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<75 75 106 150 250 > 500 Particle size [µm]

## Milling Characteristics of Coal and Torrefied Biomass Blends in a Roller Mill

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the larger particles of the milled product and in the samples collected inside the roller mill than those in the input feedstock. The biomass-derived part with the highest X/G ratio accumulated inside the roller mill, which is probably why the mill power and differential pressure increased with the TP content. The TP, with poor grindability, was discharged from the mill with a larger particle size than that of coal. Although torrefaction treatment pyrolyzed the biomass, the degree of torrefaction can vary within the pellets depending on the torrefaction conditions. To ensure stable operation of the roller mill and an effective reduction in the size of the coal−TP blend, the selection and use of uniformly and sufficiently torrefied wood pellets are important.

### **ENTRODUCTION**

Replacing some of the coal by biomass is an effective way to reduce  $CO<sub>2</sub>$  emissions from pulverized-coal-fired power plants and integrated coal gasification-combined cycle power plants. However, compared with coal, biomass has lower grindability in the existing roller mills<sup>1</sup> and a lower calorific value per weight. $2,3$  $2,3$  $2,3$  The maximum biomass co-firing ratio for stable operation is limited by the design of the coal mill and the grindability of the feed biomass. Researchers have recently investigated the potential of pelletization and torrefaction technologies<sup>4</sup> to improve the fuel properties of biomass and make it suitable for use in the existing power plants.

products also increased. Furthermore, the biomass-derived neutral sugar content and the xylose/glucose  $(X/G)$  ratio were higher in

Torrefaction is extensively studied as a method for upgrading the fuel properties of biomass $5,6$  and involves a thermochemical treatment that pyrolyzes some of the biomass components in an inert atmosphere and reduces the total oxygen content.[7](#page-4-0) Torrefaction of the biomass significantly increases its calorific value per unit weight, water repellency, and grindability.<sup>[1,3,8](#page-4-0)−[10](#page-4-0)</sup> Therefore, commercial-scale torrefaction facilities have been developed, which currently provide torrefied wood pellet (TP) products to the market.

In pulverized-coal-fired power plants, the solid fuels (coal and biomass) are milled to reduce the particle size, which aids subsequent combustion and gasification. Current power plants typically mill biomass and coal blends in a conventional vertical

roller mill or mill each component separately in dedicated mills. $11,12$  It is possible to achieve a high biomass co-firing ratio when the biomass and coal are milled separately because the mill conditions can be optimized for the biomass with poor grindability. However, this requires additional infrastructure. In contrast, when a conventional vertical roller mill is used, although the maximum biomass co-firing ratio is limited by the equipment specifications, operating conditions, and the grindability of the feedstock (biomass/coal), the capital costs of installing new equipment are reduced.<sup>12,[13](#page-4-0)</sup> Therefore, most power plants that co-mill coal with biomass use existing vertical roller mills developed for pulverizing coal. $^{14}$  $^{14}$  $^{14}$ 

The particle size of the coal/biomass-milled products significantly affects the combustion efficiency, combustion stability, and unburned ash content.<sup>15,[16](#page-4-0)</sup> To control the particle size of the milled products, the roller mills are usually equipped with a rotary classifier.<sup>[1](#page-4-0),[17](#page-5-0)</sup> Typically, coal feedstocks are milled

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**Roller mill** 



<span id="page-1-0"></span>until over 80 wt % of the milled coal passes through a 200 mesh  $(\langle 75 \mu m \rangle)$ . Because torrefied biomass has higher reactivity and volatile content than coal, a larger milled particle size is acceptable.<sup>[18,19](#page-5-0)</sup> To appropriately increase the biomass co-firing ratio, it is necessary to grind the coal and biomass into particle sizes corresponding to their combustibility.

We previously reported the milling characteristics of blends of coal and wood pellets (WPs) without torrefaction treatment using the neutral sugar present only in plant biomass, but not in coal, as a tracer in a roller mill. $^{20}$  $^{20}$  $^{20}$  In that study, the WPs were selectively accumulated inside the roller mill and discharged with a particle size larger than that of the coal particles. In another study, we observed that the grindability of the coal− biomass blend was highly dependent on the degree of torrefaction of the biomass.<sup>[21](#page-5-0)</sup> Considering this background, in the present study, blends of coal and commercially available TP were milled using a bench-scale roller mill. The neutral sugar content (NSC) was measured to evaluate the milling performance of various coal−TP blends. We expect that the findings of this study will provide an important foundation for the selection and milling of torrefied biomass toward applications enabling the sustainable use of biomass.

#### ■ MATERIALS AND METHODS

Feedstock Materials. Commercially available TPs (Biomass Fuel Co., Ltd., Tokyo, Japan) made from Acacia cultivated in Vietnam were used as a feedstock. Bituminous coal mined from Hunter Valley, Australia, was used as the coal sample. The analytical methods of the Japanese Industrial Standard (JIS) were adopted for higher heating value characterization, proximate analysis (ash, volatile matter, and fixed carbon), and elemental analysis (carbon, hydrogen, nitrogen, and oxygen) of the TPs and coal, which were also used in our previous study.<sup>20,21</sup>

Neutral Sugar Analysis. To evaluate milling performance, the NSC of the feedstocks, milled products, and samples from inside the roller mill were determined using the two-stage sulfuric acid hydrolysis method<sup>22</sup> with slight modifications.<sup>2</sup> Briefly, each sample was hydrolyzed and the neutral sugar in solution was quantified using high-performance liquid chromatography (HPLC; Prominence, Shimadzu Corporation, Japan) with SP0810 columns (Showa Denko K. K., Japan) and a charged aerosol detector (Corona Veo RS; Thermo Fisher Scientific). The mobile phase was acetonitrile/water (13.0/ 87.0  $(v/v)$ ) with a flow rate of 0.5 mL/min. The experiments were performed three times for each sample.

Milling of Coal-TP Blends. Milling tests with various coal−TP blends [13.2 wt % (case 1), 25.5 wt % (case 2), and 36.8 wt % (case 3) TP] were performed using a bench-scale vertical roller mill (UM3.6, Ube Machinery Corporation, Ltd., Japan). The coal samples were coarsely milled (<10 mm) using a hammer mill (H-15, Hosokawa Micron Corporation, Japan) before the milling tests.

A schematic of the roller mill is shown in Figure 1. The major parts of the roller mill are grinding rollers, a rotating table, and a rotating classifier. The feedstock falls from the top of the roller mill onto the center of the table and is moved to the rollers by centrifugal force caused by the rotation of the table. Under a flow of hot air from the bottom of the mill, the milled products are blown up into a rotating classifier. The milled products below a certain particle size are classified and conveyed by air to the mill outlet. The coarse particles fall back onto the table for further grinding. The input enthalpy of the





feedstocks is kept constant in coal-fired power plants. To ensure a constant heating value  $(2.39 \text{ MJ/s})$ , the feed rate of the coal−TP blend was controlled with a conveyor belt speed.

The operating conditions during the roller mill experiments were as follows: rotational speed of the rotary classifier of 120 rpm; rotational speed of the table of 60 rpm; a roller pressure of 5 MPa; a carrier-gas flow rate of 600 N  $m^3/h$ ; gas temperature at the mill outlet of 70 °C; a milling time of 120 min.

The milling conditions for coal alone and the coal−TP blends are shown in Table 1. The milling tests were run for

Table 1. Milling Conditions for Coal Alone and the Coal− TP Blends<sup>a</sup>

parameter	case $0^{20}$	case 1	case 2	case 3	
feedstock	coal	$\text{coal/TP}$	$\text{coal/TP}$	$\text{coal/TP}$	
ratio $(coal/TP)$ [J/J]	100/0	90/10	80/20	70/30	
ratio (coal/TP) [wt %]	100/0	86.8/13.2	74.5/25.5	63.2/36.8	
feed rate [kg d.b./h]	300	333	345	362	
<sup>a</sup> TP, torrefied wood pellet.					

120 min to allow the conditions within the roller mill to stabilize. Stabilization of each milling test was confirmed by monitoring the differential pressure of the mill, the power consumption of the rotating table and classifier, and the production rate of the milled products every 2 s.

Herein, the differential pressure of the mill is defined as the difference between the gas inlet and outlet pressures. The total power consumption is the sum of the power consumption of the rotating table and classifier. The differential pressure and total power consumption of the mill are presented as the average values around 110 min after the test started. At the end of the test, when the mill differential pressure and the total power consumption of the mill were stable, the sample inside the mill was aspirated from the sampling port near the center of the rotating table and collected. The milling performance was also assessed in terms of the particle-size distribution of the products after milling. The finished milled products and samples taken from inside the roller mill were sieved using a shaker for 30 min to classify them as follows: <75 (collected in a pan), 75−106, 106−150, 150−250, 250−500, and >500 μm. After classification, the milled and sieved samples were weighed.

# ACS Omega<br>■ RESULTS

Feedstock Properties. The results of the heating value characterization, proximate analysis, and elemental analysis of the coal and TPs are provided in Table 2. Compared to coal,

Table 2. Heating Values and the Results of Proximate and Ultimate Analysis of the Coal and TP Feedstocks $a$ 



 ${}^a$ TP, torrefied wood pellet.  ${}^b$ Dry basis. <sup>c</sup>As received.  ${}^d$ Dry ash-free basis.

TP has a lower high/low heating value, moisture content, ash content, and fixed carbon content, and higher volatile matter content. TP has a higher oxygen content than coal. Therefore, TP is considered an easily combustible fuel but with a low calorific value. In addition, the TP samples had a lower nitrogen content than coal, which are sources of nitrogen oxides. Compared to the reported properties of raw and torrefied woody biomass,  $24,25$  the TP used here was mildly torrefied.

The NSC of the TP and coal samples was measured. The data for the coal sample were below the limit of detection of the instrument. The total NSC of the TP sample was 58.6  $\pm$ 3.1 wt %, indicating that the TP contained mostly neutral sugars, mainly glucose (52.9  $\pm$  2.1 wt %) and some xylose (4.0  $\pm$  0.5 wt %). The TP also contained mannose (0.7  $\pm$  0.2 wt %), arabinose (0.6  $\pm$  0.3 wt %), and galactose (0.3  $\pm$  0.0 wt %), which were considered negligible as they were all below 1 wt %. Compared to previously reported properties of plant biomass,  $23,26$  $23,26$  although most of the hemicellulose was pyrolyzed in our TP sample, cellulose (a polymer of glucose) was present. The NSC results further prove that the TP had a very mild degree of carbonization (torrefaction).

Milling Performance. The generation rate of the milled products reached a constant value after ∼30 min. For cases 0, 1, 2, and 3 (defined in [Table 1](#page-1-0)), the average generation rate of the milled products (around 110 min) was  $287 \pm 9$ ,<sup>[20](#page-5-0)</sup> 343  $\pm$  9,  $350 \pm 10$ , and  $356 \pm 17$  kg w.b./h, respectively. Table 3 presents the power consumption and differential pressure of

#### Table 3. Power Consumption and Differential Pressure of the Mill for the Four Milling Cases



the roller mill. The ratios were calculated by dividing the power consumption or differential pressure of the blend tests with those of the control (coal only). Compared to milling coal alone, the power consumption increased as the TP content increased.

Figure 2 presents the particle-size distributions of the coal− TP blends. The highest weight yield was for the  $\langle 75 \mu m$ 



Figure 2. Particle-size distribution of the products from the four milling tests.

fraction in all cases, and the weight yields decreased with increasing particle size. As the TP blending ratio increased, the yield of smaller particles decreased, and the yield of larger particles increased.

Compared to the weight yield of the  $\langle 75 \mu m \rangle$  fraction for case 0 (89.4 wt %), the yields of case 1 (51.5 wt %), case 2  $(40.6 \text{ wt } %)$ , and case 3  $(38.5 \text{ wt } %)$  corresponded to a reduction by 37.9, 48.8, and 50.9 wt %, respectively. The decrease in the weight yield of the  $\langle 75 \mu m$  fraction was higher than the corresponding addition of TP. Therefore, the coal particle size in the milled product significantly increased in the presence of TP. The weight yield of the  $>500 \mu m$  fraction was undetectable for case 0, but was 2.9, 4.4, and 4.7 wt % for cases 1, 2, and 3, respectively. Therefore, when TP with low grindability is blended with coal, the average particle size of the final milled product is higher. Furthermore, the particle size of the coal in the milled product was significantly larger than that achieved when the coal was milled alone.

NSC. NSC of various coal−TP blends was measured to evaluate their milling performance and neutral sugar composition. [Figure 3a](#page-3-0) presents the NSC values of the coal− TP blends corresponding to each particle-size range after milling. Regardless of the TP content, the NSC tended to be higher for the larger particle-size fraction. Comparing each case, the NSC of the  $\langle 75 \mu m$  fraction was higher in the case of higher TP contents. In contrast, the NSC of the  $>500 \mu m$ fraction was lower for higher TP contents. Therefore, increasing the TP content facilitated the preferential milling of TP in the mixture.

Xylose/glucose  $(X/G)$  ratio of the NSC for each particlesize range of the milled product is shown in [Figure 3b](#page-3-0). The X/ G ratio tended to be higher for the larger particle-size ranges. It was considered that the fraction of TP with lower X/G ratios was milled preferentially. This trend was particularly obvious for mixtures with lower TP contents. Compared with the X/G ratio of the input feedstock (0.076), the ratios of the >500  $\mu$ m fraction were 2.72, 1.74, and 1.52 times higher for cases 1, 2,

<span id="page-3-0"></span>

Figure 3. Neutral sugar analysis for each particle-size range of the coal−TP blends. (a) NSC; (b) xylose/glucose ratio.

and 3, respectively. The  $X/G$  ratio of the largest fraction was particularly high for case 1 with the lowest TP content.

Table 4 shows that the NSC values of the samples collected from inside the roller mill were higher than those of the input

Table 4. NSC of the Input Feedstocks and Samples from Inside the Mill $a$ 

milling test	NSC input feedstock [wt %]	NSC sample from mill table $\begin{bmatrix} \text{wt } \% \end{bmatrix}$		
case 1	7.7	13.4		
case 2	14.9	18.5		
case 3	21.6	23.1		
<sup>a</sup> NSC, neutral sugar content.				

feedstocks for all blends. Like the feedstock, the NSC values of the samples collected from inside the roller mill increased with increasing TP content ratio. For cases 1, 2, and 3, the samples from inside the mill showed NSC values that were higher by a factor of 1.74, 1.24, and 1.07 than those of the corresponding feedstock. In particular, milling with a low TP content resulted in a significant accumulation of neutral sugar in the mill.

Table 5 shows that the X/G ratios of the samples collected from inside the mill were higher than those of the input feedstock in all cases, especially for the blends with low TP

Table 5. X/G Ratios of the Input Feedstocks and the Samples from Inside the Mill<sup>a</sup>

milling test	$X/G$ feedstock $[-]$	$X/G$ samples from mill $[-]$
case 1	$0.076 \pm 0.014$	$0.133 \pm 0.010$
case 2	$0.076 \pm 0.014$	$0.091 \pm 0.011$
case 3	$0.076 + 0.014$	$0.078 \pm 0.010$

 ${}^a$ X/G, xylose/glucose.

blending ratio. The X/G ratio of the samples from inside the mill was higher by a factor of 1.77, 1.21, and 1.04 than those of the corresponding feedstocks for cases 1, 2, and 3, respectively.

#### ■ DISCUSSION

The power consumption and differential pressure of the mill measured during milling the coal−TP blends were higher than when using coal alone. The particle size of the milled product tended to increase with increasing TP content in the blend. The degree of torrefaction and the TP content have been shown to greatly influence the milling performance,  $21,27,28$  $21,27,28$  $21,27,28$ where well-torrefied biomass is milled in preference to coal.<sup>[20](#page-5-0)</sup>

Because the TP used in this study had a low degree of torrefaction and lower grindability in the roller mill than the coal, increasing the TP content resulted in an increase in the mill power consumption and differential pressure and a larger particle size of the final milled product. These tendencies are consistent with those obtained for milling woody biomass and coal blends.<sup>[20,29](#page-5-0)</sup> Because the feed rate of the coal−TP blend was set as equal-calorie based for case 0, the weight-based feed rate of the blends was higher than that of coal alone. Therefore, the different feed rates may have contributed to the increase in power consumption and differential pressure. The addition of TP considerably increased these factors, even for case 1 with the lowest TP content. These differences could also be attributed to the different milling properties of the TP blends.

Blending coal with TP increased the final particle size of the product compared to coal alone. Although it is considered that TP does not need to be milled as much as coal owing to its higher volatile content and reactivity,  $24,25$  $24,25$  $24,25$  the evaluation of the milling performance of TP with low grindability is important for the selection of TP feedstocks that enable stable operation of the mill and reduction of the particle size of the milled product. Both the milled products and those collected from inside the roller mill had higher NSC values and X/G ratios than the input feedstock.

In our previous study of raw  $WPs<sub>1</sub><sup>20</sup>$  $WPs<sub>1</sub><sup>20</sup>$  $WPs<sub>1</sub><sup>20</sup>$  we observed no difference in the X/G ratios between the input feedstock and samples collected from inside the roller mill and no variation in X/G between the different particle sizes of the milled products. Because it has been reported that xylan, which is a polysaccharide of xylose, pyrolyzes during torrefaction at lower temperatures than cellulose, which is a polysaccharide of glucose, $30^2-32$  $30^2-32$  $30^2-32$  parts of the biomass with different compositions could have different grindability.<sup>[33](#page-5-0)</sup> Because the TP used here had a low degree of torrefaction, there were probably parts of the sample that were not sufficiently pyrolyzed and had a high X/G ratio. In the coal−TP blends, the parts of the TP with low grindability were not discharged well as milled products and accumulated in the roller mill.

It is considered that coal was preferentially milled and discharged from the mill, resulting in a relatively high ratio of TP accumulated in the mill. This accumulated TP probably contributed to the increase in mill power consumption and differential pressure and was discharged from the mill without being sufficiently milled. Because the general methods for evaluating the fuel properties (i.e., industrial analysis, elemental analysis, and heating value characterization) are measured using the fine fraction of the product, the measured values are an average of the solid fuel. Although the TP composition depends on that of the raw biomass, color differences between the TPs derived from thermal nonuniformity treatment and packed-bed torrefaction treatment have been reported.<sup>[34](#page-5-0)</sup> In <span id="page-4-0"></span>addition, there is also a report on the compositional difference between the surface and bulk of the TP owing to the thermal gradient across the particles during torrefaction.<sup>[35](#page-5-0)</sup> Therefore, even a small amount of TP with a low degree of torrefaction in the blend could result in an increase in the particle size of the milled products, as well as increase the mill power consumption and differential pressure. To ensure the stable operation of the roller mill and an effective reduction in the size of the coal−TP blend, the selection and use of uniformly and sufficiently torrefied wood pellets important.

Blending biomass with coal is an attractive way to reduce CO<sub>2</sub> emissions from existing pulverized-coal-fired power plants without the need to install major new facilities. Therefore, there have been many attempts to use torrefied biomass as a solid fuel. Although large-scale experiments require large amounts of torrefied biomass and large experimental mills, the accumulation of experimental results using large-scale systems is important to further increase the biomass co-firing ratio. It should be noted that the milling performance (i.e., the particle-size distribution, the accumulation of TP in the roller mill, and the mill differential pressure and power consumption) in actual industrial roller mills depends on the equipment specifications, operating conditions, and the properties of the feedstock (biomass/coal).

In this study, it was shown that increasing the content of TP with a low degree of torrefaction increased the particle size of the milled products, the mill power consumption, and differential pressure, while the portion of the TP with a high X/G ratio that was difficult to mill accumulated inside the mill and was discharged with large particle size. To ensure stable operation of the roller mills and to sufficiently reduce the size of the milled products, it is considered optimal to use fuels that have been uniformly and sufficiently torrefied. Torrefied fuels are produced from various biomass materials with a range of torrefaction conditions, and the sugar composition depends on the biomass feedstock. Hence, further studies are required to provide more detailed information regarding the types of biomass and the effects of the degree of torrefaction on the milling performance of roller mills.

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#### Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

#### Notes

The authors declare no competing financial interest.

The data sets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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#### **B** ABBREVIATIONS

TP, torrefied wood pellet; WP, wood pellet; d.b., dry basis; w.b., wet basis; wt %, weight-basis %; HPLC, high-performance liquid chromatography; JIS, Japanese Industrial Standard; NSC, neutral sugar content

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