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Effect of Woody Biomass Type on Gasification Rate of Char
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Biomass has been recognized as a renewable energy source alternative to fossil fuels, because it doesn't increase CO₂ amount in the atmosphere. Among various ways of woody biomass use, IGCC power generation is known as the most possibly efficient technology. In the process, biomass is firstly pyrolyzed and the produced volatile and char are secondary converted; hence it is necessary to know the production process and yields of pyrolysis gas, tar, producing char and char gasification kinetics for effective energy conversion. In this study, many woody biomass species (four softwoods, nine hardwoods and four barks) were pyrolyzed at 1000 °C at the rapid heating rate up to around 1000 °C /s. Then the produced char is gasified in an experimental fluidized bed. After that, the produced char was gasified by CO₂ at 1000°C. We computed char conversion and its rate from CO production rate with time and conversion rate was plotted against conversion. Different gasification rate curves are found for various biomass samples. The gasification rate of char was evaluated by the first order rate constant based on the volume reaction model. Hardwood char gasification rates were faster than those of softwood samples. Most of the gasification rates of char from the bark samples were nearly same as those of woody part samples. As for the hardwood, the char produced from biomass with lower compressive strength generally gave faster reaction rate.

バイオマスは大気中の二酸化炭素濃度を増やさない、化石燃料の代替となりうる再生可能エネルギーである。木質バイオマス使用の様々な方法の中で、IGCC発電は最も高効率技術として期待されている。本プロセスでは、最初に熱分解により熱分解ガスとチャーが生成され、次にチャーがガス化される。そのため、エネルギー変換の高効率化のためには、熱分解ガス、タール、チャーの生成挙動や収率、チャーのガス化速度を知る必要がある。本研究では多くの樹種（針葉樹4種、広葉樹9種、バーク4種）を流動層を用いて 1000°C /s 程度までの高速昇温で 1000°Cで熱分解した。その後生成チャーを 1000°Cでガス化をした。我々は転化率とその変化速度をCO生成速度の経時変化から計算し、両者の関係を図示した。バイオマス種により様々なガス化速度曲線が得られた。チャーのガス化速度は体積反応モデルによる一次反応速度定数を用いて評価した。広葉樹チャーのガス化速度は針葉樹にくらべ速かった。ほとんどの樹種について、バークチャーのガス化反応性は本部のそれとほぼ同じだった。一般的に広葉樹について、低い圧縮強さを有するバイオマスから得たチャーほど速い反応速度を与えた。

Key Words

Woody biomass char, Gasification rate, Fluidized bed, Compressive strength

1. Introduction

Biomass has been recognized as a renewable energy source alternative to fossil fuels. So many conversion processes have been proposed for the biomass energy use as energy source. Most convenient way is combustion using fixed beds or moving beds. However there remain various problems of lower efficiency and indoor pollution.

In order to solve these problems, the use of large scale high temperature continuous processes is suggested as one direction. In these reactors, the biomass is fed continuously and is usually rapidly heated in the reactor.

In our previous papers, the basic behavior of biomass introduced to a bed fluidized under the nitrogen atmosphere was investigated. By using a fluidized bed, it was demonstrated that *in situ* gasification of char produced at rapid heating rate is possible¹⁾. We also applied this to the

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biomass pyrolysis²⁾. We found that only under the conditions of rapid pyrolysis between 600 and 1000 °C, porous alumina particles as the bed material adhered on the surface of the obtained char³⁾. Zhang *et al.* reported that the char yields by rapid pyrolysis of Hinoki cypress sawdust in a lab-scale drop-tube furnace (DTF) was decreased above 1100°C, which was suggested to be attributed to the enhancement of the gasification reactions between solid products (char and coke) and reactive gases⁴⁾. We also reported that char yield from *Eucalyptus camaldulensis* was influenced by the pyrolysis temperature and heating rate. Under rapid heating rate, char yield was smaller than that under the slow heating and almost constant at about 15%, between 500 and 900°C, while it was decreased at higher temperature than 900°C²⁾⁵⁾.

Hence the pyrolysis condition of temperature and heating rate should give serious effects on the downstream processes such as combustion and gasification.

Dall'Ora *et al.* found that the char oxidation reactivity in TG was increased by the fast pyrolysis conducted in a separate reactor and decreased as pyrolysis temperature increased up to 1573 K⁶⁾.

Among the various high temperature processes, taking the usability of the electricity into the account, the electricity generation by combined cycle using gas produced from biomass is thought to be promising. Char gasification is the last and the slowest step of biomass conversion into the gas. The gasification rate of char is known to be slow and it limits process efficiency. Hence the information on gasification rate is needed for design of efficient energy conversion processes.

For the gasification, though so many papers reported the gasification kinetics and morphology change during the gasification, most of the studies were conducted at relatively low temperature and for chars produced at the low heating rate. Furthermore most of the char used for gasification reactivity test was prepared in a separate reactor. The various kinds of biomass species have so far been tested by many researchers while under different conditions, so the unified comparison between various kinds of biomass under the unified condition was difficult.

Zhang *et al.* reported that the experiments with continuous or sudden supply of steam were conducted using a fluidized-bed/fixed-bed quartz reactor⁷⁾. The structural features of chars were then characterized using Fourier transform (FT)-Raman spectroscopy. The effects of the biomass particle size on char structure were minimal when the biomass was heated slowly while being in continuous contact with steam. However the temperature range was up to 900 °C and pyrolysis was conducted under the slow heating of 10°C/min.

Tinaut *et al.* proposed a one-dimensional stationary model of biomass gasification in a fixed bed downdraft gasifier, in which biomass was heated slowly and gasified at low temperature⁸⁾.

Fushimi *et al.* found that a higher heating rate (up to 100 °C/s) substantially increased the reaction rate of lignin char in steam gasification because porous char was produced during devolatilization due to rapid evolution of volatiles⁹⁾. However the heating rate was not so high compared with commercial fluidized beds, estimated as up to 1000°C/s, and temperature was as low as 800°C.

Okumura and Okazaki found that the gasification rate of biomass char increased as pressure increased between 700-1100°C, while the heating rate (up to 100°C/s) was not so high compared with commercial fluidized beds and woody biomass was not tested¹⁰⁾.

Marquez-Montesinos *et al.* reported the CO₂ gasification kinetic of grapefruit skin carbonized at 700°C in a conventional quartz tube furnace, with and with steam at the temperature between 725 and 800°C¹¹⁾.

As are shown in the above, many experimental results on biomass gasification have so far been reported but most of them were under the slower heating rate than that in commercial continuous fluidized beds or entrained beds and the data at the higher temperature than 1000°C were not so many.

Yuan *et al.* prepared the char of three typical biomasses, rice straw char (RS char), chinara leaves char (CL char), and pine sawdust char (PS char) in a high-frequency furnace, which could efficiently reduce secondary reactions under rapid pyrolysis conditions at 800 to 1200 °C and the char was isothermally gasified in a thermogravimetric analyzer (TGA) under a CO₂ atmosphere¹²⁾. They found that gasification rates decreased with the increasing pyrolysis temperature.

As mentioned above some paper reported the gasification results of char produced under rapid heating rate. However the produced char was generally taken out after the char was cooled and then it was gasified. Furthermore, in most of the biomass gasification experiments, a limited number of biomass species were tested and the results are difficult to be directly compared with the results under the different experimental condition in other reports.

In the present study, the authors conduct *in situ* gasification of char produced under the high heating rate in an experimental fluidized bed without cooling, by changing the gas from nitrogen to 25% CO₂. Seventeen biomass samples are gasified and their gasification rates are measured under the unified condition of the above gas

concentration and temperature of 1000°C which is relatively high compared with conditions of previous papers.

2. Experimental

2.1 Biomass Samples

The biomass types we used were four softwoods (Japanese red pine (MATSU), *Chamaecyparis obtuse* (HINOKI), *Cryptomeria japonica* (SUGI), *Cedrus deodara* (HIMARAYASUGI)**), four evergreen hardwoods (*Eucalyptus camaldulensis* (YUKARI)*, *Quercus phillyraeoides* (UBAMEGASHI), *Quercus myrsinaefolia* (SHIRAKASHI)**), *Rhizophora mucronata* (OBAHIRUGI)) five deciduous hardwoods (*Celtis sinensis* var *japonica* (ENOKI), *Quercus serrata* (KONARA), *Aphananthe aspera* (MUKU), *Zelkova* (KEYAKI), *Quercus acutissima* (KUNUGI)) and four barks (*Cedrus deodara* (HIMARAYASUGI), *Rhizophora mucronata* (OBAHIRUGI)***, *Celtis sinensis* var *japonica* (ENOKI), *Quercus serrata* (KONARA)).

They were supplied as follows: * from our experimental site¹³⁾; ** from Seikei University at Tokyo; *** from mangrove plantation site in Thailand¹⁴⁾; others from lumbermill factory of Saitama Mokuzai Tip Co-op, in Nagatoro. Their properties are shown in Table 1.

2.2 Experimental Setup and Condition

Fig. 1 shows the details of the fluidized bed device used. Experimental conditions were as follows: static bed

height, 50 mm; particle density, 1170 kg/m³; mean diameter of alumina bed particles, 215 μm; range of particle diameter of biomass, 1.7< d_p <2.8 mm (by sieving); gas flow rate, 6 U_{mf} [m/s]; biomass amount, 250 mg; pressure, 1 atm (around 1.01×10^5 Pa); gasification temperature, 1000°C; holding time, 10 min.

When biomass samples (1.7< d_p <2.8 mm) are dropped into the fluidized bed kept at 1000°C under nitrogen atmosphere, the biomass samples are heated rapidly with vividly moving bed materials. According to the

Fig. 1 Fluidized bed devices

Table 1 Property of woody biomass samples

Group	Biomass species (Popular name)	Biomass species (Scientific name)	Biomass species (in Japanese)	Proximate analysis (wt % dry)			Ultimate analysis (wt % dry)			
				Volatile Matter	Fixed Carbon	Ash	C	H	N	O (dif.)
softwood	Japanese pine	<i>Pinus japonica</i>	(MATSU)	82.18	17.25	0.57	49.40	6.33	0.14	43.56
	Japanese cypress	<i>Chamaecyparis obtusa</i>	(HINOKI)	84.75	15.08	0.17	48.33	5.91	0.00	45.59
	Japanese cedar	<i>Cryptomeria japonica</i>	(SUGI)	83.39	16.41	0.20	48.69	6.09	0.00	45.02
	Cedar	<i>Cedrus deodara</i>	(HIMARAYA SUGI)	83.30	16.40	0.26	47.97	5.58	0.10	46.09
evergreen hardwood	River red gum	<i>Eucalyptus camaldulensis</i>	(YUKARI)	78.92	18.50	2.58	48.02	5.99	0.37	43.04
	Ubamegashi	<i>Quercus phillyraeoides</i>	(UBAMEGASHI)	80.10	17.90	1.95	45.71	6.02	0.26	46.06
	White oak	<i>Quercus myrsinaefolia</i>	(SHIRAKASHI)	82.90	14.70	2.40	43.99	5.72	0.18	47.71
	Red mangrove	<i>Rhizophora mucronata</i>	(OBAHIRUGI)	81.16	17.95	0.89	45.18	6.03	0.10	47.80
deciduous hardwood	Nettle tree	<i>Celtis sinensis</i> var <i>japonica</i>	(ENOKI)	82.20	16.80	1.08	45.25	6.20	0.29	47.18
	Konara	<i>Quercus serrata</i>	(KONARA)	84.30	15.00	0.69	45.12	4.56	0.26	49.37
	Aphananthe	<i>Aphananthe aspera</i>	(MUKU)	80.40	17.00	2.62	46.23	6.24	0.51	44.40
	Japanese zelkova	<i>Zelkova serrata</i>	(KEYAKI)	79.70	19.18	1.12	47.42	5.50	0.15	45.81
	Sawthorn oak	<i>Quercus acutissima</i>	(KUNUGI)	83.20	16.20	0.59	45.79	6.04	0.23	47.35
bark	Cedar	<i>Cedrus deodara</i>	(HIMARAYA SUGI)	-	-	1.02	48.72	5.93	0.28	44.05
	Red mangrove	<i>Rhizophora mucronata</i>	(OBAHIRUGI)	-	-	4.59	42.59	5.30	0.52	47.00
	Nettle tree	<i>Celtis sinensis</i> var <i>japonica</i>	(ENOKI)	-	-	13.52	42.97	5.62	1.09	36.80
	Konara	<i>Quercus serrata</i>	(KONARA)	-	-	8.42	46.18	5.91	0.72	38.77

-: NA (because of their small sample amount)

numerical calculation of temperature profile inside the particle assuming the spherical shape with constant surface temperature of 1000 °C, we got rapid heating rate of averaged temperature inside the particles up to around 1000°C/s (at the first stage of heating)¹⁵⁾.

After 10 minutes' pyrolysis, the *in situ* produced char was started to be gasified by introducing 25% CO₂ gas diluted with nitrogen. We sampled produced gas using a 10 mL syringe at 5 s to 15 min intervals for 60 min and CO concentration was analyzed by a gas chromatograph. When oxygen was detected the nitrogen and CO concentrations were corrected assuming that it was regarded to be caused by the leaking-in of air. The CO production rate was given by multiplying the molar nitrogen flow rate by concentration ratio of CO to nitrogen, and plotted against the time.

2.3 Analyses of time variation of CO production

From the time variation curve of an experiment, we computed char conversion and conversion rate assuming $C + CO_2 \rightarrow 2CO$. Hence the mole of carbon converted was given as the half of the accumulated value of time variation of CO produced, f_{CO} . The accumulated value was calculated by trapezoidal integration. The total carbon gasified was given from the above integration through the experiment. The conversion at a time, X , was given by dividing the integrated value of CO produced until the time, t , by the total carbon gasified. The half value of CO produced at the time, t , was divided by the value of total carbon gasified, and this gave the conversion rate at the time, dX/dt .

In the present study, the gasification experiments were iterated for several times under the same experimental condition using a sample. In order to give the averaged dX/dt vs. X curve, first of all, time variation curve of f_{CO} was given by the spline method. The numerical data of time variation of f_{CO} were taken for every one second. When the negative value was given by spline method, which was often found before 20 s or 30 s of the first sampling method, the value was corrected to be zero. Then all of the data of f_{CO} of one second intervals under the same condition were averaged and then dX/dt and X were calculated as above in a continuous fashion.

From the data of dX/dt vs. X , the regression line was given only for the data by least square method. In order to get the regression line the data of initial 60 s were excluded because the time delay by initial gas mixing was estimated to be around 60 s from the gas flow rate and the volume of the reactor. The data for $X > 0.98$ were also excluded because the so many data points gathered around $(X, dX/dt) = (1, 0)$. The absolute value of the slope was defined as the

reaction rate constant of the gasification.

2.4 Char properties

The chars of the biomass species were separately prepared and taken out from the fluidized bed after cooling. Their weight decrease was measured by combustion at 815°C. The equivalent carbon yield (dry biomass basis) was measured by dividing the decrease in weight by carbon molecular weight.

The cross sectional SEM images were also taken with JEOL JSM-5200. The SEM images of bark char samples were not taken because they were too weak to prepare cross sectional cutting surface.

3. Results and discussion

3.1 SEM images of cross section of char produced under the rapid pyrolysis

Fig. 2 shows the SEM images of cross sections of softwood chars produced from the biomass samples under the rapid pyrolysis. Figs. 3 and 4 show SEM images of cross sections of respectively evergreen and deciduous hardwood chars produced from under the rapid pyrolysis. All of the images are taken with the same magnitude (200 times magnification). In general, pore diameter of various softwood chars in Fig. 2 looks larger than that of that of various kinds of hardwoods. The difference in pores of chars between ever green and deciduous hardwoods is not so clear. Rather the pore diameter looks different one by one irrespective of ever green and deciduous. In general larger pore diameter with same wall stricture makes char density less, which may make reaction rate larger. In this paper this point of discussion will be done later in section 3.5.

3.2 Carbon yield

The carbon yield was calculated from the CO production rate during the gasification assuming $C + CO_2 \rightarrow 2CO$. Linear change was assumed for the period between the two sampling times. The averaged value of the carbon yield is given in Fig. 5 using a black bar for each biomass species. The data for bark samples are placed just next right of the data of the corresponding data for woody portion.

The carbon yield calculated from the weight decrease by combustion of char is also shown in Fig. 5 using a white bar for each biomass species. Though the carbon yield by produced char combustion, which includes weight loss of other component than carbon, gave a little larger values than those calculated from gasification data, almost same values are obtained, which suggests the reliability of the gasification experiment. It was found that the carbon yields

Fig. 2 SEM photos of cross section of softwood char produced under the rapid pyrolysis

Fig. 3 SEM photos of cross section of evergreen hardwood char produced under the rapid pyrolysis

Fig. 4 SEM photos of cross section of deciduous hardwood char produced under the rapid pyrolysis

Fig. 5 Carbon yields from CO production by gasification and from weight reduction by produced char combustion

of the woody portion were almost the same for all of the biomass species while the carbon yield of bark is generally larger than that of corresponding woody portion, which suggests smaller volatile matter contents.

3.3 Variation of gasification rate with carbon conversion

Fig. 6 (a-q) shows relation between carbon conversion and gasification rate for all of the 17 chars produced under the rapid heating rate condition from 17 biomass species including four samples of bark [Fig. 6 (n-q)]. All of the data points correspond to the gas sampling time. The curve in each figure shows the averaged result of the relation between conversion rate and conversion by applying the spline method. All of the curves by averaging data using

spline method look to shift to left compared with the point data. This is caused by the the treatment way of averaging data as follows. In the process of the averaging, the negative value given by spline method was often observed before 20 s or 30 s of the first sampling and the value was corrected to be zero. On the other hand, in case of the data point, the conversion before the first measured point is calculated from the linear approximation line connecting with the origin of the figure of time vs. f_{CO} relation. The reason why the averaged curves give larger maximum values of dX/dt than those of the data points is explained by the same reason. Namely, the total released amount of CO of the average by spline method is a little smaller than those from the average of the experiments given by the points data. Regardless the above discrepancy, the averaged curve is expected to give better description of the dX/dt curve, not only because it is the averaged value but also because it gives better value of CO release by smoothing of initial disturbance of the data. Hence the following discussion is conducted using the averaged curves.

In case of softwood char gasification [Fig. 6 (a-d)], all of the species gave almost the same maximum dX/dt value around 0.003/s. Furthermore after the peak around $X=0.1$, drastic decrease in the rate was observed and then for $X>0.2$, the X vs. dX/dt relation was almost expressed by the following liner relation of $dX/dt = k(1-X)$ which is usually termed as the volume reaction model.

On the other hand, in case of hardwood char, irrespective of evergreen [Fig. 6 (e-h)] or deciduous [Fig. 6 (i-m)], the maximum gasification rate depended on species: dX/dt ranged from 0.0035 to 0.007 /s. Furthermore, the relatively rapid gasification rate was kept up to higher

(a) Japanese red pine

(b) *Chamaecyparis obtuse*

(c) *Cryptomeria japonica*

(d) *Cedrus deodara*

(e) *Eucalyptus camaldulensis* (f) *Quercus phillyraeoides* (g) *Quercus myrsinaefolia* (h) *Rhizophora mucronata* (i) *Celtis sinensis var japonica* (j) *Quercus serrate* (k) *Aphananthe aspera* (l) *Zelkova* (m) *Quercus acutissima*

Fig. 6 Char gasification reactivity (CO_2 25%, 1000°C , rapid heating: SW, Softwood, a, b, c, d, n: EGHW, Evergreen Hardwood; e, f, g, h, o, p: DHW, Deciduous Hardwood; i, j, k, l, m, q)

conversion than the case of softwood. The drastic drop in the rate was usually found only at higher conversion than the case of softwood [Fig. 6 (a-d)], namely around $X = 0.8$ or so. Therefore even when the maximum dX/dt value was almost same, the gasification rate of hardwoods [Fig. 6 (e-m)] is still larger than that of softwoods.

In case of bark samples [Fig. 6 (n-q)], the gasification rate was also same (for *Celtis sinensis var japonica* bark, ENOKI(p)) or larger (*Quercus serrate* bark, KONARA(q)), the maximum gasification rate was shifted to higher conversion, and the dX/dt curve became more flat, as or than that for woody portion.

3.4 Gasification rate constant based on the volume reaction model

Irrespective of the non-linearity of dX vs. X curve, especially for hardwood and bark samples, the regression straight line for dX/dt vs. X averaged curve excluding initial 60 s and $X > 0.98$ was determined and the obtained regression line equation is given in each figure. As shown in all of figures of Fig. 6, all of the straight lines are from the origin of (1, 0), and approximately given as the equation of $dX/dt = k(1-X)$.

Hence the reaction rate constants based on the volume reaction model were given as -(slope of the regression line), and shown in Fig. 7 as bar graph. It was clearly shown that all of the softwood chars gave the rate constant around 0.003 1/s , and most the hardwood chars gave larger values of gasification rate constants than most of the values for softwood chars. Among softwood chars,

Fig. 7 reaction rate constant k and several biomass

only *Quercus myrsinaefolia* (SHIRAKASHI)(g) gave almost same tendency and same maximum value as those of softwoods samples.

3.5 Relation between physical properties and reaction rate constant k

It is well known that various physical properties are different among species and they are published in a webpage¹⁶⁾ collecting data from various literatures such as ones by Osaka Mokuzai Seihin Keieisha Kyougikai¹⁷⁾ and Aida¹⁸⁾. In the present paper, the mutual relations between reaction rate constant and various properties of density and various strengths were examined. Fig. 8 shows comparison between density and reaction rate constant k . The higher density is, the higher reaction rate constant k is, in general, because softwoods have low densities and

Fig. 8 Relation between density and reaction rate constant k Fig. 9 Relation between longitudinal compressive stress¹⁶⁾ and rate constant k

low reaction rates and hardwoods, vice versa. It is also interesting by referring the SEM images of chars shown in Fig. 2 that pore diameter of chars of the softwood looks large. Namely all of the softwoods used in the present paper have relatively low density, and the produced chars show large pore diameter and low gasification reactivity. On the other hand, some hardwoods used in the present paper have relatively high density, and the produced chars show small pore diameter and high gasification reactivity, while some do not show the above tendency.

As we wrote in section 3.1, smaller pore diameter makes mass transfer rate slow in the char, which may affect the reaction rate. However the present results indicate that larger pore structure gives slower gasification rate, which indicates the gasification rate is not controlled by the intra-particle diffusion.

As various strengths, longitudinal compressive stress, Yang's modulus, share strength, vertical tension strength and bend strength have been reported^{16)~18)} for the species

tested in the present study. In general, only among the hardwoods species, the species with higher strength show lower reaction rate constant while softwoods show lower reaction rate constants than hardwoods irrespective of their almost same range of strength values as those of softwoods. Among these figures, only Fig. 9 is shown here, giving relation between longitudinal compressive stress and reaction rate constant k , which shows the strongest relationship with k among various wood strength values we examined. The present tendency suggests the strong structure of the wood reduces the gasification rate, though this relation is only applied to hardwood.

4. Conclusions

Cross sectional pore diameter of char from softwood was generally larger than that from hardwood. The char prepared from softwoods showed lower gasification reactivity with smaller rate constants than that from hardwoods. The gasification rate of char from softwoods generally showed relatively rapid rate at initial stage of gasification and then the rate was drastically decreased around conversion of 0.1, while that of char from hardwoods was kept higher until higher conversion, suggesting the small amount of existence of low reactive parts. Bark reaction rate was almost similar to wood. Reaction rate constants of char from hardwoods were well correlated with longitudinal compressive stress.

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