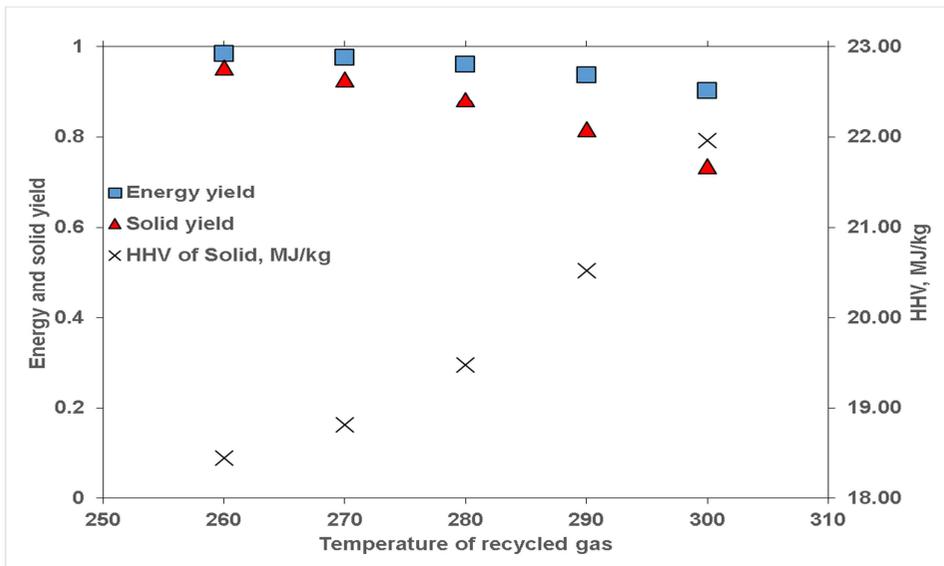


Figure 3-2 Sensitivity analysis of energy yield, solid yield and HHV of solid with varying the flow rate of the recycled gas.

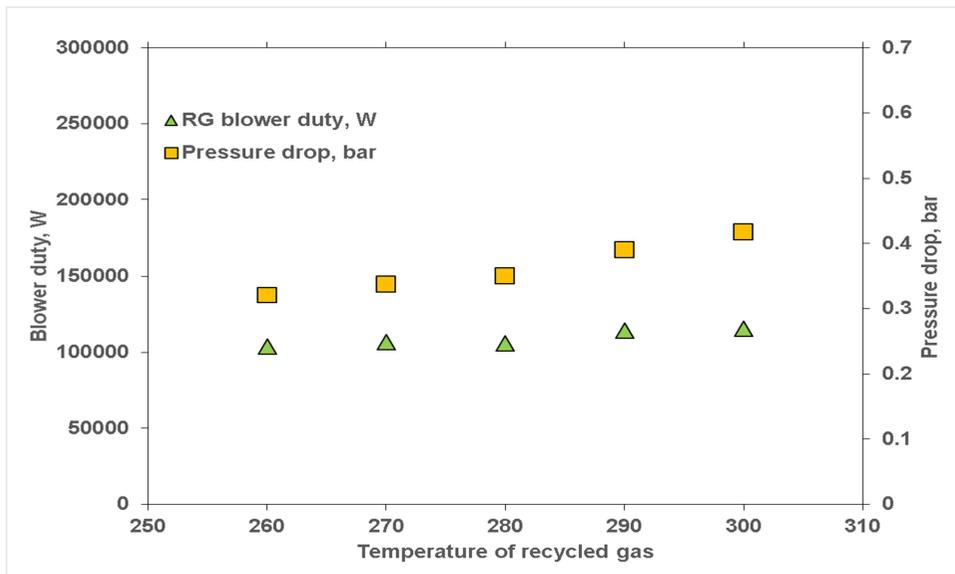


Figure 3-3 Sensitivity analysis of pressure drop and blower duty with varying the flow rate of the recycled gas.

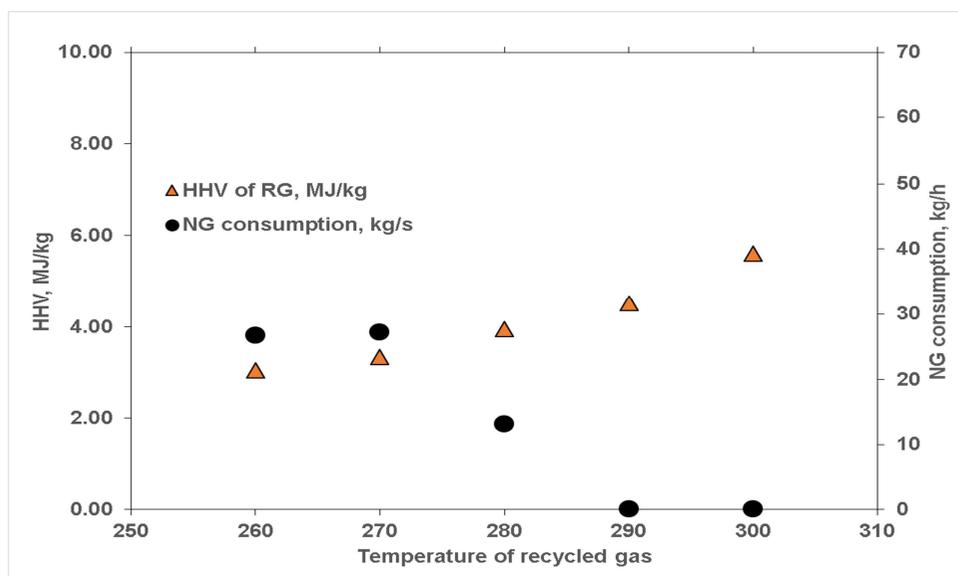


Figure 3-4 Sensitivity analysis of HHV of recycled gas and NG consumption with varying the flow rate of the recycled gas.

3.3.2. Effect of flow rate of the recycled gas on the process parameters

An increase in the flow rate of the recycled gas, which is the second manipulated variable, increases the amount of heat energy in the recycled gas, so the energy capacity of the recycled gas increases. However, in aspect of the torrefaction process, it increases blower duty, which is proportional to the flow rate of the stream through the blower. The flow rate of the recycled gas has additional effects on the blower duty. This manipulated variable increases the gas flow velocity in the reactor, which increases the pressure drop in the reactor. The increase in blower

duty is therefore more than proportional to the recycled gas flow rate. Moreover, to supply the heat energy for a higher recycled gas flow rate, more natural gas needs to be consumed. Figure 3-5 shows the results of the sensitivity analysis for the recycled gas flow rate. The blue curve represents the base case. When a higher flow rate of the recycled gas is used, on the red and green curves, the energy yield lowers. Conversely, when a lower flow rate of the recycled gas is used, i.e., the yellow and black curve, the temperatures of solid doesn't reach to the temperature of recycled gas (280 °C) and torrefaction is barely proceeded. The final energy and solid yield and HHV of torrefied biomass are described in Figure 3-6. As explained in section 3.2, the energy and solid yield decrease with increase in the flow rate of recycled gas, and HHV of torrefied biomass increase with the intensity of torrefaction progress. Figure 3-7 represents the pressure drop and blower duty with varying the flow rate of the recycled gas. The pressure drop of the recycled gas in reactor increase more than that with variations of the temperature of recycled gas. As previously explained, the pressure drop is influenced by the velocity of gas stream. Since the increase in the flow rate of recycled gas causes increase in the velocity, the pressure drop is increased. Also, because the handling capacity of blower increase with the flow rate of recycled gas, the blower duty increases more than the curve of pressure drop. The HHV of recycled gas increases with the flow rate of recycled gas due to progress of torrefaction as shown in Figure 3-8. The NG consumption decreases with the increase in the flow rate of recycled gas.

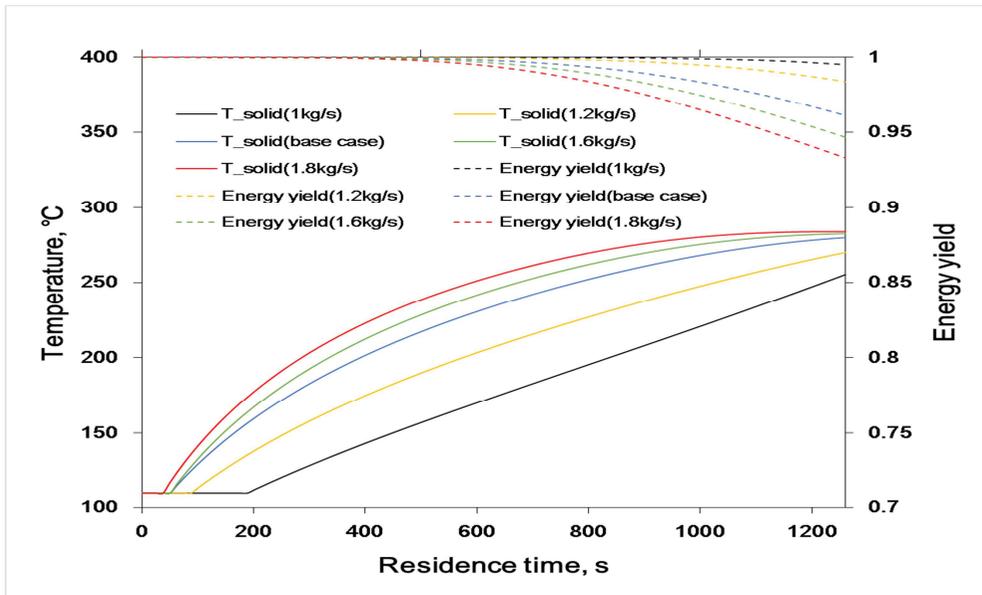


Figure 3-5 Sensitivity analysis of temperatures and energy yield profile with varying the temperature of recycled gas

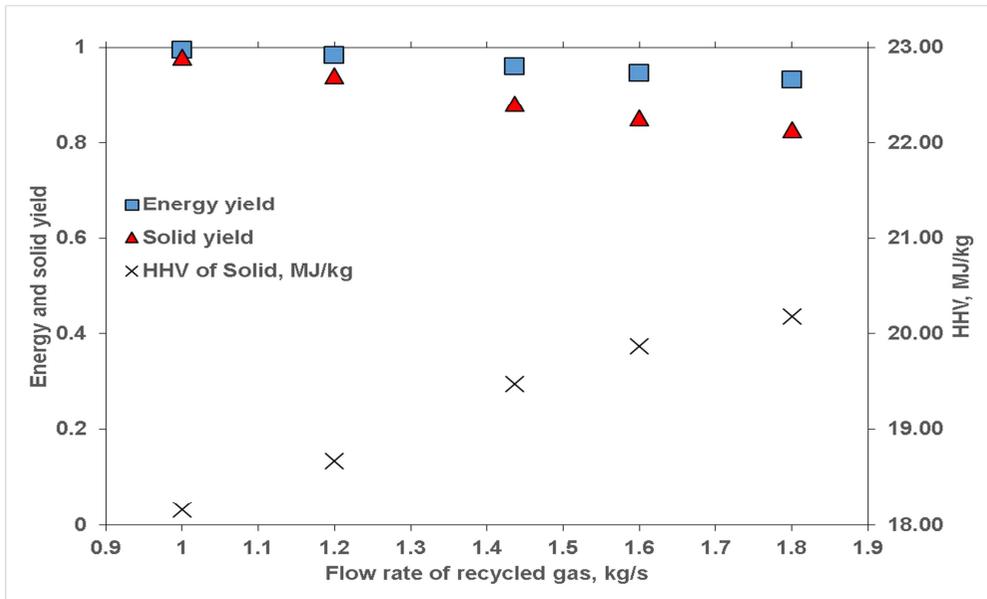


Figure 3-6 Sensitivity analysis of energy yield, solid yield and HHV of solid with varying the temperature of recycled gas

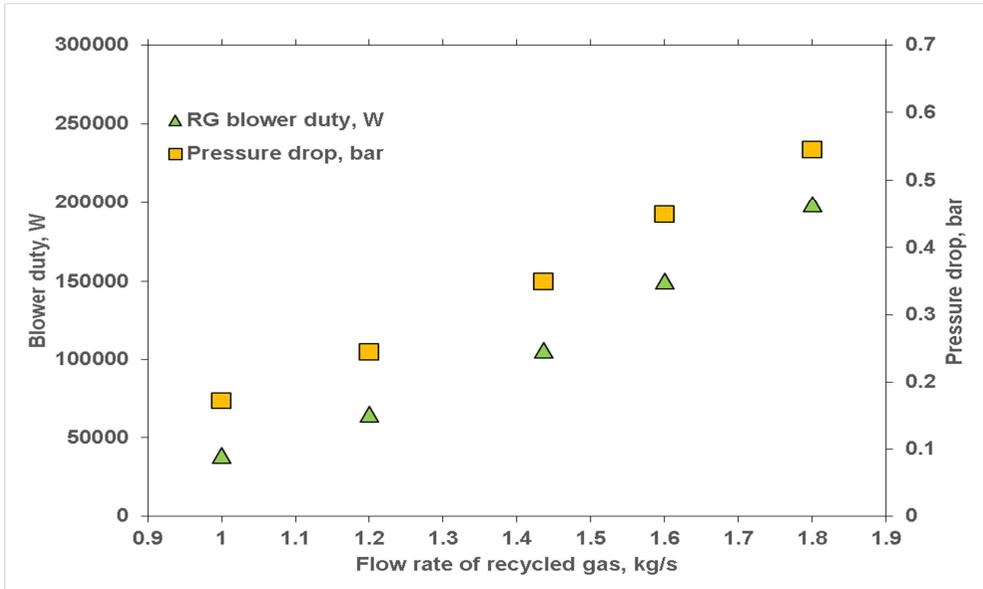


Figure 3-7 Sensitivity analysis of pressure drop and blower duty with varying the temperature of recycled gas

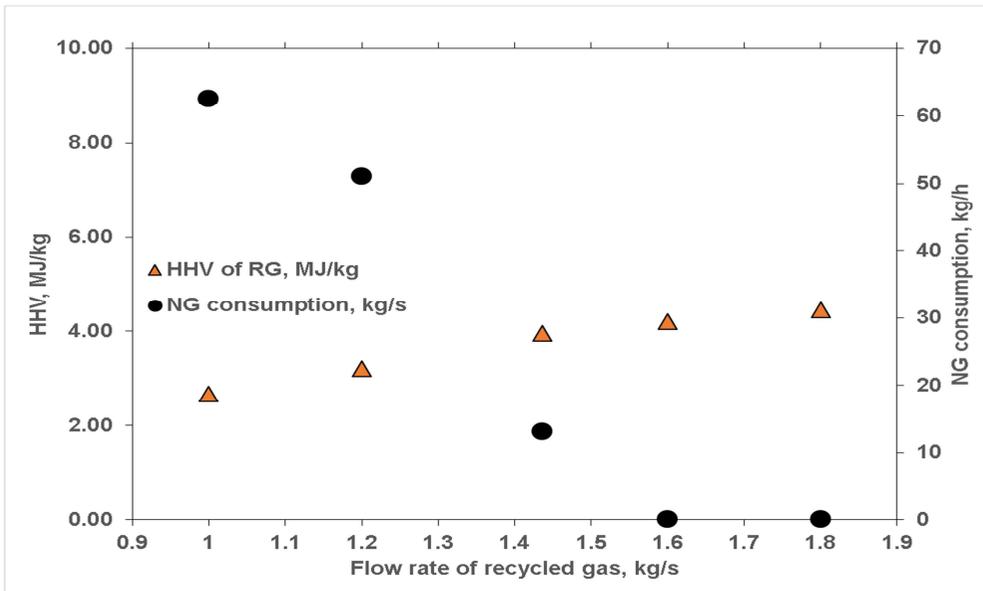


Figure 3-8 Sensitivity analysis of HHV of recycled gas and NG consumption with varying the temperature of recycled gas

3.3.3. Effect of residence time on the process parameters

The last manipulated variable in this sensitivity analysis is the residence time. Most torrefaction studies also consider the residence time as an operating variable. The torrefaction progress increases with the residence time. An increased residence time requires a larger reactor, which increases the reactor cost and affects the process economics. Moreover, because an increase in the residence time causes a decrease in the gas flow velocity and an increase in the reactor length, this operating variable affects the blower duty. Figure 3-9 shows the results of the sensitivity analysis for the residence time. The yellow curve represents the base case. Energy yield of torrefied biomass decrease with increase in the residence time. However, effects of residence time are lower than those of the temperature and flow rate of recycled gas, as also shown in Figure 3-10. Because the flow rate of recycled gas is not enough, the temperature of gas phase starts to drop from the initial temperature of recycled gas (280 °C) in temperature profile and the isothermal section mostly not exists along the reactor. Thus, the torrefaction temperature section (more than 250 °C) is lightly influenced by residence time, and the progress of torrefaction is also lightly varied. If the flow rate of recycled gas is higher in the base case, the energy and solid yield would be seriously affected. Figure 3-11 represents the pressure drop and blower duty with varying the flow rate of the recycled gas. Note that because the

residence time is a term for solid, the residence time does not affect to the velocity of gas flow but only the reactor length. Since the residence time of solid increase with the reactor length, the pressure drop and blower duty increase with the residence time. The HHV of recycled gas slightly increases with the flow rate of recycled gas due to progress of torrefaction as shown in Figure 3-12. The NG consumption slightly decreases with the increase in the flow rate of recycled gas.

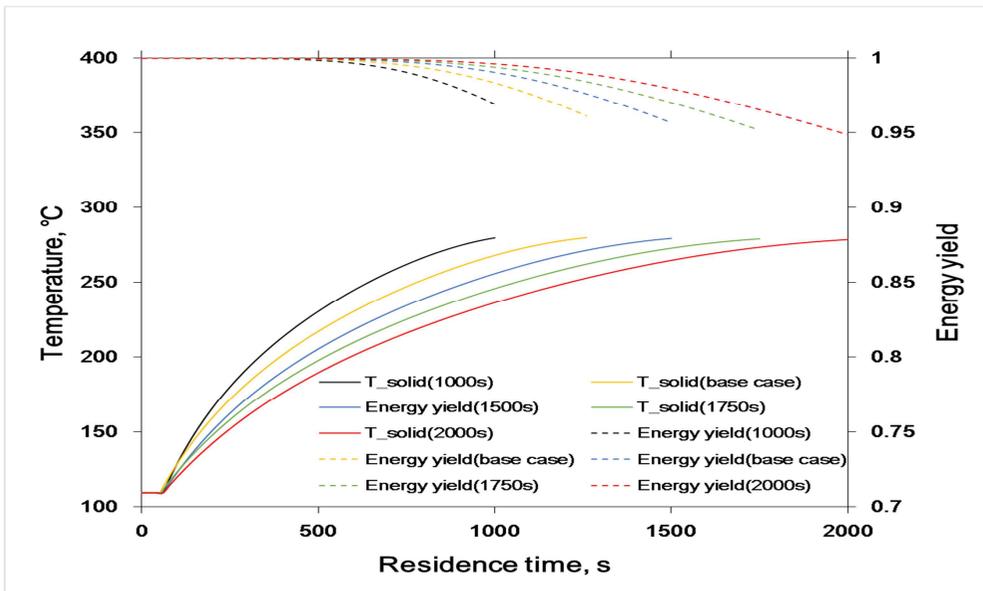


Figure 3-9 Sensitivity analysis of temperatures and energy yield profile with varying the residence time

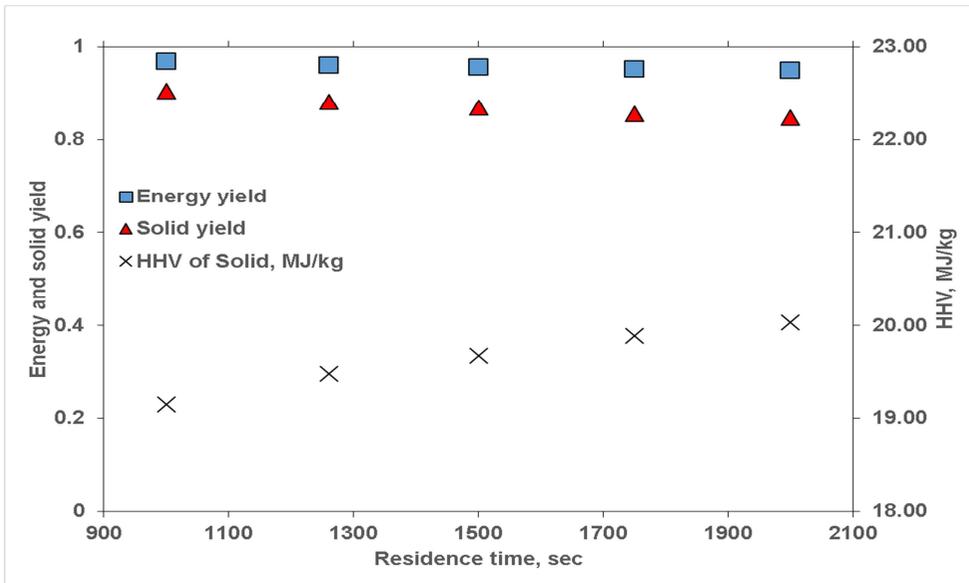


Figure 3-10 Sensitivity analysis of energy yield, solid yield and HHV of solid with varying the residence time

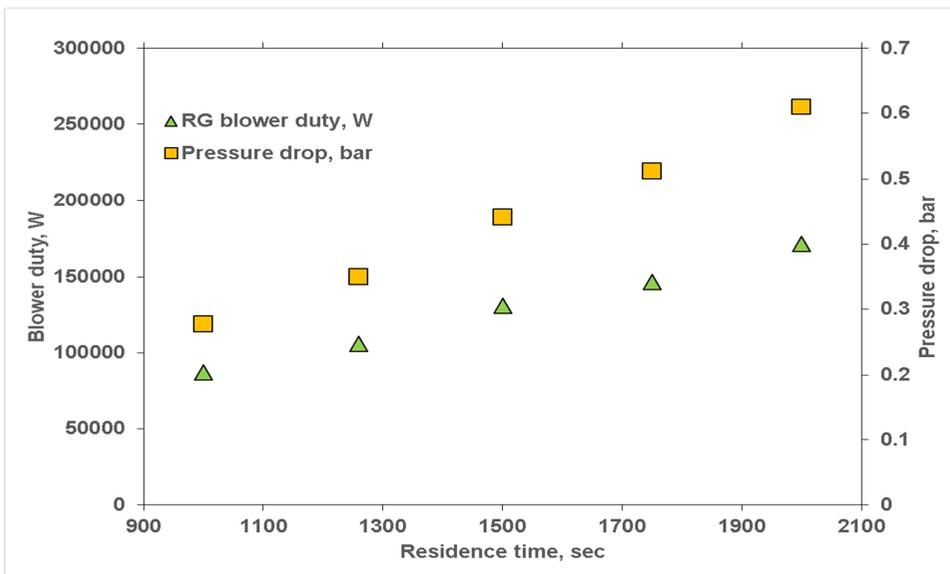


Figure 3-11 Sensitivity analysis of pressure drop and blower duty with varying the residence time

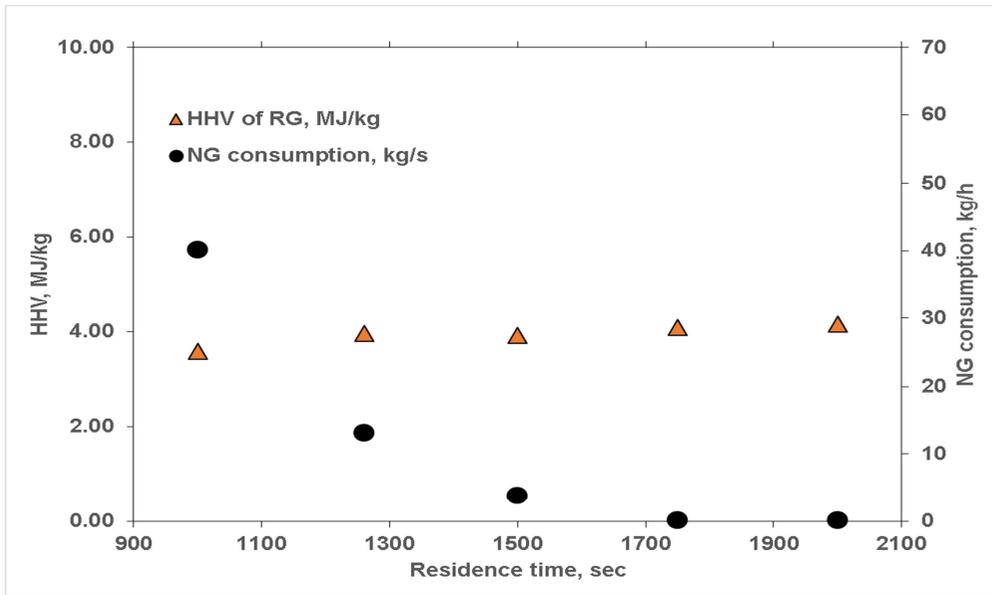


Figure 3-12 Sensitivity analysis of HHV of recycled gas and NG consumption with varying the residence time

To analyze the influence of operation variables on the product properties, the percentage of product properties variation were calculated with variation of 1% in operation variables. Table 3-3 represented the comparison of operation variables.

Table 3-3 comparison of influence on the product properties of operation variables

	Temperature of recycled gas	Flow rate of recycled gas	Residence time
Energy yield	0.84	0.11	0.024
Solid yield	0.23	0.24	0.077
HHV of solid	0.18	0.14	0.054

3.3.4. Effect of feed condition on the process parameters

Effect of feed conditions were represented in this section. Moisture contents of feedstock are varied in the range between 5%~25% in this analysis. Figure 3-13 shows the temperature profile in the reactor with varying the moisture contents of biomass feedstock. Because the drying occurs rapidly in the top of reactor, the moisture content of feedstock do not influence to the temperature profile and product qualities. Figure 3-14 represents the temperature profile in the reactor with varying the temperature of biomass feedstock. The temperature of feedstock are varied in the range between 25°C~110°C. As the temperature of feedstock is lower, the temperature profile of reactor is decreased and severity of torrefaction is reduced. Hence the prevention of heat loss are essential during movement of biomass from dryer to the torrefaction reactor.

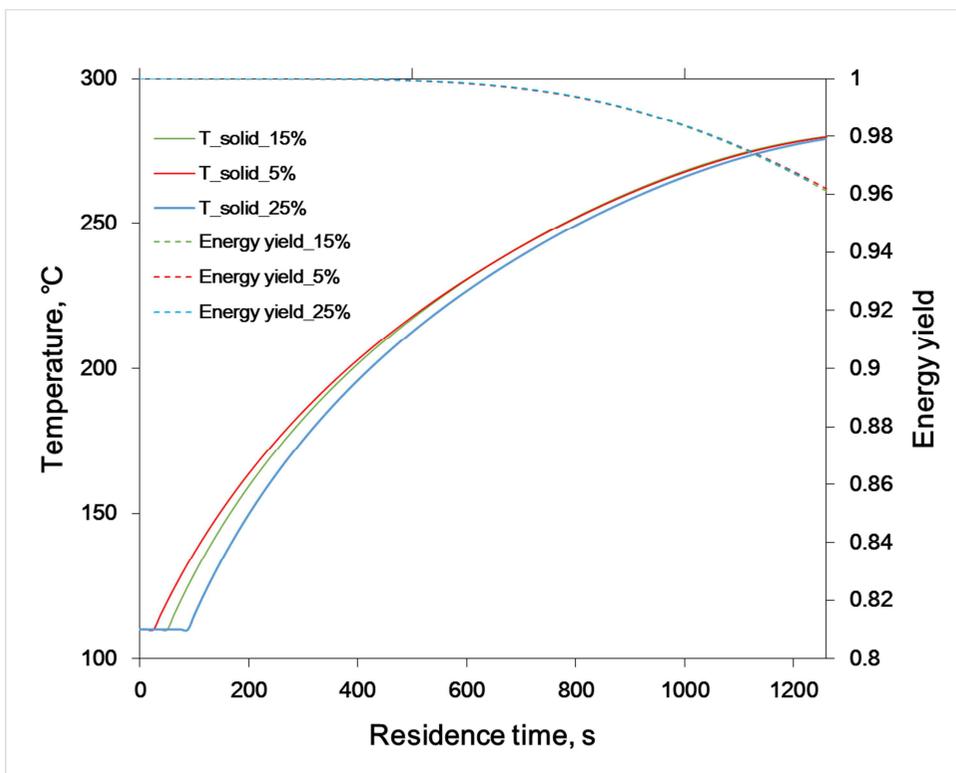


Figure 3-13 effect of moisture content of biomass feedstock

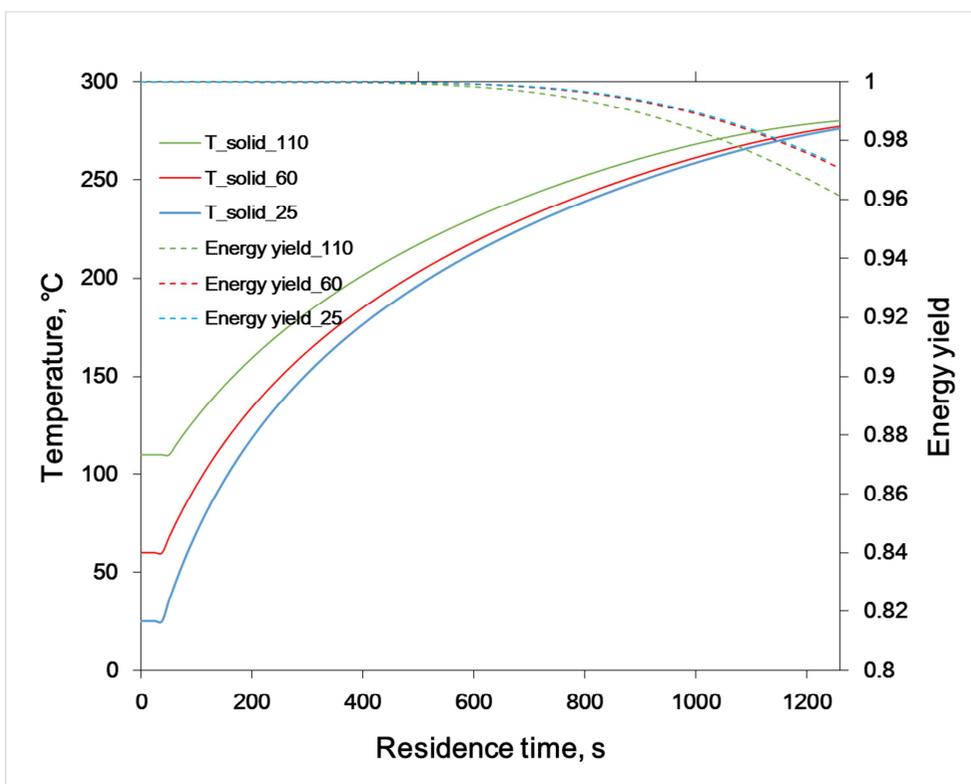


Figure 3-14 effect of temperature of biomass feedstock

3.3.5. Effect of operating conditions on the process parameter with energy yield of 0.961

Because the purpose of torrefaction process is to produce a fuel, the energy yield of torrefied product is specified for process design. Since the analysis on effects of the operating variables will influence design decisions in practical processes, a sensitivity analysis of operating conditions on the process parameters with specified energy yield is required. Among the manipulated variables, the recycled gas

temperature is selected as the variable that should be determined for meeting the energy yield target decided in this chapter. In other words, when the flow rate of the recycled gas and the residence time vary independently, the temperature of the recycled gas is calculated to satisfy the energy yield target. The decrease of the recycled gas flow rate and the residence time results in harsh heat transfer conditions. A higher recycled gas temperature is required with lower values of the recycled gas flow rate and residence time.

Calculated temperature of the recycled gas with varying of the flow rate of recycled gas and the residence time are represented in Figure 3-15 (with specified energy yield of 0.961, which is from base case). When the energy yield is specified, higher temperature of recycled gas is needed for lower flow rate of recycled gas and residence time. The grinding energy of torrefied biomass, which is important properties to evaluate the product, is shown in Figure 3-16. The grinding energy was estimated depending on the solid loss of torrefied biomass. Bates [27] represented the correlation of grinding energy with mass loss of torrefied solid using experimental data for the torrefied willow from Bridgeman et al. [28] as follow:

$$E_{g,t} = 10a(171.6m_l - 6.0767)^b \left(\frac{1}{\sqrt{P}} - \frac{1}{\sqrt{F}} \right) \quad (21)$$

where $E_{g,t}$ is the grinding energy for torrefied biomass in kWh/ton, m_l is the mass loss for torrefied biomass, P is the 80% passing size of the production in μm , F is the 80% passing size of the feed in μm , and values for a and b are provided of 1622

and -1.08 by McIntyre and Plitt [29]. Values for P and F were assumed as 30,000 μm and 200 μm in this chapter. The mass loss of solid increases with the temperature of recycled gas; the mass loss with higher temperature of recycled gas and lower flow rate of recycled gas and residence time is higher than that with lower temperature of recycled gas and higher flow rate of recycled gas and residence time. Because the energy yield was specified, the HHV of solid increases with decrease in the flow rate of recycled gas and residence time as shown in Figure 3-17. Note that higher temperature of recycled gas with lower flow rate of recycled gas and residence time produces the attractive properties of torrefied biomass; higher HHV, higher grindability, and lower mass yield with same energy yield. The blower duty increases with the flow rate of recycled gas and residence time as shown in Figure 3-18. As previously described, an increase in the recycled gas flow rate causes an increase in the pressure drop in the reactor because the increased velocity of the gas phase increases the friction in the reactor. Moreover, the pressure drop in the reactor is increased by a high residence time. The pressure drop, which the blower should cover, increases with increasing recycled gas flow rate and residence time. An increase in the flow rate of the recycled gas increases the amount of fluid that the blower needs to handle. The cost of the blower duty therefore increases with increasing recycled gas flow rate and residence time. Figure 3-19 shows the NG consumption with varying the recycled gas flow rate and residence time. Although the trend of NG consumption are not clearly represented due to the influence of various variables, the NG consumption are seen to increase with the flow rate of

recycled gas and residence time. As previously described, the NG consumption is calculated to satisfy the 2 specification. In this operating range, the specification of adiabatic flame temperature of 1000 °C has superiority due to low flow rate of recycled gas. Moreover, because the torrefaction gas is maintained at a particular heating value at a constant energy yield, variations in the heat from torrefaction gas are small, and also variation of the NG consumption should be small. However, since the temperature of recycled gas after reactor increase with the temperature of recycled gas before reactor, the temperature of torrefied gas toward furnace is higher. It causes that required energy for temperature change from the temperature of torrefied gas to the adiabatic flame temperature of 1000 °C is lowered. Thus, the NG consumption with specified energy yield increase with the flow rate of recycled gas and the residence time.

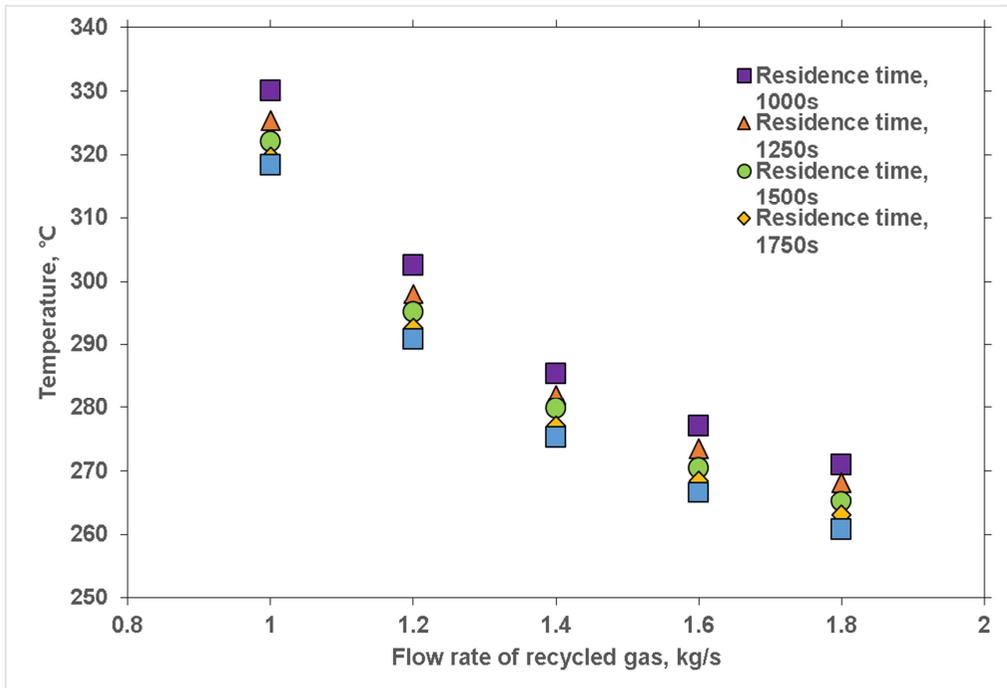


Figure 3-15 Sensitivity analysis of calculated temperature of recycled gas with specified energy yield of 0.961 (base case)

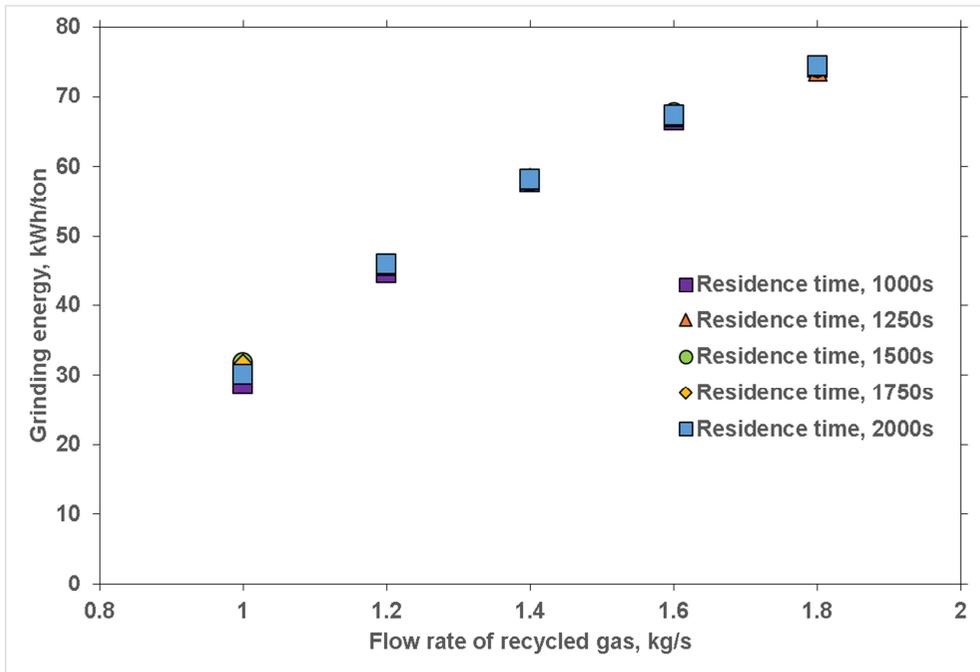


Figure 3-16 Sensitivity analysis of grinding energy of solid with specified energy yield of 0.961 (base case)

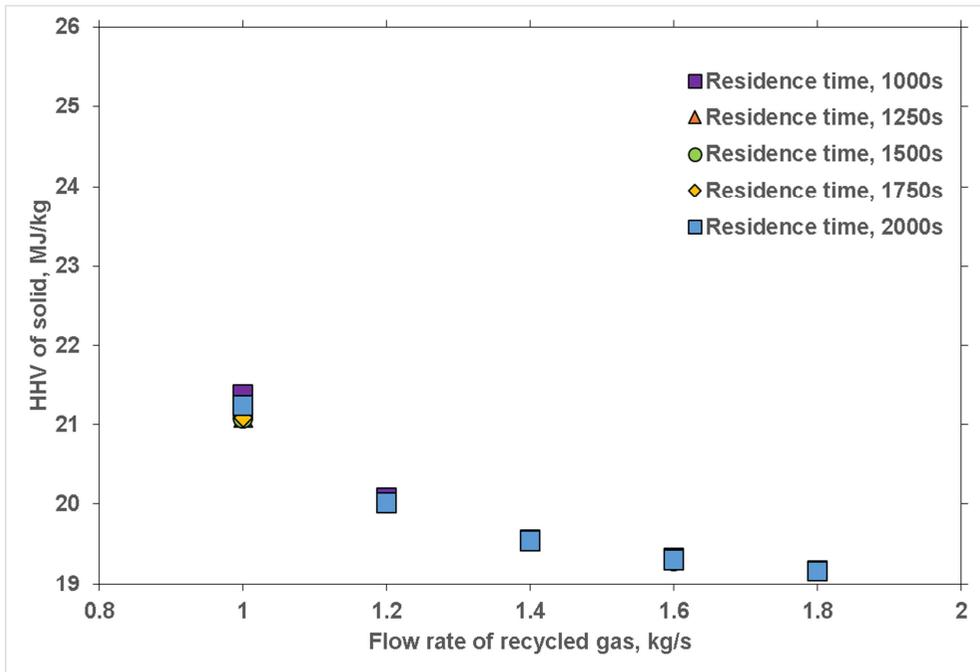


Figure 3-17 Sensitivity analysis of HHV of solid with specified energy yield of 0.961 (base case)

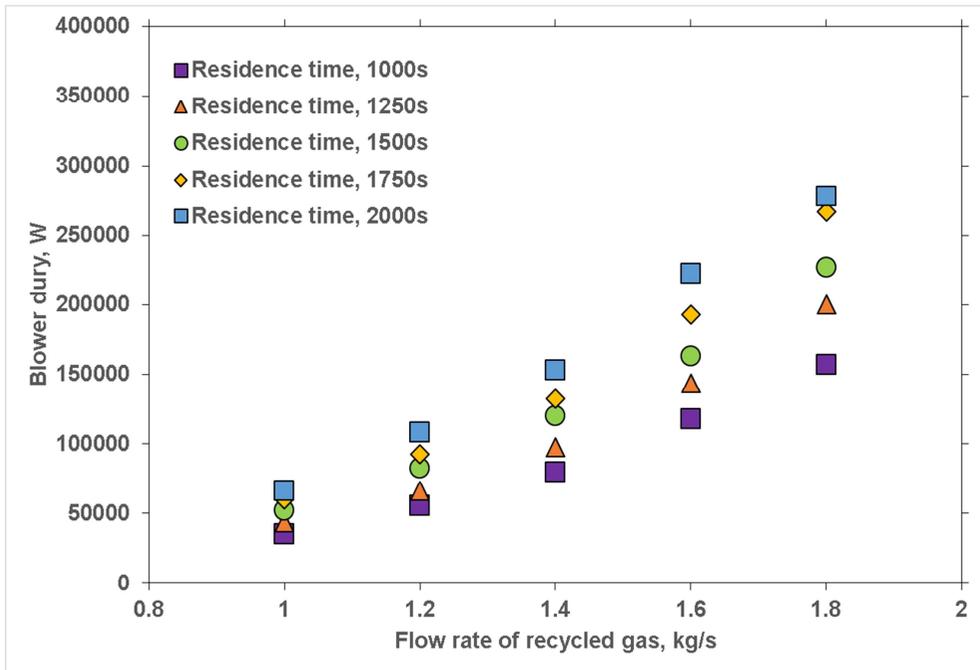


Figure 3-18 Sensitivity analysis of blower duty with specified energy yield of 0.961 (base case)

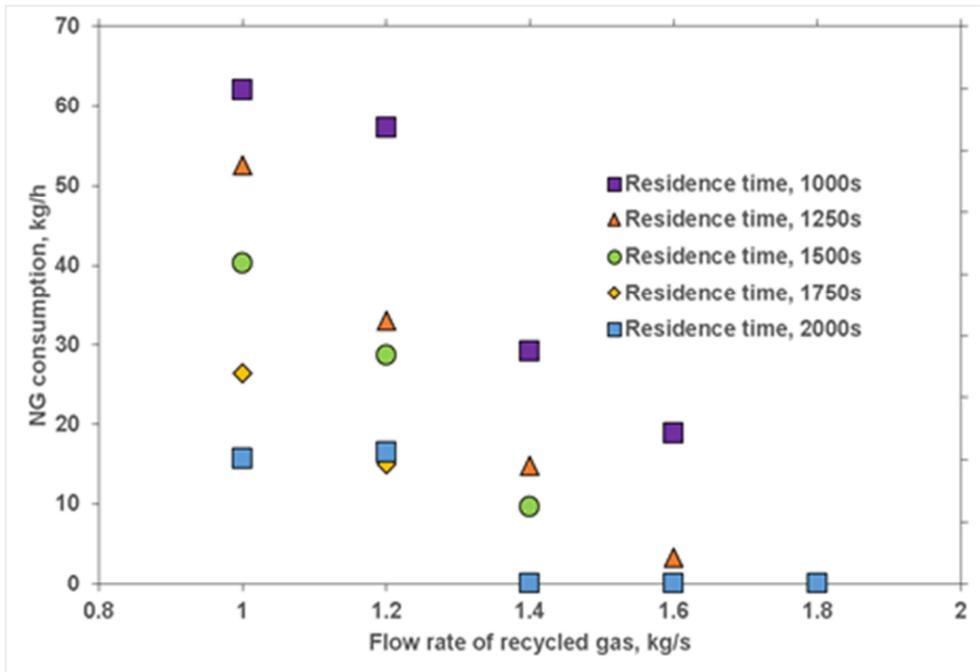


Figure 3-19 Sensitivity analysis of NG consumption with specified energy yield of 0.961 (base case)

3.3.6. Effect of operating conditions on the process parameter with energy yield of 0.9

Figure 3-20 shows the results of sensitivity analysis with the specified energy yield of 0.9, which means more torrefaction progress than the base case. Calculated temperature of recycled gas is generally higher than that with the energy yield of 0.961 as represented in Figure 3-20. Because the mass loss of solid is higher than that with the energy yield of 0.961, the grinding energy is also lower than that, as shown in Figure 3-21. Also, the HHV of solid is improved as shown in Figure 3-22.

The blower duty is slightly higher owing to the same flow rate of recycled gas and residence time, as shown in Figure 3-23. Although the specification of adiabatic flame temperature has superiority in this operating range, there are no NG consumption in this range; the autothermal operation is achieved due to enough heating value of torrefied gas in this operating range.

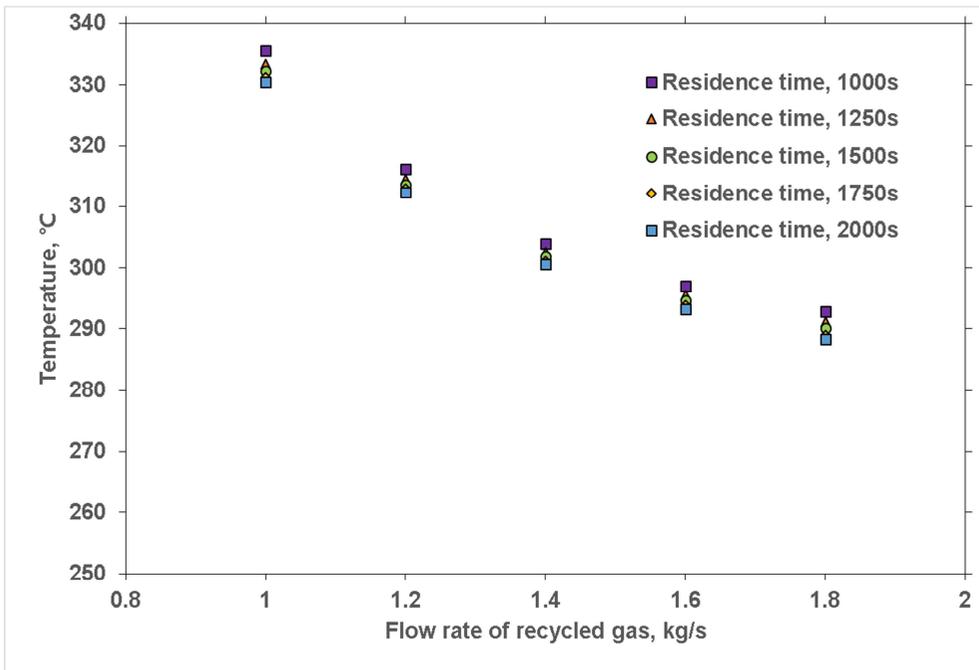


Figure 3-20 Sensitivity analysis of calculated temperature of recycled gas with specified energy yield of 0.9

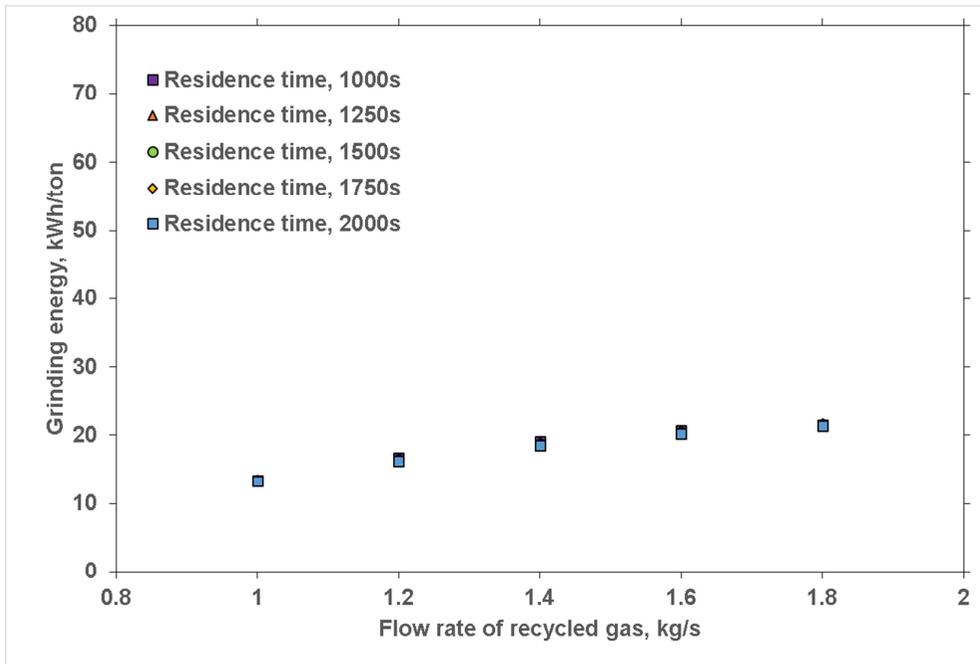


Figure 3-21 Sensitivity analysis of grinding energy of solid with specified energy yield of 0.9

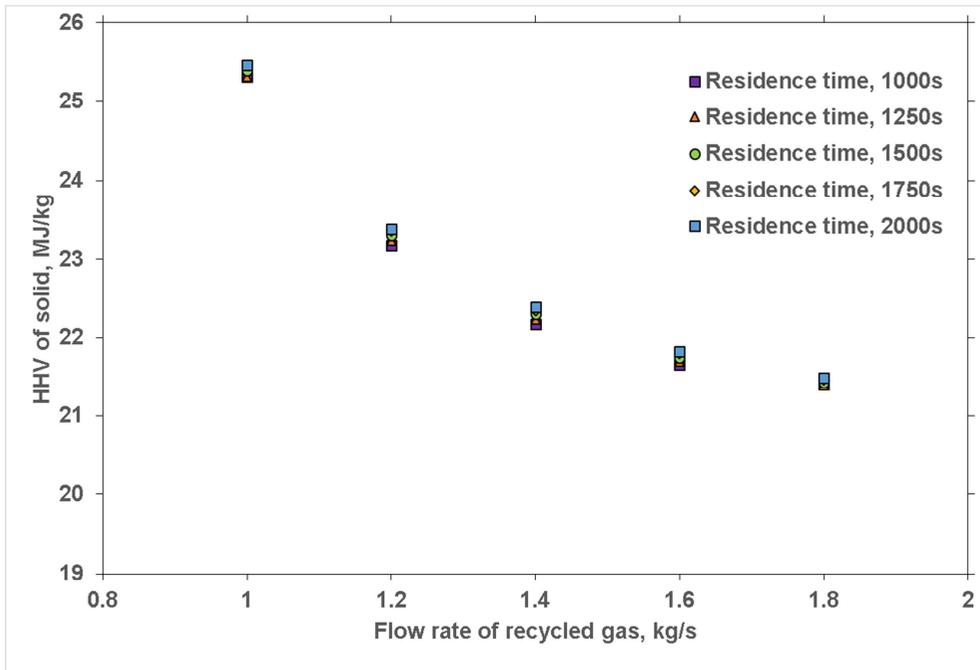


Figure 3-22 Sensitivity analysis of HHV of solid with specified energy yield of 0.9

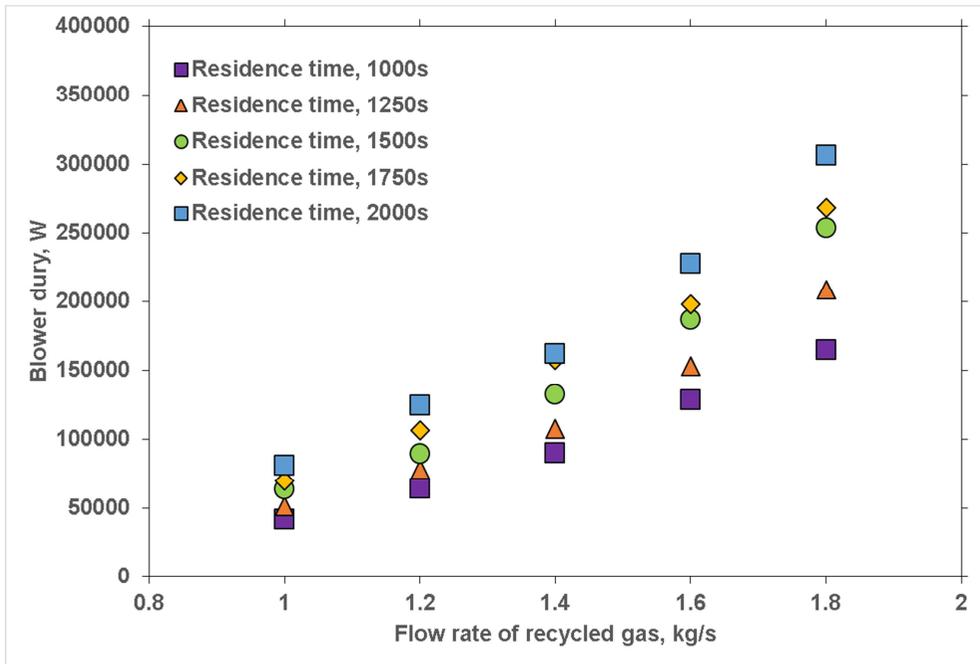


Figure 3-23 Sensitivity analysis of blower duty with specified energy yield of 0.9

The effects of operating conditions for the torrefaction reactor were analyzed using sensitivity analysis considering the parameters for the whole process. In this chapter, the results of sensitivity analysis represented notable discussions which cannot be obtained from the previous researches. One is that the residence time insignificantly influenced the energy yield when the flow rate of recycled gas is low. Another is that higher temperature of recycled gas with lower flow rate of recycled gas and residence time produces the attractive properties, including HHV and grindability, of torrefied biomass when the energy yield is specified. It can be useful for design of commercial torrefaction process, although these discussions need to be investigated more with experimental study.

CHAPTER 4 : Optimization and economic comparison of process alternatives for biomass torrefaction

4.1. Introduction

The process to handle the solid product is simply consists of dryer, torrefaction reactor and grinder, as shown in Figure 2-1. Bergman et al. proposed the heat recovery scheme in which the volatiles generated from solid during the torrefaction are used as a fuel for combustion [3]. After proposed process scheme by Bergman, unfortunately, few studies on the development for biomass torrefaction process were conducted. Several studies developed a flowsheet model using the commercial process simulation tool Aspen plus [30, 31]. However, they used the simple lumped reactor model provided by Aspen plus, which does not consider a kinetics model of the solid and volatile production in the torrefaction reactor. Yash Joshi et al. developed the tool to simulate a steady state model of the torrefaction process by linking unit operation blocks of drying and torrefaction along with auxiliary process equipment [14]. They used the reactor model assuming isothermal condition instead of detailed heat transfer model. In an aspect of systematic process, that assumption significantly influences accuracy of the product properties and process parameters, as the isothermality in reactor depends on the operating variables such as a flow rate

of recycled gas [32]. Other researches on the torrefaction process has mainly focused on the effects of operating conditions on the product properties based on experimental studies [8, 33]. No further research on development of the process design for torrefaction has been published since Bergman proposed the heat recovery concept.

Torrefied product quality and process efficiency is affected by various parameters including the properties of biomass feedstock and operating conditions. Optimization of operating conditions in torrefaction process is essential to achieve economic feasibility for a commercial process. Several study on optimization of torrefaction process conditions were carried out. Lee et al. investigated the optimal conditions for the torrefaction of mixed softwood by response surface methodology [26]. They used a severity factor to integrate the effects of reaction times and temperature into a single variable. Chin et al. presented the optimum torrefaction condition for the production of high energy density biofuels [25]. They used the technique of response surface methodology to find the optimal conditions for high density solid fuel. Most studies with the aim of improvement of torrefaction conditions considered only product quality like energy density and grindability, not the process parameters which determine the process economics.

In this paper, base case of process design and process alternatives for the torrefaction process were developed. The base case of torrefaction process was established considering the heat recovery concept by Bergman using process model developed from our previous work [32]. Operating variables of base case were

determined from literatures. The base case of torrefaction process is analyzed to find out the opportunities to improve the process efficiency related to economics. The process alternative is built to overcome the inefficiency found from the analysis result of base case in this paper. In order to optimize the operating variables, assessment methods for torrefaction process with varying the operating conditions were compared and analyzed. Based on selected assessment method, the economics of torrefaction process was evaluated and the optimization problem of operating variables is formulated. The operating variables were optimized to minimize the capital and operating costs in the torrefaction process. The optimized process alternatives were compared on economics of process.

4.2. Development of process alternatives

4.2.1. Analysis on base case of process design

The volatile gas includes a significant amount of water evaporated from the biomass feedstock and from the volatile products during the torrefaction as presented in Table 4-1. Since this high water content results in inefficient combustion of the wet torrefaction gas, it is questionable whether the torrefaction gas will be combustible [3]. For completely dry biomass, the water concentration in the volatile gas varies from 50 to 80 wt.% when the amount of water yielded by the torrefaction reaction changes from 5 to 15 wt.% [3]. High water contents of volatiles caused low

temperature of the flue gas from the combustion, as the flue gas has high heat capacity. It leads to require additional fuel to satisfy the target temperature of recycled gas. Furthermore, the water content of the combustion gas is maintained until it moves to the dryer as a flue gas. The flue gas from the furnace passes through the heat exchanger and then reaches the dryer to deliver its residual heat to the raw biomass. A high moisture content of the flue gas will prevent the drying of the biomass, making a larger dryer area necessary. Thus, it is necessary to reduce the water content in the volatile gas obtained during the torrefaction so that it can be utilized efficiently.

Table 4-1 composition of volatiles from the simulation results of base case

Component	Composition
Acetic acid	0.065
Water	0.762
Formic acid	0.023
Methanol	0.025
Lactic acid	0.013
Furfural	0.005
Hydroxy acetone	0.005
CO₂	0.084
CO	0.017

The temperature distribution in the reactor is affected by operating conditions. The temperature distribution of base case was non-isothermal as shown in Figure 4-1(a). Since most experiment results of torrefaction are with isothermal condition, the experimental results are difficult in application to the reactor model and design with the non-isothermality in the reactor obtained from simulation results. The experiment with non-isothermal condition is difficult to be carried out. Also, torrefaction is not progressed with non-isothermal condition when the range of high temperature is short in the whole distribution.

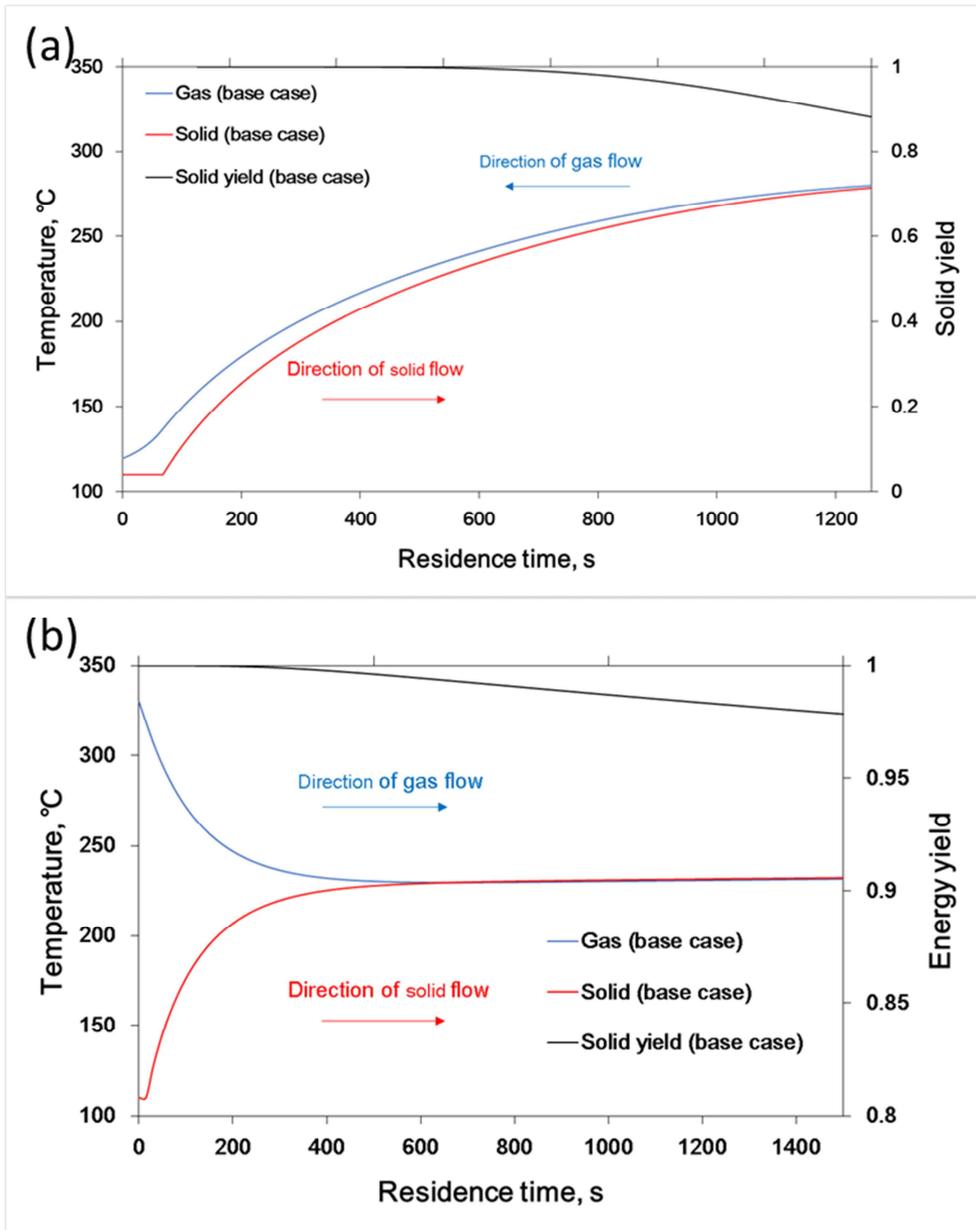


Figure 4-1 temperature distribution of (a) base case and (b) co current with 1.5 kg/s of flow rate of recycled gas, 330°C of temperature of recycled gas and 1500s of residence time

4.2.2. Opportunities to enhance the process

There are two options for the arrangement of the recycled gas stream in the torrefaction reactor: counter current and co current. Commercial torrefaction reactors such as TORSPYD have counter current flow arrangements [18]. Conversely, some torrefaction experiments have used a co current flow arrangement to heat up the biomass in the reactor [12]. The flow arrangement of torrefaction reactor is commonly counter current flow. It is known that the counter-current flow arrangement of the hot stream and the cold stream require less heat exchanger area because of the higher LMTD (log mean temperature difference) between the hot and cold streams. This flow arrangement causes the non-isothermal condition. On the other hand, as shown in Figure 4-1 (b), isothermality of temperature distribution is achieved in the co current flow of reactor. In the early part of reactor, the temperature of solid and gas phase are rapidly converged and maintained at converged temperature and achieve the isothermal conditions. Furthermore, the outlet stream for the recycled gas in the torrefaction reactor has a higher mass flow rate than the inlet stream, since the volatile gas is added to the recycle gas during torrefaction. When the recycle gas flows in the same direction as the solid, the effect of increase in the mass flow rate of recycled gas is taken. Therefore, this arrangement has a better capacity to remove the higher heats of reaction at higher reactor temperatures. In this study, in order to maintain isothermal conditions, a co current flow arrangement was considered as process alternative.

Table 4-1 shows the composition of volatiles in the base case of process design.

High water content in the volatiles cause the low heating value and low temperature of flue gas in the furnace. The volatiles from the torrefaction reactor is divided into a stream that acts as a heat carrier gas that moves to the torrefaction reactor, called the recycled gas, and a stream that is used as fuel, called combustion gas, in the furnace to supply heat energy to the recycle gas. High water contents mainly come into question in the combustion gas. In order to reduce the water contents in combustion gas, condensation of combustion gas can be applied. When the combustion gas is under condensing condensation, the water contents of gas is reduced and heating value of gas is improved, although the temperature of volatiles is reduced. Thus, the condenser is added to the combustion volatiles in the process alternative.

A process for improving the combustibility by reducing the water content in the volatile gas is proposed as a process alternative. This process takes advantage of the fact that the composition of the generated volatile gas varies with length of the torrefaction reactor. As shown in Figure 4-2, the noncombustible water and carbon dioxide species are mainly generated in the early part of reactor, whereas lactic acid and furfural, which have higher heating values, are mainly located in the late part of the reactor. Therefore, in the process alternative, additional volatile outlet streams was created along the length of the torrefaction reactor. The additional stream at the early part of reactor mainly contains water, because the evaporation of water occurs in the early part of reactor. The removal of this stream therefore results in a decrease in the water content of the volatiles.

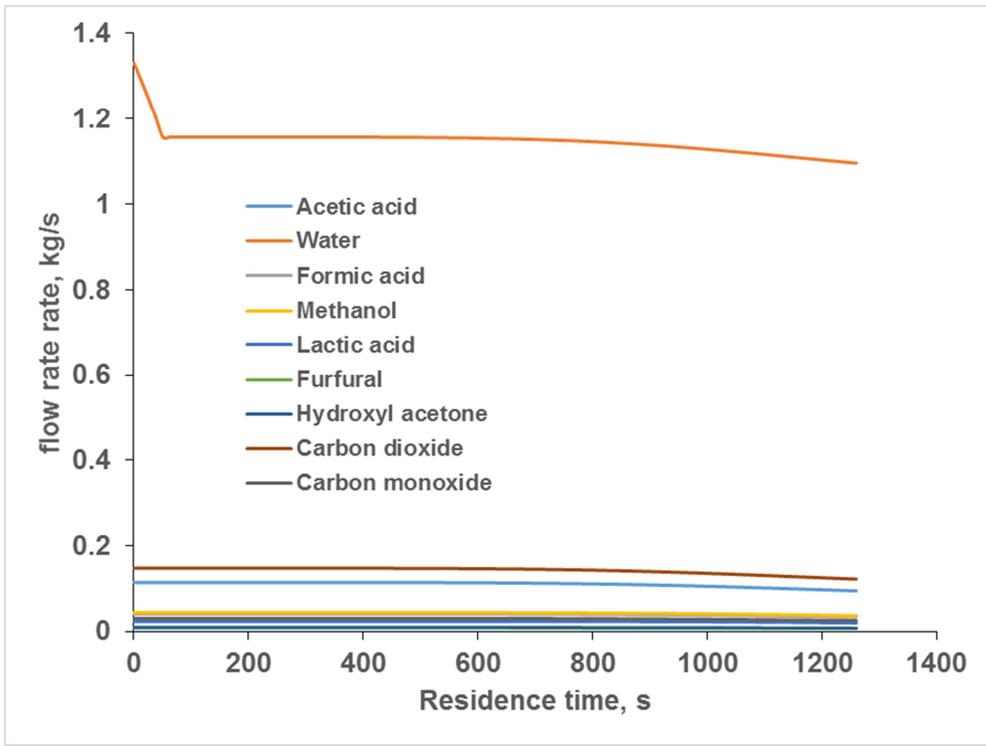


Figure 4-2 flow rate of component in gas phase obtained from simulation results of base case

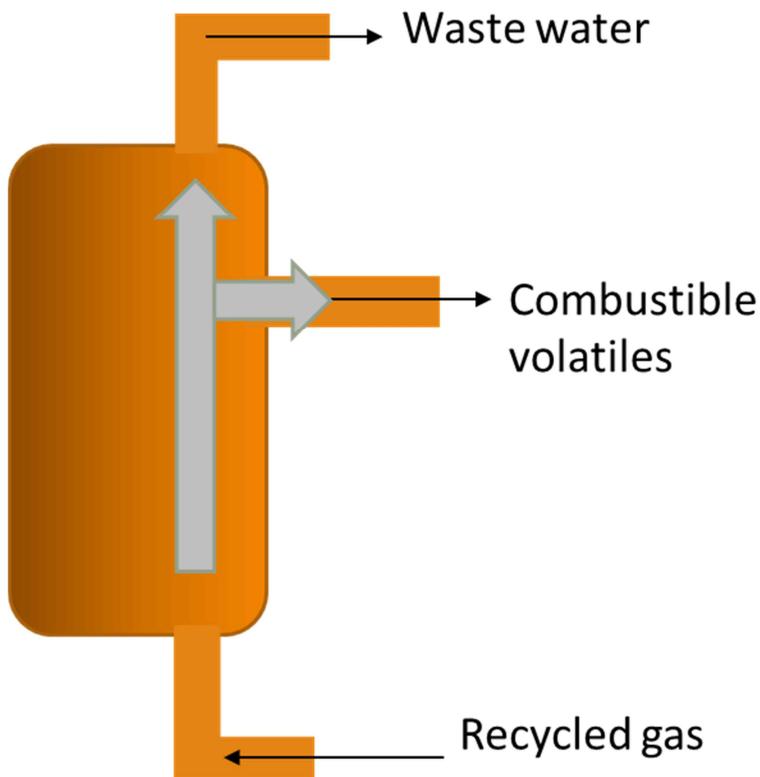


Figure 4-3 scheme of moving bed reactor with additional volatile outlet streams

4.2.3. Summary of process alternatives

Table 4-2 presents the conditions of process alternatives proposed in this study. The base case has reactor with counter-current, one volatile outlet stream and no condenser. The process structure of alternative 1 is equal to that of base case. The operating variables of the base case are optimized in alternative 1. The optimization of operating variables is described in section 3. The process structure of alternative 2 is changed for the co current flow arrangement as shown in Figure 4-4. The

process structure of alternative 3 is changed to add the condenser on the combustion gas as shown in Figure 4-5. The process structure of alternative 3 is changed on the reactor structure into 2 volatiles outlet streams. The operating variables of process alternatives 1-4 are optimized as explained in section 3.

Table 4-2 summary of process alternatives

	Flow arrangement	Volatiles outlet	condenser	Operating variables
Base case	Counter current	1	No	-
Alternative 1	Counter current	1	No	Optimized
Alternative 2	Co current	1	No	Optimized
Alternative 3	Counter current	1	Yes	Optimized
Alternative 4	Counter current	2	No	Optimized

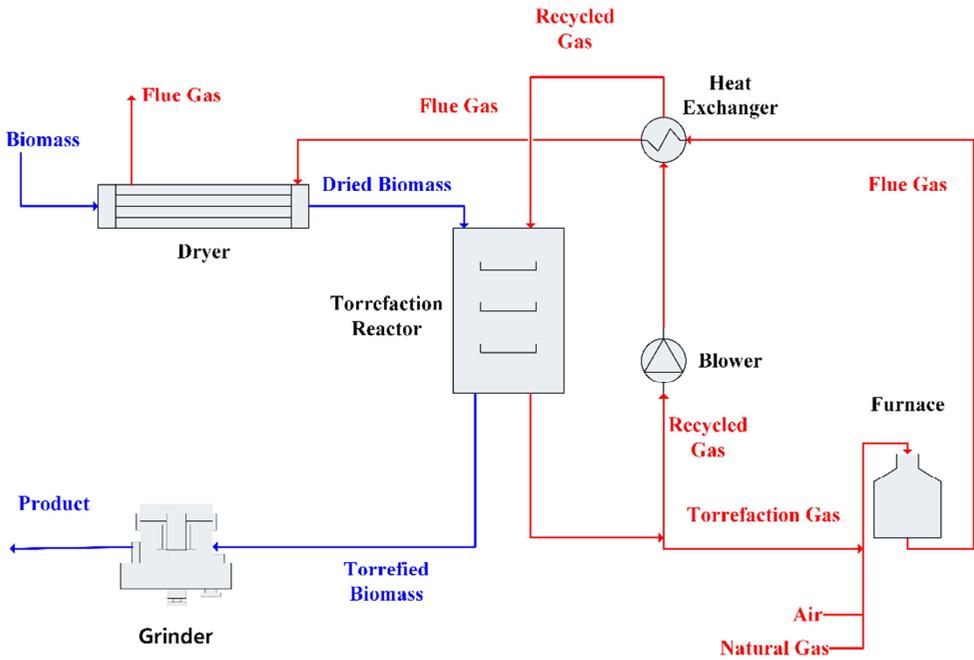


Figure 4-4 scheme of alternative 2

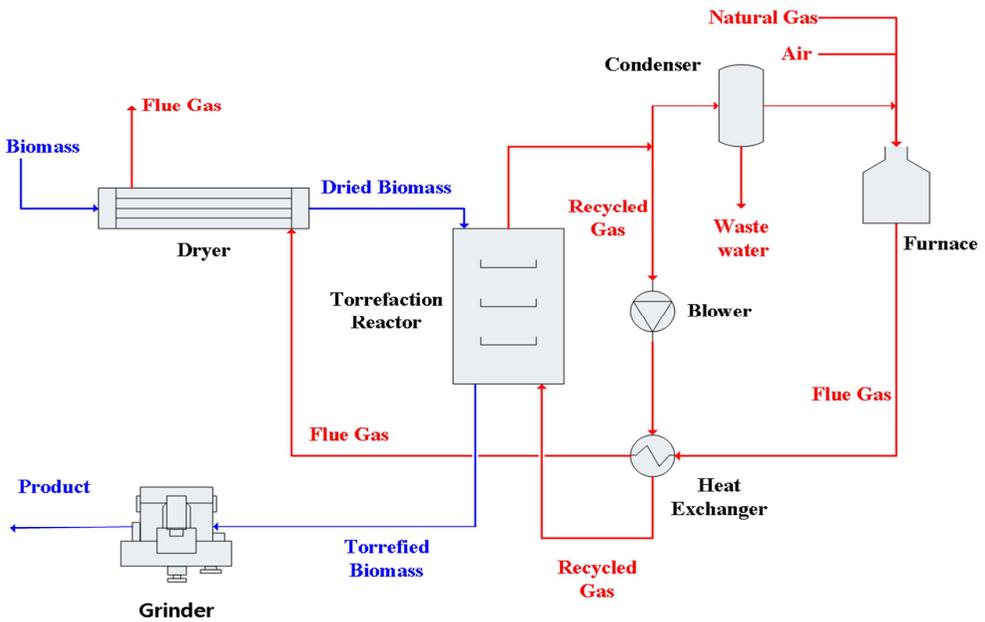


Figure 4-5 scheme of alternative 3

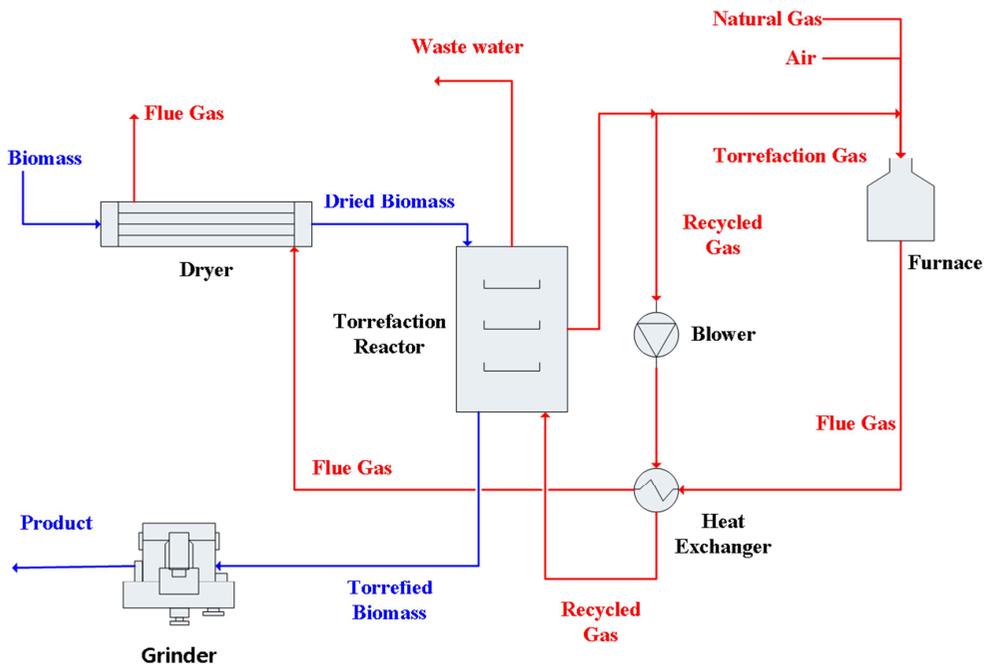


Figure 4-6 scheme of alternative 4

4.3. Optimization problem formulation

4.3.1. Objective function

In order to formulate the optimization problem, the method to assess the process economics should be determined, which become the basis on the objective function of the optimization problem. Table 4-3 shows the considerations to choose the assess method. When the assessment of cost basis is chosen, several assumptions for cost model of entire process, such as return on investment, are used. Because the

assumptions for cost model become different depending on the researcher's propensity, optimal point obtained from cost model with wrong assumptions can be poor results in real plant. Additionally, when the product properties is not specified in assessment of cost basis, it is impossible to estimate the cost varying with the product properties. On other hand, assessment of energy basis use energy yield to evaluate the performance of process. Definition of energy efficiency in torrefaction process was represented in Bergman's report [3].

$$\eta_p = \frac{F_{prod} LHV_{prod}}{F_{feed} LHV_{feed} + E_{util}} \quad (22)$$

As shown in Eq. 22, energy efficiency becomes highest value at conditions that torrefaction do not occurs, when the product properties is not specified in assessment of energy basis. If the product energy yield is specified, all variables except for energy of utility in the Eq. 22 are fixed. Thus, optimization of energy efficiency become minimization of utility consumption and searching the autothermal conditions. Although utility consumption is minimized in the autothermal conditions, it cause to increase the capital cost of units such as torrefaction reactor. Although energy efficiency is useful for simple comparison of processes, it is not suitable for objective function of optimization problem for the torrefaction process. As described above, four assessment method were analyzed to check its limitation. As a result, assessment method of cost basis with product properties specification was suitable for the objective function in this study, if the assumptions for cost mode are reasonable.

The energy yield, solid yield and grindability are commonly used to evaluate the quality of torrefied product. In conditions for the base case of process design, torrefied product is applied for co-firing as a fuel. Hence, the energy yield of torrefied product is selected for specified product properties of chosen assessment method.

Table 4-3 assessment methods for torrefaction process

Base index	Constraint for product properties	Limitations
Cost	Yes	Optimum is dependent on several assumptions
Cost	No	It is difficult to evaluate the cost for varying product properties
Energy	Yes	Optimum is condition that torrefaction not occurs
Energy	No	Energy efficiency is function of only utility

The operating variables of process alternatives intricately affect economics of the torrefaction process. The objective function in the optimization problem should include the cost function affected by operating variables, as follows:

$$\text{Minimize } C_T = C_{NG} + C_E + ROI \times (C_R + C_F + C_{HX} + C_B) \quad (23)$$

where the objective function is to minimize the total cost, C_T , which is the sum of the annual utility costs and annualized equipment costs affected by the manipulated variables. Utility costs include the annual cost of natural gas (C_{NG}) in the furnace and electricity (C_E) in the blowers and grinding.

The prices of natural gas and electricity are taken from the literature, as shown in Table 4-4. The flow rate of natural gas was calculated to satisfy the specification, from simulation results by developed process model. The specification is to satisfy the recycled gas temperature in the heat exchanger by varying the consumption of natural gas. The blower cost is obtained by multiplication of the electricity price and the blower duty, which is calculated by process simulation by inputting the flow rate of the recycled gas and pressure drop obtained from the reactor model. In this study, the capital cost includes the cost of the torrefaction reactor (C_R) and the furnace (C_F). Although there are several units in the biomass process, the manipulated variables affect the capital costs of the reactor and the furnace; moreover, these are the most expensive pieces of equipment used in the biomass process. The cost of the moving-bed torrefaction reactor was obtained from Bergman's work (Table 4-4). The cost of the furnace is calculated using Eq. (3) though Eq. (5) in Table 4-4 (Seider et al., 2009), in which heat duty of the furnace includes the heat energy from natural gas and the combustion gas. In those equations, C_B is the base cost of furnace, F_P is the pressure factor, and the material factor, F_M , is 1.4 for Cr-Mo alloy steel, Q is the heat duty in Btu/h, and P is the design pressure of furnace in psig. To annualize the capital cost, the reactor cost and furnace cost are multiplied by the return on investment (ROI), which is assumed to be 0.2 year.

Table 4-4 input data for economic evaluation of torrefaction process

Conditions	Value or approach	Reference
Natural gas	\$ 43/MWh	Bauer

price		(2014)
Electricity price	\$ 120/MWh	Bauer (2014)
Purchase cost of torrefaction reactor	M€ 0.275 for a moving-bed reactor of diameter of 1.3 m and length of 6.7 m	Bergman (2005)
	$C_F = F_P F_M C_B$	
Purchase cost of furnace	$C_B = \exp\{0.08505 + 0.766[\ln(Q)]\}$ $F_P = 0.986 - 0.0035\left(\frac{P}{500}\right) + 0.0175\left(\frac{P}{500}\right)^2$	Seider (2009)
Purchase cost of heat exchanger	$C_p = \exp\{11.667 - 0.8709[\ln(A)] + 0.9005[\ln(A)]^2\}$	Seider (2009)
Purchase cost of blower	$C_p = 0.6 \exp\{6.6547 + 0.7900[\ln(P_C)]\}$	Seider (2009)
Annual operation time	8000 h	Bergman (2005)
Return on investment	0.2	Assumed

The grinding energy of torrefied biomass, which is important properties to evaluate the product, is shown in Fig 7(b). The grinding energy was estimated depending on the solid loss of torrefied biomass. Bates [27] represented the correlation of grinding energy with mass loss of torrefied

solid using experimental data for the torrefied willow from Bridgeman et al. [28] as follow:

$$E_{g,t} = 10a(171.6m_l - 6.0767)^b \left(\frac{1}{\sqrt{P}} - \frac{1}{\sqrt{F}} \right) \quad (24)$$

where $E_{g,t}$ is the grinding energy for torrefied biomass in kWh/ton, m_l is the mass loss for torrefied biomass, P is the 80% passing size of the production in μm , F is the 80% passing size of the feed in μm , and values for a and b are provided of 1622 and -1.08 by McIntyre and Plitt [29]. Values for P and F were assumed as 30,000 μm and 200 μm in this study.

4.3.2. Manipulated variables

The torrefaction progresses more intensely with increasing temperature and flow rate of the recycled gas and the residence time. However, a higher recycled gas temperature and flow rate and a longer residence time increase the operating and capital costs and affect the economics of the process design. The recycled gas temperature is increased by increasing the natural gas flow rate. A higher recycled gas flow rate causes a higher pressure drop and increases the blower duty. A long residence time needs the torrefaction reactor to be lengthened, so the equipment is more expensive. The operating variables of the torrefaction reactor therefore need to be optimized to take account of the economics of the torrefaction process. Thus,

three manipulated variables of the torrefaction reactor were chosen to formulate the optimization problem. The recycled gas temperature, which is the first of the manipulated variables, affects the driving force of heat transfer between the solid and the recycled gas in the reactor. The flow rate of natural gas, which is one aspect of the economics of the torrefaction process, is influenced by the temperature of the recycled gas. An increase in the flow rate of the recycled gas, which is the second manipulated variable, increases the amount of heat energy in the recycled gas, so the energy capacity of the recycled gas increases. However, in terms of the economics of the torrefaction process, it increases blower duty, which is proportional to the flow rate of the stream through the blower. The flow rate of the recycled gas has additional effects on the blower duty. This manipulated variable increases the gas flow velocity in the reactor, which increases the pressure drop in the reactor. The increase in blower duty is therefore more than proportional to the recycled gas flow rate. Moreover, to supply the heat energy for a higher recycled gas flow rate, more natural gas needs to be consumed, and this affects the economics of the torrefaction process. The last manipulated variable in this optimization problem is the residence time. Most torrefaction studies consider the residence time as an operating variable. The torrefaction progress increases with residence time. An increased residence time requires a larger reactor, which increases the reactor cost and affects the process economics. Moreover, because an increase in the residence time causes a decrease in the gas flow velocity and an

increase in the reactor length, this operating variable affects the blower duty, and therefore affects the process economics.

4.3.3. Constraints

The constraints in this optimization problem should be identified to find feasible ranges of manipulated variables. Hard constraints on the heat and mass balances of the gas and solid phases in the reactor were taken into account using the reactor model. The energy yield specification of product in the torrefaction reactor should be considered as a constraint in this problem.

4.3.4. Solution procedure

The optimization problem was solved using active-set algorithm in optimization toolbox of MATLAB, as shown in Figure 4-7. The cost of the reactor, blower duty, furnace, and natural gas were calculated for the entire range of recycled gas flow rates, temperature and residence times.

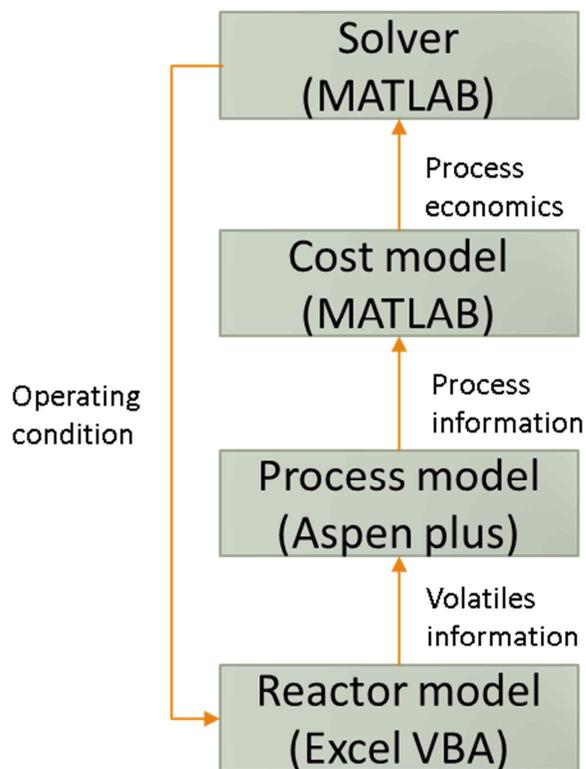


Figure 4-7 solution procedure

4.4. Results and discussion

4.4.1. Alternative 1 (optimized base case)

The operating variables of base case were optimized in alternative 1. The optimal operating variables were compared with the operating variables of base case to demonstrate the effects of optimization in this study. Table 4-6 shows comparisons between various conditions and costs for the optimal case and the base case. The

temperatures of recycled gas and residence time of alternative 1 is higher than that of base case, on the contrary the flow rate of recycled gas in alternative 1 is lower than that in base case. That fact cause the difference of reactor temperature distribution between both cases as shown in Figure 4-8. As discussion in our previous work [8], solid loss is increased with longer range of high temperature at particular energy yield specification. The results that solid loss of alternative 1 is higher in Table 4-7 are reasonable due to a temperature distribution of solid in alternative 1. Blower duty of alternative 1 is lower due to lower flow rate of recycled gas and NG consumption of alternative 1 is higher so that it is influenced various factors. The energy yield distribution in both cases are converged to 96.1%. Those results lead to the economic comparison as shown in Table 4-8. The electricity cost for blower in alternative 1 is lower and the cost for NG consumption is higher. The grinding energy of alternative 1 is lower due to higher solid loss. Higher residence time of alternative 1 results in higher purchase cost of reactor and the purchase cost of furnace follows the tendency of NG consumption cost.

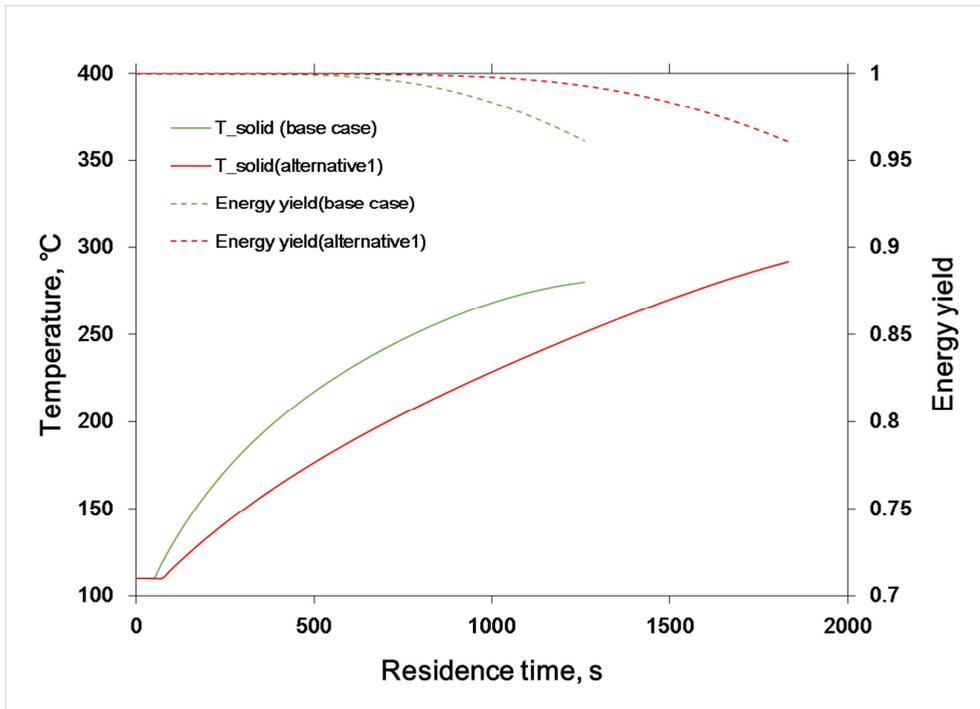


Figure 4-8 temperature and energy yield distribution of base case and alternative 1

4.4.2. Alternative 2 (co current)

Temperature distribution of alternative 2 has different profile, as shown in Figure 4-9. Temperature of solid and gas are converged in early part and maintain the converged temperature. Due to difference of flow arrangement in both case, isothermality of temperature distribution is achieved in alternative 2. But Table 4-6 shows the high temperature of recycled gas is required to satisfy the target energy yield in alternative 2 because of low heat exchange efficiency. Also, high flow rate of recycled gas is required for temperature distribution to be converged in torrefaction temperature. This temperature distribution cause low solid loss and the high flow rate of recycled gas leads to high blower duty as presented in Table 4-7.

However, NG consumption of alternative 2 become zero because the temperature of volatile from reactor is as high as that of torrefied gas. Since the difference of temperature between inlet and outlet stream of recycled gas is low, additional fuel is not required in the alternative 2. It means that the recycle gas poorly convey the heat energy in reactor. It demonstrate that heat exchange with co current has poor efficiency, again. Therefore, high cost for process economics, except for NG consumption and furnace, are required as presented in Table 4-8.

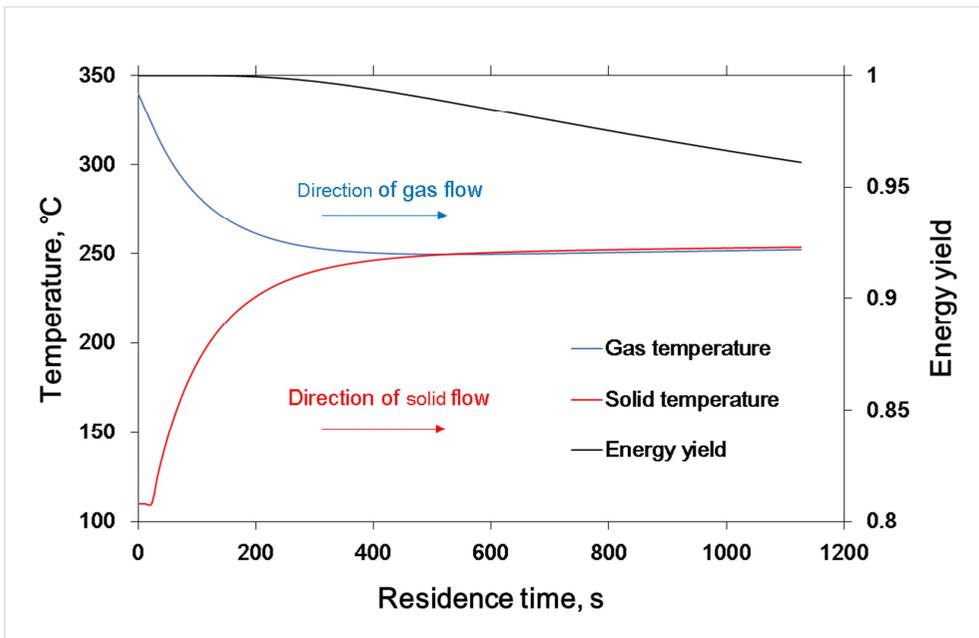


Figure 4-9 temperature of gas and solid phase and energy yield distribution of alternative 2

4.4.3. Alternative 3 (adding condenser)

Temperature distribution of alternative 3 is similar to that of base case as shown in Figure 4-10. The operation variables are similar to those of base case as shown in Table 4-6. Since the combustion gas from the volatiles is condensed before moving toward furnace, HHV of volatile gas is higher than other case as shown in Table 4-7. It cannot accomplish the considerable reduction of NG consumption due to many volatile, which has a heating value, are removed in condenser. However, the furnace cost is significantly reduced in Table 4-8, because the energy to heat up the water is not required. The blower duty and solid yield of alternative 3 are similar to those of base case. Therefore, NG consumption cost and furnace purchase cost are reduced and other cost are not increased.

Table 4-5 stream data of in/out stream of condenser

	Feed	Liquid	Vapor
Temperature, °C	194	60	60
Pressure, bar	1.5	1.0	1.0
Flow rate, kg/s	0.355	0.271	0.083
Composition (mass basis)			
Acetic acid	0.075	0.013	0.275
Water	0.722	0.921	0.076
Formic acid	0.027	0.001	0.109

Methanol	0.032	0.032	0.033
Lactic acid	0.019	0.025	0.000
Furfural	0.005	0.002	0.016
Hydroxy acetone	0.007	0.006	0.009
CO₂	0.094	0.000	0.399
CO	0.019	0.000	0.082

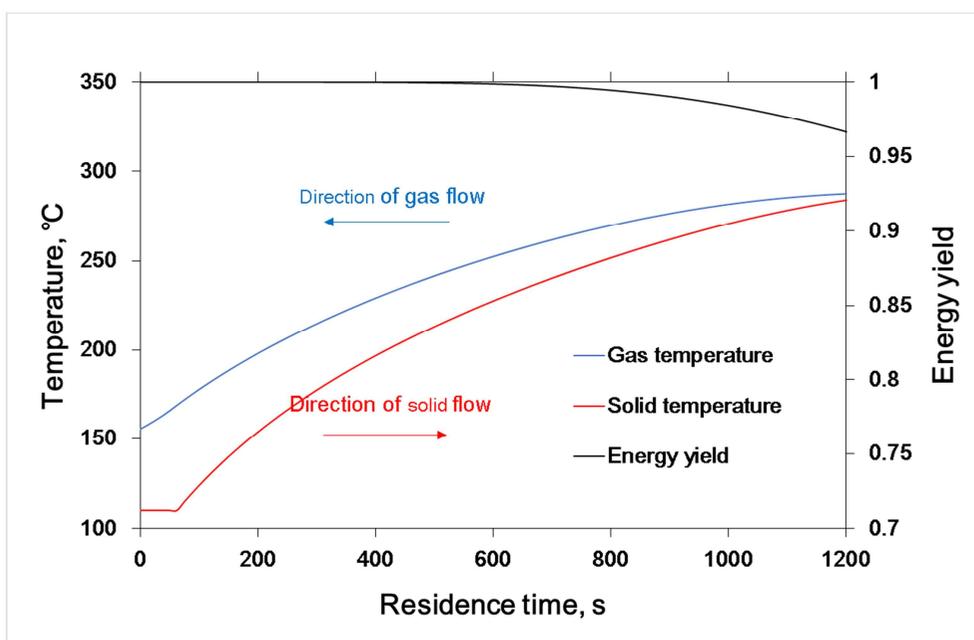


Figure 4-10 temperature of gas and solid phase and energy yield distribution of alternative 3

4.4.4. Alternative 4 (adding rich water volatile stream)

Operating variables of alternative 4 include split ration for additional stream and the split point in the reactor as presented in Table 4-6. The temperatures of recycled gas and residence time of alternative 4 is lower than that of base case, on the contrary the flow rate of recycled gas in alternative 4 is higher than that in base case. Temperature distribution of alternative 4 is shown in Figure 4-11. Since gas stream is moved out at early stage, drop of temperature profile in gas phase occurs. It causes that low temperature of gas phase of early part make the range for solid drying section long. Also, high flow rate of recycled gas is required due to reduced flow rate in early part. However, it achieve high HHV of volatiles as shown in Table 4-7. Since the difference of operating variable, the blower duty has difference. Although addition of rich water volatiles stream achieve the increase in HHV of volatiles, the NG consumption is not reduced. Instead, the blower duty is reduced because additional stream is taken away without blower. Therefore, the costs in alternative 4 are minimum in alternatives

	Combustible volatiles	Waste water
Temperature, °C	160.9	110.5
Flow rate, kg/s	1.42	0.21
Acetic acid	0.0945	0.0698
Water	0.6516	0.7429

Formic acid	0.0335	0.0248
Methanol	0.0388	0.0286
Lactic acid	0.0223	0.0165
Furfural	0.0065	0.0048
Hydroxy acetone	0.0081	0.0060
CO₂	0.1199	0.0885
CO	0.0247	0.0183

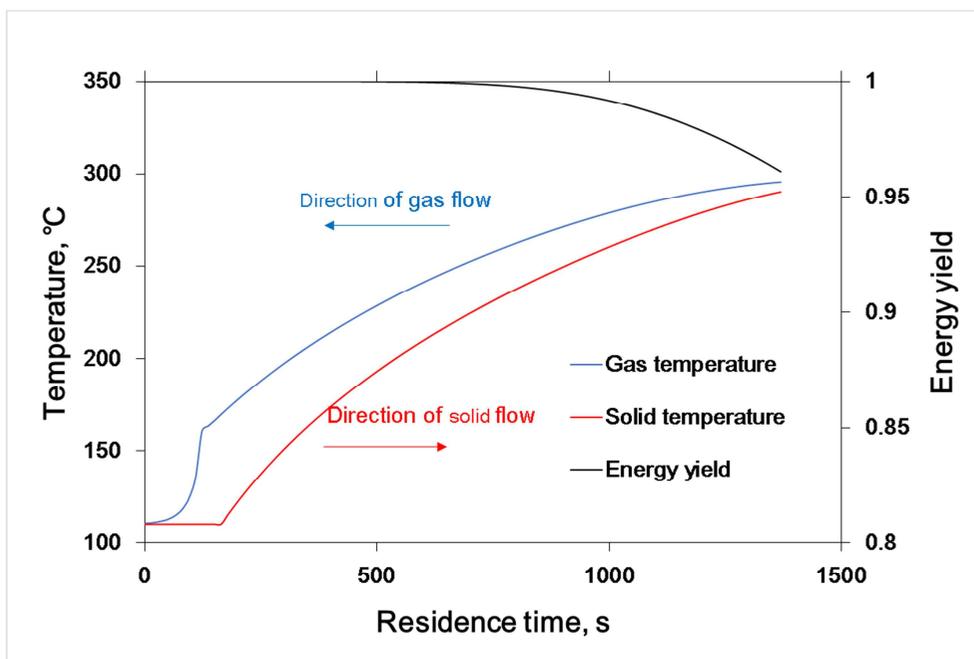


Figure 4-11 temperature of gas and solid phase and energy yield distribution of alternative 4

Table 4-6 operating variables of base case and alternative

Method	Flow rate of RG, kg/s	Temperature of RG	Residence time, s	Split ratio for water reduction	Geometrical point for water reduction, %
Base case	1.436	280	1260		
Alternative 1	1.008	312.47	1848		
Alternative 2	1.689	351.77	1136		
Alternative 3	1.295	290.35	1212		
Alternative 4	1.307	299.12	1381	0.908	9.8%

Table 4-7 product properties and process parameters

Method	Basic process	Alternative 1	Alternative 2	Alternative 3	Alternative 4
Energy yield	0.961	0.961	0.961	0.961	0.961
Solid yield	0.882	0.842	0.910	0.868	0.858
HHV of Solid, MJ/kg	19.48	20.39	18.78	19.75	19.98
Pressure drop, bar	0.350	0.358	0.401	0.308	0.251
RG blower duty, W	105931	79084	178711	85894	66362
TG blower duty, W	31212	33568	30833	33642	8749
air blower duty, W	19407	27267	10754	20977	11674
NG consumption, kg/s	13.1	16.9	0.1	13	16.5
HHV of volatiles, MJ/kg	4.0	4.5	3.8	16.2	5.2
Water contents of volatile	0.762	0.717	0.789	0.079	0.652

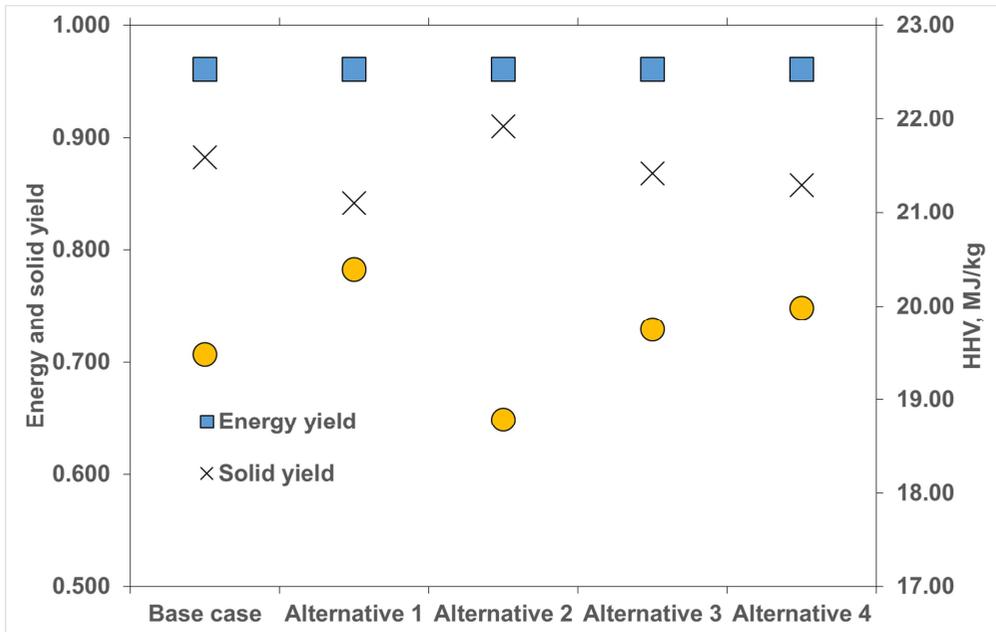


Figure 4-12 energy and solid yield and HHV of product of process alternatives

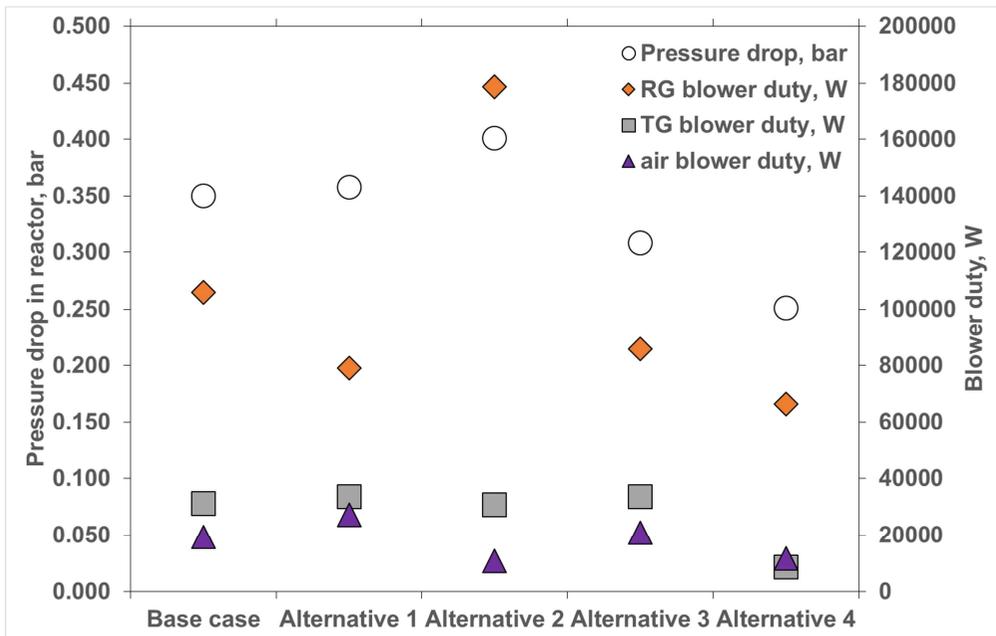


Figure 4-13 pressure drop in reactor and blower duties of process alternatives

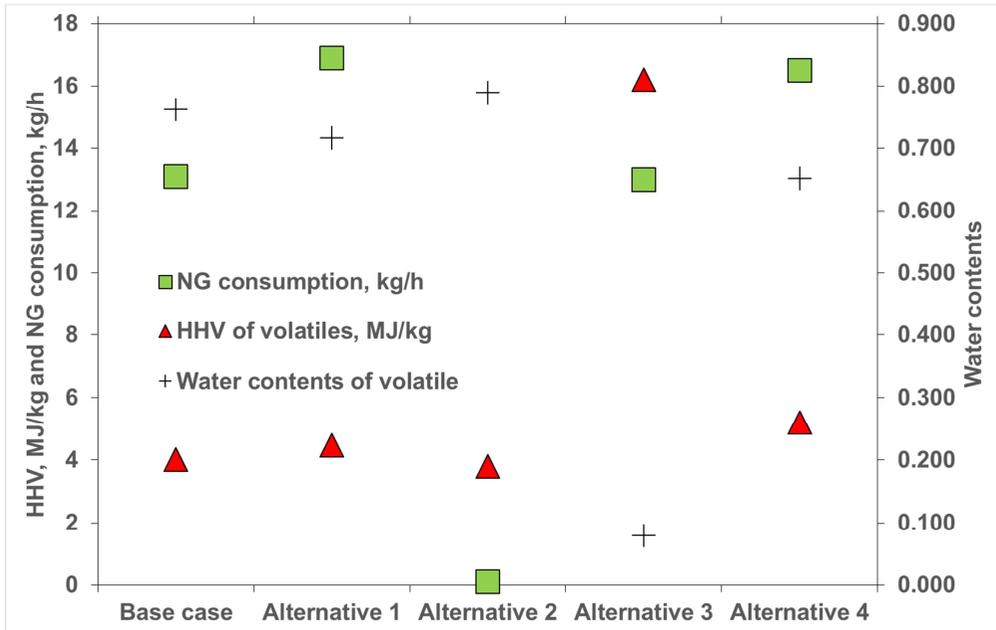


Figure 4-14 NG consumption and HHV of volatiles and water contents in volatiles of product of process alternatives

4.4.5. Economic comparison of process alternatives

Economics of process alternatives are represented in Table 4-8. Alternative 1 has large electricity consumption due to high flow rate of recycled gas. Also, the grinding energy is highest in the alternatives. Alternatives 4 has lower furnace cost than other case because the water contents of volatiles are reduced. Since other cost for alternatives 3 is not increased, this 3 alternatives have good economic potential.

Table 4-8 economic comparison of process alternatives

	Basic process	Alternative 1	Alternative 2	Alternative 3	Alternative 4
Electricity	\$149,877	\$134,211	\$211,134	\$134,857	\$83,212
NG consumption	\$69,441	\$86,624	\$0	\$66,517	\$86,017
grinding energy	\$206,121	\$130,207	\$373,305	\$179,511	\$159,380
Reactor	\$376,750	\$471,768	\$352,359	\$374,593	\$395,980
Furnace	\$291,765	\$351,189	\$217,587	\$152,493	\$164,662
Blowers	\$67,181	\$50,205	\$113,409	\$54,543	\$42,142
Heat exchanger	\$209,136	\$183,471	\$118,473	\$253,711	\$187,424
Total	\$614,405	\$562,369	\$744,805	\$547,953	\$486,651

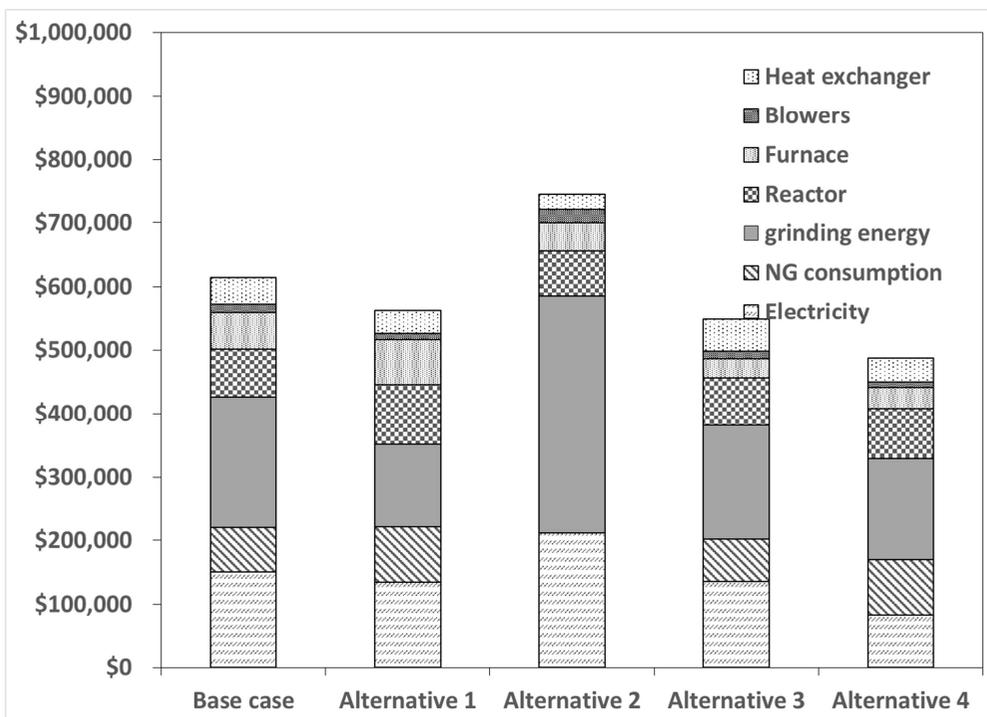


Figure 4-15 cost distribution of process alternatives

In this chapter, the base process design was analyzed to find the potential for improvement of the process efficiency. The non-isothermality of reactor and high water contents of volatiles are found as inefficient factor in the basic process design. The qualitative solution for the inefficient factor were proposed and applied to the process alternatives. The operation variables of process alternatives were optimized and compared based on the economic evaluation. Finally, the optimal process was proposed. The base case of process design for biomass torrefaction was analyzed to find out the potential for improvement of process economics. Since the drawbacks found in this study are key to improve the process efficiency, the solutions will be more investigated to overcome it in further researches. The process alternatives to reduce the drawbacks of base case were proposed. Previous researches on the

torrefaction mostly assumed process of Bergman's concept, but proposed alternatives in this study can be applied to various condition of biomass torrefaction for adaptation to each feature. Economic assessment method and optimization procedure used in this study are suitable to biomass torrefaction process and various biomass application process. Furthermore, through results of comparison of process parameter and economics, effects on the operating variables and process design are in-depth understood. As a results, the optimal process design achieved cost saving of 9.05% than base case.

CHAPTER 5 : Conclusion and Future Works

5.1. Conclusion

This thesis has addressed the design and optimization of biomass torrefaction process using the reactor and process model developed in this study. The reactor model was developed by integrating the kinetics model of torrefaction and thermochemistry model, heat transfer model for pyrolysis, drying, pressure drop model. The process design for biomass torrefaction were conducted and optimized using the process mode.

At first, a one-dimensional reactor model that considered the practical thermal conditions for biomass torrefaction was developed, based on an existing kinetic model and the thermochemistry of the reaction. The reactor conditions in the base case were generated using the developed model based on Bergman's case study. The reactor model developed in this chapter can predict the temperature profiles of the gas and solid phases in the reactor. One feature of the reactor model is that it can be applied to practical operating conditions, including the recycled gas temperature and flow rate, and residence time, to calculate the temperature profile. This allows us to estimate not only the product quality but also to obtain information on the factors that influence the process units, such as the composition and flow rate of the volatile gas and the energy required by the torrefaction reactor. Such predictions are required for the design of industrial processes, because industrial torrefaction plants use recycled gas as a heat carrier instead of using an electric heater. Although the reactor model is based on willow experiments performed by Prins [10] , it will be

useful for the design of general torrefaction processes.

Secondly the effects of operating conditions for the torrefaction reactor were analyzed using sensitivity analysis considering the parameters for the whole process. In this chapter, the results of sensitivity analysis represented notable discussions which cannot be obtained from the previous researches. One is that the residence time insignificantly influenced the energy yield when the flow rate of recycled gas is low. Another is that higher temperature of recycled gas with lower flow rate of recycled gas and residence time produces the attractive properties, including HHV and grindability, of torrefied biomass when the energy yield is specified. It can be useful for design of commercial torrefaction process, although these discussions are needed to be investigated more with experimental study.

Lastly, the basic process design was developed using the process model based on the Bergman's case study. In order to formulate the optimization problem, the assessment method which is suitable to torrefaction process is selected. Selected assessment method is based on the cost basis with specification for energy yield of torrefied product. After formulation of optimization problem, the cost contours of each cost are represented and analyzed. Based on the results of cost contour, optimal operating variables are calculated and compared with operating variables of basic process design. The basic process design was analyzed to find the potential for improvement of the process efficiency. The non-isothermality of reactor and high water contents of volatiles are found as inefficient factors in the basic process design. The qualitative solutions for the inefficient factors were proposed and applied to the process alternatives. The operation variables of process alternatives were optimized and compared based on the economic evaluation. Finally, the optimal process was

proposed.

5.2. Future Works

Future studies about process design and optimization of biomass torrefaction can be considerably complemented by carrying out the experimental study. The reactor model developed in the thesis was not validated at same operating conditions for torrefaction. The proposed process design was also validated by construct the pilot plant for proposed design of biomass torrefaction. After the validation of reactor model and process design, the studies on the process of biomass torrefaction can be extended and applied to the various process for biomass application such as biomass gasification process and bio diesel process. The in-depth analysis including properties of biomass and operating conditions for the reactor and process will give an inspiration to design various biomass application process.

References

- [1] Bates R, Ghoniem A. Biomass torrefaction: Modeling of volatile and solid product evolution kinetics. *Bioresource Technology*. 2012;124:460-9.
- [2] Bates RB, Ghoniem AF. Biomass torrefaction: Modeling of reaction thermochemistry. *Bioresource technology*. 2013;134:331-40.
- [3] Bergman P, Boersma A, Zwart R, Kiel J. Torrefaction for biomass co-firing in existing coal-fired power stations. Technical Report, ECN Biomass. 2005.
- [4] Petroleum B. BP statistical review of world energy. 2003.
- [5] Meijer R. Overview of European torrefaction landscape. Conference Overview of European torrefaction landscape.
- [6] Dutta A. Torrefaction and other processing option. Conference Torrefaction and other processing option.
- [7] Tumuluru JS, Boardman RD, Wright CT. Response surface analysis of elemental composition and energy properties of corn stover during torrefaction. *Journal of Biobased Materials and Bioenergy*. 2012;6(1):25-35.
- [8] Medic D, Darr M, Shah A, Potter B, Zimmerman J. Effects of torrefaction process parameters on biomass feedstock upgrading. *Fuel*. 2012;91(1):147-54.
- [9] Zanzi Vigouroux R, Tito Ferro D, Torres A, Beaton Soler P, Björnbom E. Biomass torrefaction. Conference Biomass torrefaction. p. 859-62.
- [10] Prins MJ. Thermodynamic analysis of biomass gasification and torrefaction. PhD Thesis. 2005.
- [11] Medic D, Darr M, Shah A, Rahn S. The effects of particle size, different corn stover components, and gas residence time on torrefaction of corn stover. *Energies*. 2012;5(4):1199-214.
- [12] Ohliger A, Förster M, Kneer R. Torrefaction of beechwood: A parametric study including heat of reaction and grindability. *Fuel*. 2012;104:607-13.
- [13] Peduzzi E, Boissonnet G, Haarlemmer G, Dupont C, Maréchal F. Torrefaction modelling for lignocellulosic biomass conversion processes. *Energy*. 2014;Accepted.
- [14] Joshi Y, de Vries H, Woudstra T, de Jong W. Torrefaction: Unit Operation Modelling and Process Simulation. *Applied Thermal Engineering*. 2014;Accepted.
- [15] Tumuluru JS, Sokhansanj S, Wright CT, Boardman RD. Biomass torrefaction process review and moving bed torrefaction system model development. Idaho National Laboratory (INL); 2010.
- [16] Di Blasi C, Lanzetta M. Intrinsic kinetics of isothermal xylan degradation in inert atmosphere. *Journal of Analytical and Applied Pyrolysis*. 1997;40:287-303.
- [17] Jalan R, Srivastava V. Studies on pyrolysis of a single biomass cylindrical pellet—kinetic and heat transfer effects. *Energy Conversion and Management*. 1999;40(5):467-94.
- [18] Ratte J, Fardet E, Mateos D, Héry J-S. Mathematical modelling of a continuous biomass torrefaction reactor: TORSPYD™ column. *Biomass and Bioenergy*.

2011;35(8):3481-95.

[19] Li L, Ma W. Experimental study on the effective particle diameter of a packed bed with non-spherical particles. *Transport in porous media*. 2011;89(1):35-48.

[20] Pach M, Zanzi R, Björnbom E. Torrefied biomass a substitute for wood and charcoal. *Conference Torrefied biomass a substitute for wood and charcoal*, vol. 20.

[21] Arias B, Pevida C, Feroso J, Plaza M, Rubiera F, Pis J. Influence of torrefaction on the grindability and reactivity of woody biomass. *Fuel Processing Technology*. 2008;89(2):169-75.

[22] Khezami L, Chetouani A, Taouk B, Capart R. Production and characterisation of activated carbon from wood components in powder: cellulose, lignin, xylan. *Powder Technology*. 2005;157(1):48-56.

[23] Yang H, Yan R, Chen H, Lee DH, Zheng C. Characteristics of hemicellulose, cellulose and lignin pyrolysis. *Fuel*. 2007;86(12):1781-8.

[24] Pyle D, Zaror C. Heat transfer and kinetics in the low temperature pyrolysis of solids. *Chemical Engineering Science*. 1984;39(1):147-58.

[25] Chin K, H'ng P, Go W, Wong W, Lim T, Maminski M, et al. Optimization of torrefaction conditions for high energy density solid biofuel from oil palm biomass and fast growing species available in Malaysia. *Industrial Crops and Products*. 2013;49:768-74.

[26] Lee J-W, Kim Y-H, Lee S-M, Lee H-W. Optimizing the torrefaction of mixed softwood by response surface methodology for biomass upgrading to high energy density. *Bioresource Technology*. 2012;116:471-6.

[27] Bates RB. Modeling the coupled effects of heat transfer, thermochemistry, and kinetics during biomass torrefaction. Ph.D. Thesis: Massachusetts Institute of Technology, 2012.

[28] Bridgeman T, Jones J, Shield I, Williams P. Torrefaction of reed canary grass, wheat straw and willow to enhance solid fuel qualities and combustion properties. *Fuel*. 2008;87(6):844-56.

[29] McIntyre A, Plitt L. The interrelationship between Bond and Hardgrove grindabilities. *CIM Bulletin*. 1980;73:149-55.

[30] Dudgeon R. An aspen plus model of biomass torrefaction. Electric Power Research Institute (EPRI), Charlotte, NC. 2009.

[31] Syu F-S, Chiueh P-T. Process simulation of rice straw torrefaction. *Sustain Environ Res*. 2012;22(3):177-83.

[32] Park C, Zahid U, Lee S, Han C. Effect of process operating conditions in the biomass torrefaction: A simulation study using one-dimensional reactor and process model submitted. 2014.

[33] Sadaka S, Negi S. Improvements of biomass physical and thermochemical characteristics via torrefaction process. *Environmental Progress & Sustainable Energy*. 2009;28(3):427-34.