Journal of Engineering and Technology
Vol.4 No.1 January - June 2016

Energy Properties of Torrefied Biofuel from Cellulose

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***Abstract*— Due to the rise of concern for recycling, waste paper to produce recycled paper and paperboard is recovered in individual countries of the world. The global recovery rate is around 58% in 2014. But some paper such as beverage paper containers and the low quality waste paper are not suitable to the material for recycling. The utilization of the above paper as a solid biofuel to replace coal is considered to be reasonable from the standpoint of cascade cyclic utilization of waste biomass. The waste paper is mainly composed of cellulose and has a good property of low water content, but its heating value is two-thirds of coal. In the present study, the cellulose is reformed by the torrefaction and it is assumed that the torrefied biofuel from cellulose is utilized for the biomass and coal co-firing at existing coal power plants. There are two objectives in the present study. The first one is to correlate the energy properties of torrefied cellulose with the mass yield. The second one is to clarify the optimum mass yield condition to produce torrefied biofuel from cellulose using the modified energy analysis model. From the results, it is found that torrefaction has a positive effect on the reduction of carbon dioxide emission in the case that the energy consumptions for production and transportation of torrefied biofuel are relatively high.**

***Keywords*— Solid biofuel, Torrefaction, Cellulose, Waste paper, Energy properties**

I. INTRODUCTION

Due to the rise of concern for recycling, waste paper to produce recycled paper and paperboard is recovered in individual countries of the world. According to RISI annual review 2015 [1], the global recovery rate is around 58% in 2014. But some paper such as beverage paper containers and the low quality waste paper are not suitable to the material for recycling. The utilization of the above paper as a solid biofuel to replace coal is considered to be reasonable from the standpoint of cascade cyclic utilization of waste biomass. The waste paper is mainly composed of cellulose and has a good property of low water content, but its heating value is two-thirds of coal.

As one of the solid fuels derived from waste paper, the refuse paper and plastic fuel, which is usually abbreviated RPF, is well known and utilized in industrial boilers. The RPF is a mixture of paper and plastic, and has the following advantages. The heating value of waste paper can be raised by blending the waste plastic, and moreover the heating value of RPF can be adjustable by changing the blending ratio of waste paper and waste plastic [2]. The ash content of RPF is much smaller than that of coal, and the range of ash content is 5-10 mass% [2-5]. However, the more the waste plastic is blended in RPF, the less the reduction of carbon dioxide emission becomes, because the property of carbon neutrality for waste paper is cancelled if the blending ratio of waste paper decreases.

In the present study, the utilization of upgraded solid biofuel derived only from waste paper is investigated. Among several upgrading methods, torrefaction has come to attract attention as an effective upgrading method of biomass, because the biomass reformed by the torrefaction has a feature of high energy yield that is defined as the ratio of energy content of torrefied biomass to that of untreated biomass [6, 7]. The torrefaction process was devised in 1998 by Honjyo and Sano [8, 9]. Since then, many studies have been conducted, and several detail reviews on torrefaction have already been reported [10-12].

The main component of waste paper is cellulose, and therefore the cellulose powder reagent is used as an experimental sample. The cellulose is reformed by the torrefaction and it is assumed that the torrefied biofuel from cellulose is utilized for the biomass and coal co-firing at existing coal power plants. The energy properties of torrefied cellulose such as elemental contents, higher heating value and energy yield are correlated with the mass yield using the same method proposed by Sawai et al. [7]. Based on the energy properties, the effect of torrefaction on the reduction of carbon dioxide emission is examined by modifying the previous energy analysis model [7]. In the modified energy analysis model, an increase in mixture ratio of torrefied biofuel from cellulose is incorporated, since the torrefaction process performs functions of grindability [13]. According to Ooiwa [14], it is reported that the mixture ratio of torrefied biomass can be increased by two or three times, because the torrefaction of biomass reduces the load applied on a coal mill.

There are two objectives in the present study. The first one is to correlate the energy properties of torrefied cellulose with the mass yield. The second one is to clarify the optimum mass yield condition to produce torrefied biofuel from cellulose using the modified energy analysis model.

II. EXPERIMENTAL SAMPLES AND PROCEDURES

Fig.1 shows the variation of the mass yield Y_M of torrefied cellulose with pyrolysis temperature T. As seen in the figure, the pyrolysis temperature condition is divided into three regions. In the temperature below 220 deg. C., a slight decrease of Y_{M} occurs due to the elimination of free water. In the temperature above 350 deg. C., Y_{M} is lower than 0.4, and the mass decreasing rate against T is small. In the temperature range between 220 and 350 deg. C, the drastic mass reduction occurs due to the emission of volatile organics and chemical bound water. The pyrolysis in the above temperature range is called "torrefaction"

Fig.2 shows the torrefied cellulose samples for various mass yield. The color of sample changes from light brown to black as the pyrolysis proceeds.

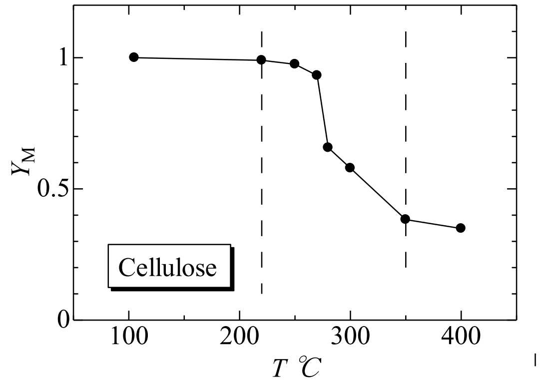


Fig. 1 Variation of Mass Yield with Pyrolysis Temperature

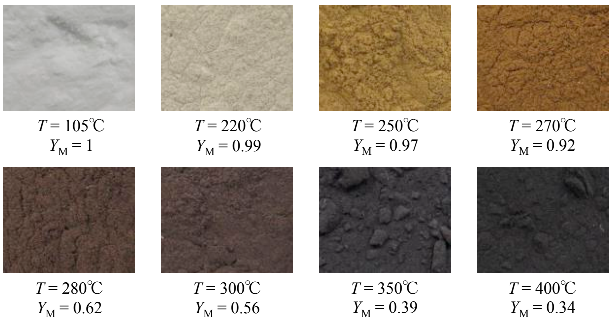

T = 105℃ Y_{M} = 1
T = 220℃ Y_{M} = 0.99
T = 250℃ Y_{M} = 0.97
T = 270℃ Y_{M} = 0.92
T = 280℃ Y_{M} = 0.62
T = 300℃ Y_{M} = 0.56
T = 350℃ Y_{M} = 0.39
T = 400℃ Y_{M} = 0.34

Fig.2 Torrefied Cellulose Samples

III. ENERGY PROPERTIES OF TORREFIED CELLULOSE

Fig.3 shows the variation of carbon, hydrogen and oxygen contents of torrefied cellulose with Y_{M}. The linear relationship between them is confirmed, and the following relations for carbon, hydrogen and oxygen are obtained. As the pyrolysis proceeds, the content of carbon increases, while the contents of oxygen and hydrogen decrease.

$$C\,[\%] = -44.4Y_{\mathrm{M}} + 86.9 \tag{1}$$

$$H\,[\%] = 3.31Y_{\mathrm{M}} + 2.97 \tag{2}$$

$$O\,[\%] = 41.8Y_{\mathrm{M}} + 9.33 \tag{3}$$

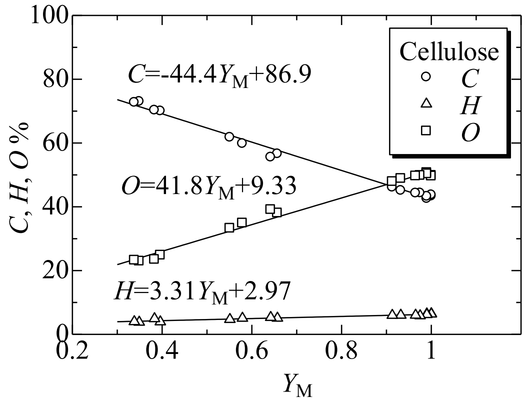


Fig. 3 Variation of C, H, O with Mass Yield

One of typical experimental correlations to estimate the higher heating value (HHV) of biomass is shown in eq.(4) where C, H, S, O, N and A represent carbon, hydrogen, sulfur, oxygen, nitrogen and ash contents and the unit of HHV is MJ/kg, which is proposed by Channiwala et al. [15].

$$\begin{aligned} HHV = 0.3491\ C + 1.1783\ H + 0.1005\ S \\ - 0.1034\ O - 0.0151\ N - 0.0211\ A \end{aligned} \tag{4}$$

Chen et al. [12] has shown that the above relation is also applicable to the estimation of HHV of torrefied biomass. Substituting eqs.(1), (2) and (3) into eq.(4), HHV of torrefied cellulose can be expressed by the following linear equation of Y_{M}.

$$HHV = -15.92\,Y_M + 32.87 \tag{5}$$

From the above equation, HHV_1 and HHV_0 which are defined by HHV at Y_{M}=1 and Y_{M}=0 can be obtained as follows.

$$HHV_1 = 16.95 \text{ MJ/kg} \tag{6}$$

$$HHV_0 = 32.87 \text{ MJ/kg} \tag{7}$$

HHV_1 represents HHV for untreated cellulose in oven-dry, while HHV_0 is a virtual HHV and defined as the limit of HHV when Y_{M} approaches zero. Using eqs. (6) and (7), HHV of torrefied cellulose can be rewritten as follows.

$$HHV = -(32.87 - 16.95)\,Y_M + 32.87 \tag{8}$$

Dividing eq.(8) by HHV_1, the improving ratio of HHV of torrefied cellulose to that of untreated cellulose, R_{H} is obtained and R_{H} is expressed by the following linear equation of Y_{M}.

$$R_H = -\ (1.94 - 1)Y_{\mathrm{M}} + 1.94 \tag{9}$$

The energy yield of torrefied biofuel, Y_E that is defined as the ratio of energy content of torrefied cellulose to that of untreated cellulose is defined by

$$Y_E = (HHV/HHV_1) \times Y_M = R_H \times Y_M \tag{10}$$

Substituting eq.(9) into eq.(10), the following quadratic equation of Y_M is obtained for Y_E of torrefied cellulose.

$$Y_E = \{-(1.94-1)Y_M + 1.94\} \times Y_M \tag{11}$$

From the above results, it is found that the energy properties of torrefied cellulose such as elemental contents, *HHV*, R_H and Y_E can be correlated with Y_M. Fig.4 shows the variation of R_H and Y_E with Y_M. The solid and broken curves represent the results of torrefied cellulose and Japanese cedar. As compare with Japanese cedar obtained in the previous study [7], R_H and Y_E of torrefied cellulose at a given Y_M are larger than those of torrefied Japanese cedar. In the case that Y_M=0.7, R_H and Y_E of torrefied cellulose are 1.28 and 0.90, while R_H and Y_E of torrefied Japanese cedar are 1.19 and 0.83.

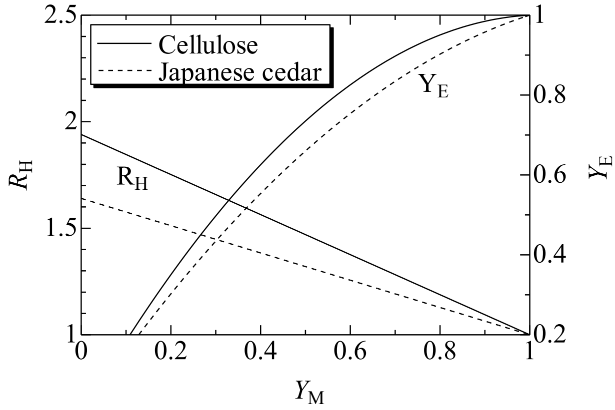


Fig. 4 Variation of R_H and Y_E with Y_M of torrefied cellulose

IV. Energy Analysis of Torrefied Biofuel from Cellulose

In the same manner as the previous study [7], the net energy of torrefied cellulose to replace fossil fuels, ΔE_N is given by

$$\Delta E_N = Y_E \times HHV_1 \times M_B - Y_M \times M_B \times E_C \tag{12}$$

where M_B is the gross mass of cellulose used to produce torrefied biofuel and E_C is the energy consumption per unit mass required for transportation, comminution, torrefaction and molding of biofuel. The first and second term in the right hand side represent the gross energy content of torrefied cellulose and the gross energy consumption for production and transportation, and the difference between them becomes the net energy. It is noted that M_B is changed by the mixture ratio of torrefied biofuel for co-firing, and given by the following relation

$$M_B = M_{B0} \times r_{CF}, \tag{13}$$

where M_{B0} is the gross mass of cellulose for the basic mixture ratio of torrefied biofuel, 3.0 mass% and r_{CF} is the increasing ratio of mixture ratio to the basic mixture ratio 3.0 mass%. Introducing r_{CF} in eq.(13) makes it possible to examine the effect of increase in mixture ratio of torrefied biofuel on ΔE_N. Dividing ΔE_N by HHV_1 and M_{B0}, the dimensionless net energy of torrefied biofuel, ΔE is obtained by

$$\Delta E = (Y_E - Y_M \times C) \times r_{CF}, \tag{14}$$

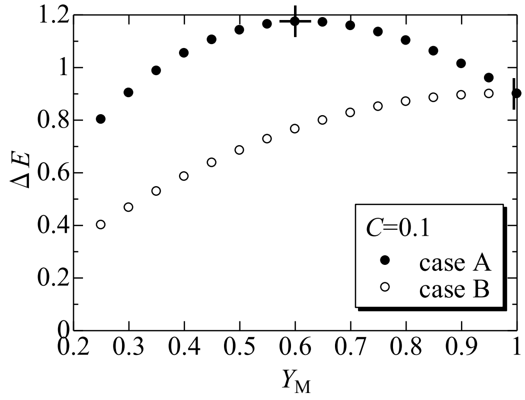
where C represents the non-dimensional energy consumption and defined by the following relation.

$$C = \frac{E_C}{HHV_1} \tag{15}$$

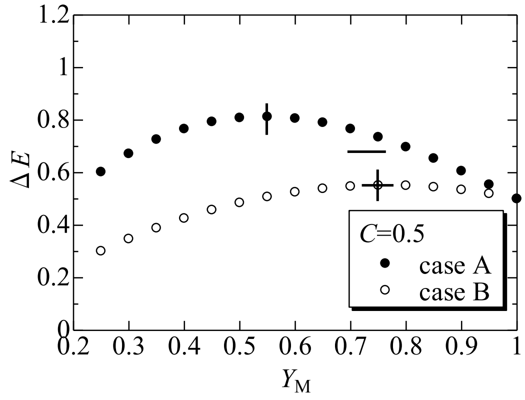
Table 1 Mixture ratio of torrefied biofuel from cellulose for co-firing

(a) caseA			(a) case B		
Y_M	Mixture ratio of torrefied biofuel [%]	r_{CF}	Y_M	Mixture ratio of torrefied biofuel [%]	r_{CF}
0.25	6.0	2.00	0.25	3.0	1.00
0.30	5.8	1.93	0.30	3.0	1.00
0.35	5.6	1.87	0.35	3.0	1.00
0.40	5.4	1.80	0.40	3.0	1.00
0.45	5.2	1.73	0.45	3.0	1.00
0.50	5.0	1.67	0.50	3.0	1.00
0.55	4.8	1.60	0.55	3.0	1.00
0.60	4.6	1.53	0.60	3.0	1.00
0.65	4.4	1.47	0.65	3.0	1.00
0.70	4.2	1.40	0.70	3.0	1.00
0.75	4.0	1.33	0.75	3.0	1.00
0.80	3.8	1.27	0.80	3.0	1.00
0.85	3.6	1.20	0.85	3.0	1.00
0.90	3.4	1.13	0.90	3.0	1.00
0.95	3.2	1.07	0.95	3.0	1.00
1	3.0	1.00	1	3.0	1.00

Table 1 shows the analysis condition to examine ΔE for various Y_M. In the case A, it is assumed that the mixture ratio of torrefied biofuel from cellulose increases linearly with decreasing Y_M, since the torrefaction process performs functions of grindability. The maximum mixture ratio is 6.0 mass% at Y_M=0.25. In the case B, the mixture ratio of torrefied biofuel from mixture ratio of torrefied cellulose is kept to be 3.0 mass% regardless of Y_M.



(a) C=0.1



(b) C=0.5

Fig. 5 Variation of ΔE with Y_M

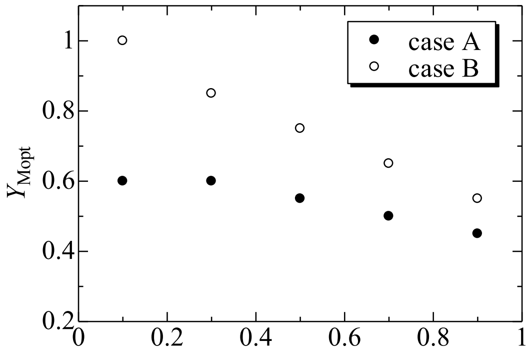


Fig. 6 Variation of optimum Y_M with C

Fig.5 shows the variation of ΔE with Y_M for the case A and case B. Fig.5a and 5b are the results for C=0.1 and 0.5. The cross symbol represents the maximum of ΔE for each analysis condition. In the case of C=0.1, shown in Fig.5a, which corresponds to the condition of low energy consumption, ΔE for the case B decreases monotonously with decreasing Y_M. This means that the carbon dioxide emission of torrefied biofuel becomes larger than that of untreated biofuel at Y_M=1, that is the torrefaction has a negative effect on the reduction of carbon dioxide emission. But ΔE for the case A has a maximum at Y_M =0.6, which means that the carbon dioxide emission of torrefied biofuel at Y_M =0.6 is smallest. In Fig.5b, it is shown that both ΔE for the case A and B have maxima at Y_M =0.55 and 0.75. When the non-dimensional energy consumption C is 0.5, the effectiveness of torrefaction in reducing the carbon dioxide emission are confirmed for the case A and B.

Fig.6 shows the variation of optimum Y_M with C for both cases. The optimum Y_M is defined as the mass yield when ΔE has a maximum. When the optimum Y_M is below unity, the torrefaction has a positive effect on the reduction of carbon dioxide emission. In the case B, the optimum Y_M decreases monotonously, and becomes below unity when C is above 0.1. Therefore, the torrefied biofuel becomes effective in reducing the carbon dioxide emission, when the non-dimensional energy consumption C is relatively high. In the case A, the optimum Y_M is in the range between 0.6 and 0.45 for all conditions of C. From the results, it is found that the torrefaction has a positive effect on the reduction of carbon dioxide emission regardless of C, when the mixture ratio of torrefied biofuel increases with decreasing Y_M.

V. CONCLUSIONS

Energy properties of torrefied cellulose correlated with mass yield, and the optimum mass yield condition to produce torrefied biofuel are investigated. The results obtained in the study are as follows.

(1) The improving ratio of *HHV* of torrefied cellulose to that of untreated cellulose is expressed by the linear equation of mass yield. The energy yield of torrefied cellulose is correlated with the quadratic equation of mass yiels. The improving ratio of *HHV* and energy yield of torrefied cellulose at a given mass yield are larger than those of torrefied Japanese cedar.

(2) The modified energy analysis model to examine the net energy of torrefied biofuel from cellulose, ΔE is proposed by taking into account the mixture ratio of biomass for co-firing. In the case of low energy consumption, ΔE decreases monotonously with decreasing mass yield, which means that the torrefaction has a negative effect on the reduction of carbon dioxide emission. But in the case of higher energy consumption, ΔE has a maximum, which shows that the torrefaction has a positive effect on the reduction of carbon dioxide emission.

(3) When the mixture ratio of torrefied biofuel for co-firing increases with decreasing mass yield, the torrefaction has a positive effect on the reduction of carbon dioxide emission regardless of the non-dimensional energy consumption.

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