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Optimal condition of torrefaction for high energy density solid fuel of fast growing tree species

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Abstract−The torrefaction properties of Acacia (*Acacia mangium*) and Albasia (*Paraserianthes falcataria*) were investigated by response surface methodology. Torrefaction was performed at 220-280 °C for 20-80 min depending on severity factor. Carbon content in the torrefied biomass increased with severity factor, whereas hydrogen and oxygen contents decreased both biomass. The calorific value of torrefied Acacia ranged from 20.03 to 21.60 MJ/kg, suggesting that the energy contained in the torrefied biomass increased by 5.09 to 13.62%, when compared with that in the untreated biomass. However, the calorific value of Albasia was relatively low, compared to that of torrefied Acacia. The weight loss of Albasia was higher than that of Acacia under a given torrefaction condition. The reaction temperature for torrefaction was an important factor to obtain high energy yield, whereas the effect of time was considerable lower. High temperature and short torrefaction time is required to obtain the highest energy yield from torrefaction using Acacia and Albasia.

Keywords: Torrefaction, Response Surface Methodology, Severity Factor, Calorific Value, Weight Loss, Energy Yield

INTRODUCTION

There is a growing interest in the use of solid, liquid and gaseous biofuels for energy. Large amounts of lignocellulosic biomass could potentially be made available for use as a source of energy. In particular, lignocellulosic biomass can be converted to heat and power. Among heat and power sources, wood pellets have a higher energy density and lower transport cost, compared to unprocessed wood. Lignocellulosic biomass has a lower risk of corrosion and fouling, due to the low ash content, compared to agricultural biomass, such as rice straw and wheat straw [1]. However, wood pellets have disadvantages, such as a relatively lower energy density, and higher moisture content, than that of fossil fuels. Wood pellets do not provide consistent assessments of calorific value and ash content because pellets are produced from different kinds of wood. To overcome these disadvantages, torrefaction has been proposed as a process for upgrading biomass by thermal treatment [2,3].

Torrefaction is a thermal pretreatment process, whereby raw material is heated in an inert or nitrogen atmosphere to avoid combustion. Typically, it utilizes a temperature range of 200 to 300 °C and torrefaction times of a few minutes to 1 h [4]. These conditions provide a hydrophobic environment and high calorific value for the biomass due to removal of hydroxyl groups during thermal treatment [5]. Therefore, the torrefied biomass provides suitable chemical and physical characteristics for long-distance transportation and long-term storage. In addition, torrefaction results in improved grindability of the biomass to produce pellets, as hemicelluloses are degraded during torrefaction [6].

In general, torrefaction induces 10% of energy loss and 30% of weight loss from untreated biomass. Based on energy and weight loss in the biomass, the energy yield is defined as the ratio of the increase of energy value to the decrease of biomass weight. Therefore, torrefied biomass should be evaluated in terms of energy yield.

Acacia (*Acacia mangium*) and Albasia (*Paraserianthes falcataria*) are representative fast growing tree species. These species are the most widely planted trees in Asia and Africa, and provide paper pulp, sawn timber, and fuel wood for industry and agriculture [7]. In addition, these species acclimate well to barren soil and a wide range of soil pH; thus, their planting is increasing worldwide [8]. Therefore, fast growing tree species have been used for energy resources [9,10].

Changes occur in the chemical and physical properties of the fast growing tree species when torrefaction is performed in the temperature range of 200-300 °C with different torrefaction time. Weight loss and increase of energy value is an important factor for optimizing the design and operation of a biomass torrefaction plant. Therefore, understanding of each of the process variables for optimization is required. In this study, torrefaction was performed for these two fast growing tree species to improve energy yield. The chemical and physical properties of the torrefied biomass, and the optimal conditions to obtain a high energy yield from the torrefied biomass, were investigated by response surface methodology.

EXPERIMENTAL

1. Materials

Acacia (*Acacia mangium*) and Albasia (*Paraserianthes falcataria*)

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Table 1. The 2^2 factorial design with four axial points and three replicates in the central point matrix employed for two independent variables

Sample no.	Variables		Coded levels		Severity factor (SF)
	Time (min) X_1	Temperature (°C) X_2	Time x_1	Temperature x_2	
1	50	250	0	0	6.12
2	50	250	0	0	6.12
3	50	250	0	0	6.12
4	70	270	1	1	6.85
5	70	230	1	−1	5.67
6	30	270	−1	1	6.48
7	30	230	−1	−1	5.30
8	80	250	1.4	0	6.32
9	50	280	0	1.4	7.00
10	20	250	−1.4	0	5.72
11	50	220	0	−1.4	5.23

chips as lignocellulosic biomass were purchased from Green-Korea Inc. (Seoul, Korea). The wood chips were screened to a size of 10-30 mm using sieves (9.5 and 31.5 mesh). The mean particle size was 20(5.23)×12.5(5.09)×3.94(1.95) mm in width, length, and thickness, respectively. Wood chips were dried to below 10% moisture content before being used for experiments.

2. Torrefaction Process

The torrefaction conditions were created with a 2^2 factorial design, which included four axial points and three replicates in the central point (Table 1). Torrefaction was performed at 220-280 °C for 20-80 min, depending on severity factor. The wood chips were dried at 105 °C for 24 h before torrefaction to remove water remaining in the biomass. Dried wood chips (500 g) were placed in a batch reactor with a temperature controller, and the reactor was sealed to increase temperature with stirring. Nitrogen was purged into the reactor at a flow of 2 L/min to maintain the anoxic condition during torrefaction. After reaching the desired reaction time and temperature, the heater was turned off, and the reactor was left to cool to room temperature. A severity factor (SF) was used to integrate the effects of reaction times and temperature into a single variable during torrefaction [11]. The SF used in this study was defined as

$$SF = \mathrm{Log}\left[t \cdot \exp\left(\frac{T_H - T_R}{14.75}\right)\right] \quad (1)$$

where t is the reaction time of the torrefaction in minute, T_H the reaction temperature in °C, and T_R is the reference temperature, most often 100 °C [11].

3. Proximate, Elemental Analysis and Calorific Value of Biomass

Proximate, elemental and calorific value analyses of the untreated and torrefied biomass were performed, according to the wood pellet quality standard [12]. Calorific value was measured with a Parr 6400 isoperibol oxygen bomber calorimeter (Parr Instrument Inc. Moline, IL, USA). The elemental analysis was carried out using an elemental analyzer (CE Instruments Inc., Rodano, Italy).

4. Grinding Characteristics

The size distribution of the torrefied biomass was investigated by grinding. The torrefied biomass was placed into a four-blade cracker, and grinding was performed at 24,000 rpm. The ground biomass was passed through three different sieves (100, 200 and 325 mesh), and these were separated into four differently sized parts. The different parts were used for the geometric mean diameter of ground the torrefied biomass, which was measured as the size at 50% cumulative distribution.

5. Surface Response Analysis

A 2^2 factorial design was used for analyzing the torrefaction characteristics of the lignocellulosic biomass. Time (X_1, 20-80 min) and reaction temperature (X_2, 220-280 °C) were used as the independent variables during torrefaction. Calorific value, weight loss of the biomass, and energy yield were used as the dependent output variables. Second-degree polynomials were calculated using Design-Expert ver. 8.0.1 software (Stat-Ease Inc., Minneapolis, MN, USA) to estimate the response of the dependent variables. Factors from each run were appropriately coded to real independent variables x_1 and x_2 for the statistical model, as shown in Table 1. The independent variables were [(condition of the run−condition at the central point)/(step change of the factor)]. Therefore, x_1 corresponded to [(time−50)/20], and x_2 was [(temperature−250)/20].

RESULTS AND DISCUSSION

1. Proximate and Elemental Properties of the Torrefied Biomass

The changes in moisture and ash content of the torrefied biomass are shown in Table 2. The moisture content decreased as the SF increased in both biomass. This is indicative of the reduction in hydroxyl groups during torrefaction [13]. Ash content increased slightly under torrefaction conditions. These behaviors have been observed in other biomass as typical features of torrefaction [13,14].

The elemental analysis of the untreated and the torrefied biomass is shown in Table 3. The carbon content in the torrefied biomass increased as the SF, while hydrogen and oxygen content decreased in both biomass. This may have induced a decrease in the H/C and O/C ratios. The H/C atomic ratio was plotted against the O/C atomic ratio for the untreated and torrefied biomass (Fig. 1).

Table 2. Moisture and ash contents of the torrefied Acacia (AC) and Albasia (AL)

SF	Temperature (°C)	Time (min)	Moisture (%)		Ash (%)	
			AC	AL	AC	AL
5.23	220	50	2.60	2.18	0.62	0.31
5.30	230	30	2.66	2.19	0.72	0.47
5.67	230	70	2.45	2.11	1.05	0.57
5.72	250	20	2.17	2.00	0.87	0.41
6.12	250	50	2.10	1.46	0.84	0.40
6.32	250	80	2.11	1.25	0.89	0.69
6.48	270	30	2.08	1.10	1.32	0.58
6.85	270	70	2.05	1.10	0.98	0.59
7.00	280	50	1.95	1.08	1.28	0.53
Raw material	-	-	5.34	7.83	0.68	0.39

Table 3. Elemental analysis of the torrefied Acacia (AC) and Albasia (AL)

SF	Temperature (°C)	Time (min)	C (%)		H (%)		O (%)		N (%)	
			AC	AL	AC	AL	AC	AL	AC	AL
5.23	220	50	49.91	47.80	5.91	6.04	27.38	26.52	0.71	0.08
5.30	230	30	49.52	47.83	5.93	5.96	27.74	26.13	0.24	0.05
5.67	230	70	50.76	48.41	5.99	5.80	27.70	25.22	0.21	0.03
5.72	250	20	51.08	49.39	5.84	5.77	27.13	25.93	0.22	0.13
6.12	250	50	51.88	50.15	5.80	5.83	27.37	24.69	0.46	0.09
6.32	250	80	52.01	50.56	5.76	5.82	26.44	23.61	0.29	0.12
6.48	270	30	53.72	51.21	5.77	5.76	25.70	23.79	0.26	0.08
6.85	270	70	53.81	52.00	5.83	5.74	25.58	23.54	0.29	0.12
7.00	280	50	54.71	52.78	5.71	5.54	24.86	23.21	0.26	0.07
Raw material	-	-	48.11	45.50	6.15	5.79	28.00	26.59	0.56	0.19

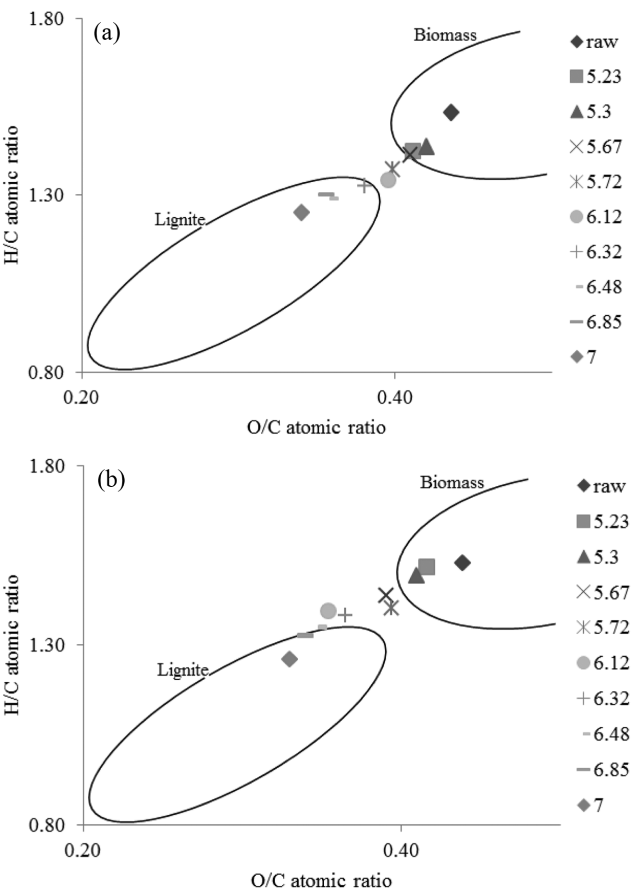

Fig. 1. H/C vs. O/C atomic ratio of untreated and torrefied biomass, depending on torrefaction severity ((a) Acacia, (b) Albasia).

Comparative solid fuel data were obtained from Van Loo and Koppejan [15]. There are three types of reaction during torrefaction: dehydration, decarbonation, and demethanation [16]. The tendency revealed in this study concerned dehydration, as the H/C atomic ratio was proportional to the O/C atomic ratio. Therefore, dehydration mainly occurred during torrefaction. The torrefied biomass at high SF had lower levels of H/C and O/C atomic ratios than those of low SF and the untreated biomass, regardless of the studied biomass. This was due to the release of hydrogen and oxygen generated from water and carbon dioxide. Torrefaction was highly effective on Acacia, compared to that of Albasia. Untreated Acacia plotted an H/C ratio of 1.53 and O/C ratio of 0.44, whereas the torrefied Acacia at SF 7 was 1.25 and 0.34, respectively. However, torrefied Albasia showed relatively higher H/C and O/C ratio compared to that of torrefied Acacia. This finding indicates that the elemental composition of torrefied Acacia at high SF was similar to that of lignite coal.

2. Analysis of Calorific Value and Weight Loss Based on SF

Table 4 shows the calorific value, weight loss and energy yield for both biomass over a range of SF. The experimental calorific value increased gradually with SF. This result was similar to the change in carbon content in the biomass during torrefaction. This result is also in agreement with the report of Lee et al. [17]. The calorific value of torrefied Acacia chips was 21.60 MJ/kg at an SF of 7.0, which was an increase of 13.62%, compared to that of the untreated biomass. The calorific value for Albasia was 21.62 MJ/kg at an SF of 7.0 (showing an increase of 10.64%, compared to the untreated biomass). Weight loss of a biomass is a good indicator of the degree of torrefaction [18]. The weight loss of the biomass increased with increasing SF. At low SF, weight loss is mainly due to thermal degradation of hemicelluloses, and the release of volatile components in the biomass. However, weight loss of biomass at high SF is attributed to the simultaneous reactions of hemicelluloses and cellulose [19].

The energy yield showed an inverse proportion to the calorific value and weight loss, based on SF. The energy yield of torrefied Acacia was relatively high, compared with that of Albasia over a range of SF. This occurred because the biomass had a relatively high calorific value and low weight loss following torrefaction.

3. Response Surface Analysis for Calorific Value, Weight Loss, and Energy Yield

The calorific value, weight loss, and energy yield obtained from the biomass depended on the torrefaction conditions, which are listed in Table 1. The calorific value of torrefied Acacia was 20.03-21.60 MJ/kg, depending on the torrefaction conditions. This result suggests that the energy content in the torrefied biomass increased by 5.09 to 13.62%, compared to that of the untreated biomass. The calorific value of torrefied Albasia was 19.73-21.62 MJ/kg, which

Table 4. Calorific value and weight loss of the torrefied Acacia (AC) and Albasia (AL)

SF	Temperature (°C)	Time (min)	Calorific value (MJ/kg)		Weight loss[a] (%)		Mass yield[b] (%)		Energy yield (%)	
			AC	AL	AC	AL	AC	AL	AC	AL
5.23	220	50	20.03	19.73	2.11	1.51	97.89	98.49	103.14	99.45
5.3	230	30	20.13	19.77	2.58	3.24	97.42	96.76	103.16	97.90
5.67	230	70	20.58	19.94	3.58	4.26	96.42	95.74	103.37	97.70
5.72	250	20	20.51	20.19	6.15	8.50	93.85	91.50	101.26	94.54
6.12	250	50	20.68	20.26	9.99	11.02	90.01	88.98	97.92	92.26
6.32	250	80	20.86	20.46	12.92	13.28	87.08	86.72	95.55	90.80
6.48	270	30	21.10	20.65	16.40	14.61	83.60	85.39	92.79	90.24
6.85	270	70	21.54	20.85	19.33	19.41	80.67	80.59	91.41	85.99
7	280	50	21.60	21.62	24.28	26.03	75.72	73.97	86.04	81.84
Raw material	-	-	19.01	19.54	-	-	100.00	100.00	100.00	100.00

[a]Weight loss (%)={(mass of untreated biomass-mass of torrefied biomass)/(mass of untreated biomass)}*100
[b]Mass yield (%)=100−weight loss of biomass

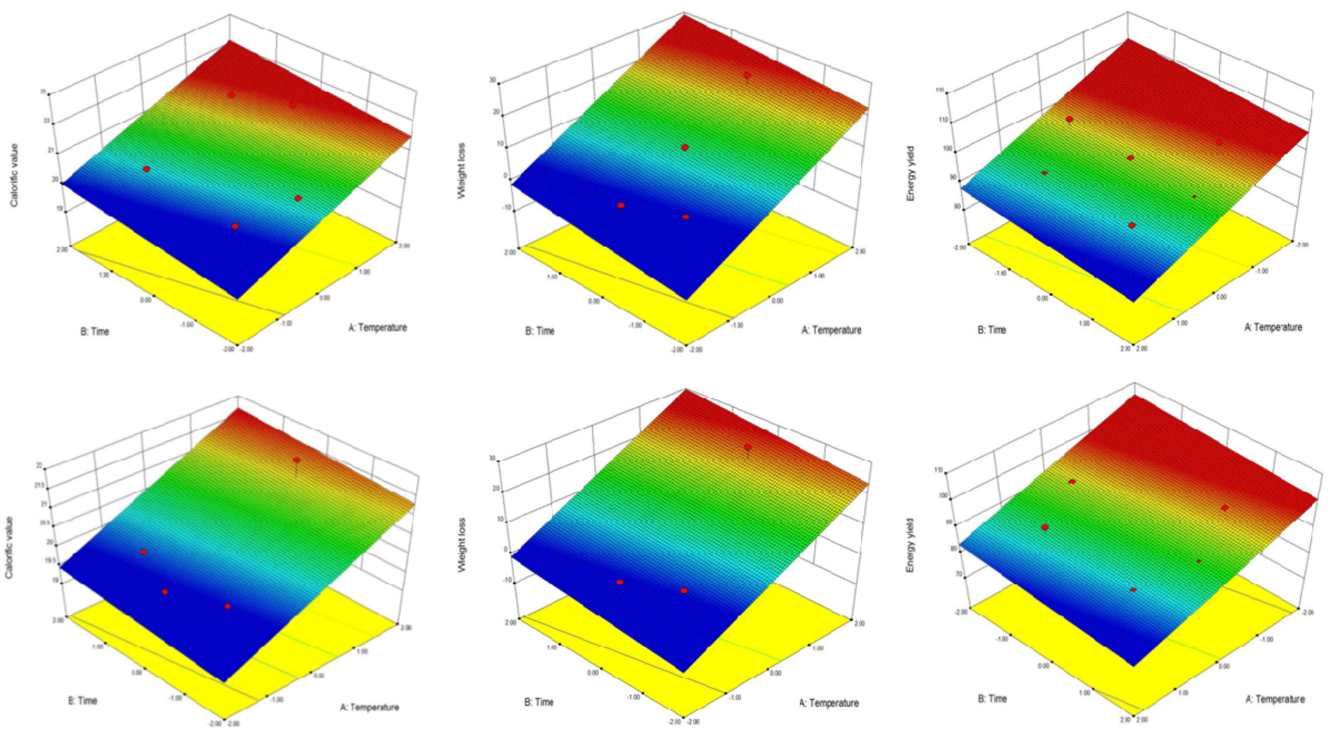

Fig. 2. Response surface and contour plot of temperature vs. reaction time, on the calorific value, weight loss and energy yield during torrefaction.

was an increase from 0.97 to 10.64%. The calorific value was affected more by the reaction temperature than by time, regardless of the studied biomass, as shown in Fig. 2.

Based on the calorific value, the response surface determined that the optimal torrefaction condition yielded the highest calorific value. The modeling results are provided in Fig. 2. A three-dimensional plot showed the predicted peak in calorific value, at optimum condition. Y_1 is the predicted calorific value (MJ/kg). The results of the analysis of variance (ANOVA) in Eqs. (2) and (3) are presented in Tables 5 and 6, respectively; x_1 and x_2 denote reaction time and temperature, respectively. This model was significant at the 99% confidence level, and the p-values for temperature and reaction time were close to 0.

$$Y_1=20.38+0.10\,x_1+0.62\,x_2-0.30\,x_1\,x_2-0.19\,x_1^2+0.31\,x_2^2 \quad (2)$$

$$Y_1=20.88+0.23\,x_1+0.94\,x_2 \quad (3)$$

Weight loss of the biomass during torrefaction is shown in Fig. 2. According to the model, temperature and time squared were the only significant parameters. The strength of the effect of the two significant process parameters on the weight loss of biomass was better revealed by the surface plot. The reaction temperature had

Table 5. Analysis of variance (ANOVA) for the adjusted model for the calorific value, weight loss and energy yield of Acacia during torrefaction

Source	Sum of squares	Degrees of freedom	Mean square	F-value	P-value (Prob>F)
Calorific value					
Model	4.53	5.00	0.91	16.51	0.0040
Residual	0.27	5.00	0.05		
Lack of fit	0.23	3.00	0.08	3.11	0.2528
Purr error	0.05	2.00	0.02		
Corrected total	4.81	10.00			
Temp.	3.09	1.00	3.09	56.22	0.0007
Time	0.08	1.00	0.08	1.41	0.2883
Time×Temp.	0.37	1.00	0.37	6.67	0.0493
$(Temp.)^2$	0.53	1.00	0.53	9.68	0.0265
$(Time)^2$	0.20	1.00	0.20	3.55	0.1181
					Adjusted R-square: 0.8858
Weight loss					
Model	486.75	2.00	243.37	80.66	<0.0001
Residual	24.14	8.00	3.02		
Lack of fit	22.20	6.00	3.70	3.81	0.2226
Purr error	1.94	2.00	0.97		
Corrected total	510.89	10.00			
Time	463.95	1.00	463.95	153.76	<0.0001
Temp.	22.80	1.00	22.80	7.55	0.0251
					Adjusted R-square: 0.9409
Energy yield					
Model	264.52	3.00	88.17	27.91	0.0003
Residual	22.11	7.00	3.16		
Lack of fit	20.98	5.00	4.20	7.39	0.1235
Purr error	1.14	2.00	0.57		
Corrected total	286.63	10.00			
Time	235.02	1.00	235.02	74.40	<0.0001
Temp.	12.97	1.00	12.97	4.11	0.0824
Time×Temp.	16.52	1.00	16.52	5.23	0.0560
					Adjusted R-square: 0.8898

an impact on biomass weight loss, during torrefaction of the studied biomass. The temperature ranged from 220 to 280 °C in this study. In this range, most of the hemicelluloses, and some of the cellulose and lignin content in the biomass were degraded during torrefaction [19]. Weight loss during torrefaction depended highly on reaction temperature, rather than time. Tables 5 and 6 show the ANOVA of the quadratic model adjustment, where the total error was classified into lack-of fit, and pure error. The F value estimated using the experimental data corresponded to the total residual and lack-of-fit values, and was lower than the tabular F value. This result indicates that the models were significant in the region studied. The model calculated for the weight loss of biomass (Y_2) is provided in the equations. Eqs. (4) and (5) are for Acacia and Albasia, respectively.

$$Y_2=10.66+1.68x_1+7.62\,x_2 \tag{4}$$

$$Y_2=11.31+1.57x_1+7.56\,x_2 \tag{5}$$

The results of the energy yield, shown in Fig. 2, were calculated from the mass yield and calorific value using Eq. (6), and are expressed as a percentage of the energy content of untreated dry biomass: $m_{torrefied}$ denotes the dry mass of torrefied biomass; $m_{initial}$, the dry mass of untreated biomass; $E_{torrefied}$, the specific energy content of biomass after torrefaction; and $E_{initial}$, the specific energy content of biomass before torrefaction.

$$\text{Energy yield (\%)}=\left(\frac{\text{Mtorrefied}}{\text{Minitial}}\right)\times\left(\frac{\text{Etorrefied}}{\text{Einitial}}\right)\times 100 \tag{6}$$

The energy yield per untreated biomass indicates the total energy preserved in the torrefied biomass. Reaction temperature had the highest impact on energy yield of the torrefied biomass, whereas the effect of time was considerable lower. The energy yields of torrefied Acacia and Albasia were fit to the response surface model provided in equations (not shown), in order to analyze the effect of the torrefaction factors on energy yield. As shown in Tables 5

Table 6. Analysis of variance (ANOVA) for the adjusted model for the calorific value, weight loss and energy yield of Albasia during torrefaction

Source	Sum of squares	Degrees of freedom	Mean square	F-value	P-value (Prob>F)
Calorific value					
Model	7.51	2.00	3.75	152.15	<0.0001
Residual	0.20	8.00	0.02		
Lack of fit	0.20	6.00	0.03	31.50	0.0311
Purr error	0.00	2.00	0.00		
Corrected total	7.71	10.00			
Temp.	7.08	1.00	7.08	286.84	<0.0001
Time	0.43	1.00	0.43	17.46	0.0031
					Adjusted R-square: 0.9680
Weight loss					
Model	487.91	2.00	243.95	62.95	<0.0001
Residual	31.00	8.00	3.88		
Lack of fit	27.95	6.00	4.66	3.06	0.2669
Purr error	3.05	2.00	1.52		
Corrected total	518.91	10.00			
Temp.	468.13	1.00	468.13	120.80	<0.0001
Time	19.78	1.00	19.78	5.10	0.0538
					Adjusted R-square: 0.9253
Energy yield					
Model	151.88	2.00	75.94	18.44	0.0010
Residual	184.82	10.00	0.00		
Lack of fit	29.66	6.00	4.94	3.01	0.2705
Purr error	3.29	2.00	1.64		
Corrected total	184.82	10.00			
Temp.	147.92	1.00	147.92	35.92	0.0003
Time	3.96	1.00	3.96	0.96	0.3557
					Adjusted R-square: 0.7772

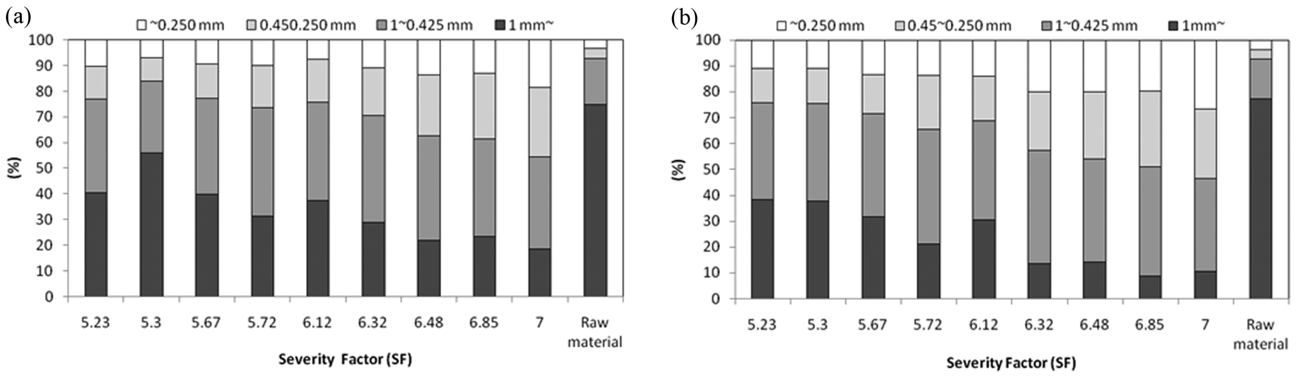

Fig. 3. Size distribution after grindability test of untreated and torrefied biomass at various SF ((a) Acacia, (b) Albasia).

and 6, equations were in good correlation with the actual data, as justified by the relatively high R-squared value. However, an accurate analysis of optimal condition for energy yield was difficult at a given torrefaction condition. The reason is that torrefaction needs a wider range of torrefaction time to obtain the highest energy yield.

4. Particle Size Distribution of the Torrefied Biomass by Grinding

The effect of the torrefaction SF with geometric mean particle size of ground torrefied biomass, through three different sieves, is shown in Fig. 3 and Fig. 4. The mean particle size of ground torrefied biomass decreased with increasing SF, regardless of the studied biomass. In particular, the mean particle size of ground torrefied Albasia decreased dramatically with increasing SF. Torrefaction severity plays an important role determining the size distribution. A similar trend in particle size distribution has also been observed

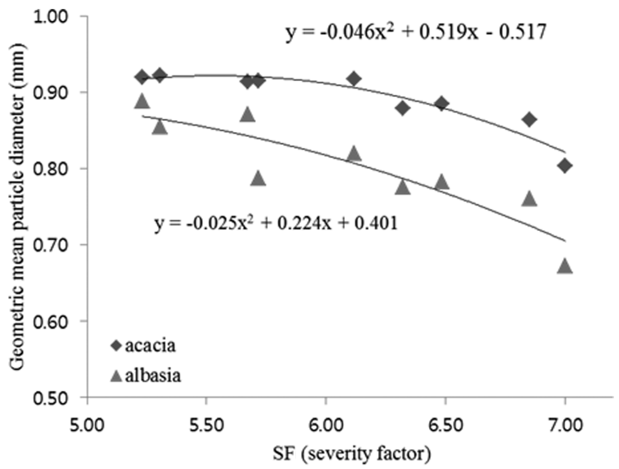

Fig. 4. Profiles of geometric mean particle size of ground torrefied biomass on the torrefaction severity factor.

in other reports [2,14]. Among the studied biomass, the mean particle sizes of Albasia at all severity factors were slightly lower than those of Acacia. Overall, the torrefied biomass produced a relatively smaller mean particle size, compared to that of untreated biomass, as the torrefied biomass was easily broken. This behavior was directly supported by the lower grinding energy consumption of torrefied biomass for pellet production.

CONCLUSIONS

The torrefaction properties of Acacia and Albasia were investigated depending on the reaction temperature and time. Reaction temperature of both biomass had a strong impact on the energy yield of the torrefied biomass, whereas the effect of time was considerably less. Between two biomass, Acacia had relatively higher energy yield, compared to that of Albasia, due to high calorific value and low weight loss during torrefaction. According to the response surface analysis of energy yield, high temperature and short torrefaction time both biomass were appropriate for biomass torrefaction and producing high energy density solid fuels. The results could be used as guide for the production of high energy density solid fuel from torrefied biomass of fast growing tree species.

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