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Communication

Water Resistance of Torrefied Wood Pellets Prepared by Different Methods

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Abstract: Torrefaction used in combination with pelletization is a promising technology to upgrade solid biofuels and has been demonstrated worldwide. In comparison with normal biomass pellets, which disintegrate under wet conditions, one of the advantages of torrefied biomass pellets is better water resistance. An understanding of the differences in water proof properties for torrefied biomass pellets by different production schemes can promote their further application. In the communication, various torrefied pellets were exposed to indoor and outdoor conditions, and changes in moisture content and diameter were examined. Two production schemes for the torrefied pellets were used for comparison: the torrefaction of wood chips followed by pelletization (pre-torrefaction) and the pelletization of wood chips followed by torrefaction (post-torrefaction). It was found that the post-torrefied pellets had much lower moisture levels than the pre-torrefied pellets in both indoor and outdoor tests. In the outdoor test with no-roof condition, the rate of increase in moisture content for the pre-torrefied pellets was more than double that for the post-torrefied pellets, and the post-torrefied pellets exhibited almost no diameter change. The results on the superior water resistance of post-torrefied pellets were nearly consistent with those reported in previous literature. Torrefied pellets have been considered for industrial use, such as in co-combustion and gasification on a large scale. Taking advantage of the different water resistances, torrefied pellets could also be used by personal and community consumers on a small scale for long-term indoor and outdoor storages as advanced solid biofuels with high waterproof performance, energy density, and lower biodegradation.

Keywords: wood pellets; torrefaction; water resistance; small-scale use

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1. Introduction

Biomass is an organic renewable energy resource with properties distinct from solar and wind energy. Fossil fuels are also organic resources originating from biomass; however, they are nonrenewable. To minimize the use of fossil fuels and establish a sustainable society, woody biomass could play an important role as an energy resource as it is compatible with construction and furniture materials, pulp and paper production, and various other applications. One disadvantage of using biomass for energy is its lower energy density compared with fossil fuels, resulting in lower thermal output per volume during combustion and inefficient storage and transportation. To increase its energy density, the densification of biomass into cylindrical pellets using minimizing variations in density and moisture content has been commercialized [1]. The benefits of such pellets have been reported not only with regard to upgrading energy density, but also for combustion [2–4]. However, pellets have hygroscopic properties, leading to disintegration, thus increasing their rotting rates and self-heating and decreasing their heating values [1,5].

Thermal treatment has conventionally been used to improve biomass properties, and Esteves and Pereira [6] have introduced numerous studies on using wood materials in this respect. Carbonization treatment aimed at conversion into charcoal is also known to upgrade the calorific values of biomass fuel. Recently, low-temperature heat treatment at 250 °C–300 °C in the absence of oxygen, called torrefaction, combined with pelletization has been demonstrated worldwide as a promising method to upgrade the characteristics of solid fuels, maximize their energy density, and achieve better hydrophobicity [7–12]. One potential application for this is to co-firing in pulverized coal-fired power plants [9–11], where the fuel demand would require up to a few hundred thousand tons annually. Economic simulations show that co-firing using coal is commercially available because of the low transportation and milling costs [10,11], and its integration with other processes makes it more economically viable than as a standalone process [12].

In Japan, the total weight of imported wood pellets in 2019 was ~1.65 million t, which was 17 times that of 5 years ago. This is due to a rapid increase in the demand for power generation. At the same time, domestic pellet production was only ~140,000 t from ~150 domestic pellet mills [13], indicating that the production of local pellet mills in Japan has been operating on a small scale. Recently, the concept of producing torrefied pellets using small-scale production in rural areas has been proposed [14–16], in which case the torrefied fuel would require heat use and stable storage. After their fundamental studies on the effects of torrefaction treatment conditions on mass yields, calorific values, and mechanical durability at a laboratory scale [17–19], Yoshida et al. [16,20,21] constructed a demonstration plant aiming at small-scale use, using a treatment amount of 20 kg/h to continuously produce torrefaction chips from raw wood chips. Burning trials of the torrefied pellets produced in this demonstration plant for a commercial pellet stove revealed that the torrefied pellets exhibited a faster ignition time than normal pellets [20]. Then, a commercial-scale plant was designed to produce torrefied wood pellets at a small scale (~3000 t of torrefied fuel annually) using local wood resources [16]. Torrefied pellets also have the advantage of better water resistance than normal pellets [7,8], which is crucial for both storage and use in hygroscopic conditions. Many papers on the hydrophobicity of torrefied biomass fuel have been published, and its behavior in terms of moisture content and dimensional change after water impregnation has been discussed [22–29]. For example, Felfli et al. [22] observed a change in moisture content after directly impregnating a torrefied briquette into water and found that the change in the moisture content of untreated pellets disturbed their shape within a few minutes. Further, Ghiasi et al. [25] measured water uptake in torrefied pellets prepared by two methods: torrefaction of wood chips followed by pelletization (pre-torrefaction) and pelletization followed by torrefaction (post-torrefatcion). They reported that post-torrefied pellets demonstrated lower water absorption than pre-torrefied ones [25]. Kubojima et al. [27] created an original apparatus to evaluate the hydrophobicity of wood pellets based on swelling behavior. They found both that torrefied wood pellets were less likely to swell and that swelling was smaller in post-torrefied pellets [27]. On the other hand, post-torrefied pellets have the disadvantage of a lower energy density compared with pre-torrefied pellets because of weight loss during torrefaction [18,30]. To promote the further application of torrefied biomass pellets, an understanding of the water resistance properties at practical levels such as high humidity or outdoor conditions is necessary.

In this communication, various torrefied wood pellets were exposed under indoor and outdoor hygroscopic conditions. The water resistances were examined, with a focus on the feasibility of these torrefied pellets for small-scale use.

2. Materials and Methods

2.1. Materials

Table 1 shows the normal (non-torrefied) and torrefied pellets used in this study. Xylems of Japanese cedar (Sugi, *Cryptomeria japonica*) and Japanese chestnut oak (Konara, *Quercus serrata*) were used as raw materials. For comparison, two methods were used in the production process of the torrefied pellets: torrefaction of wood chips followed by pelletization (pre-torrefaction) and pelletization of wood chips followed by torrefaction (post-torrefaction). In this communication, the preparation devices for the torrefied pellets were not identical for each specimen because various torrefied pellets were received from various torrefaction and pelletization devices (Table 1). The diameter of each pressing die was 6 mm, and no binders were used in the pelletization. Mass yield during torrefaction was calculated as the weight percentage of the torrefied material divided by raw material on a dry basis.

Table 1. Wood pellets used in this study.

Pellet Sample Number	Raw Material	Pellet Type [1]	Torrefaction Condition	Pelletization Condition [2]
1	Cedar	Normal	-	Flat die
2	Cedar	Normal	-	Flat die
3	Cedar	Normal	-	Flat die
4	Cedar	Normal	-	Flat die
5	Cedar	Pre-torrefied	Vacuum oven at 220 °C [3]	Flat die
6	Cedar	Pre-torrefied	Electric oven at 290 °C [4]	Flat die
7	Cedar	Post-torrefied	Vacuum oven at 220 °C [3]	Flat die
8	Cedar	Post-torrefied	Electric oven at 240 °C [4]	Flat die
9	Cedar	Post-torrefied	Super-heated steam oven at 300 °C [5]	Flat die
10	Oak	Normal	-	Flat die
11	Oak	Pre-torrefied	Electric oven at 280 °C [4]	Flat die
12	Oak	Post-torrefied	Electric oven at 240 °C [4]	Flat die
13	Cedar	Normal	-	Ring die
14	Cedar	Pre-torrefied	Rotary kiln at 215 °C [6]	Ring die
15	Cedar	Post-torrefied	Vacuum oven at 240 °C [3]	Flat die
16	Cedar	Post-torrefied	Vacuum oven at 250 °C [3]	Flat die

[1] Pre-torrefied: torrefaction of wood chips followed by pelletization. Post-torrefied: pelletization of wood chips followed by torrefaction. [2] Flat-die: flat die type pelletizer at laboratory scale (normal capacity: 30 kg/h). Ring-die: ring die type at demonstration scale (normal capacity: 500 kg/h). No binders were used in the palletization. [3] Original vacuum oven (Yasujima Corp., Kanazawa, Ishikawa, Japan). No holding time was set in the heating period. [4] Inert gas oven (Advantec, Tokyo, Japan) [17,18]. [5] Superheated steam oven was batch-type original model (Yasujima Corp., Kanazawa, Ishikawa, Japan). No holding time was set in the heating period [31]. [6] Commercial rotary kiln (Actree Corp., Hakusan, Ishikawa, Japan). The residential time was 60 min. Detailed information of the kiln has already been introduced [32].

2.2. Water Resistance Test

2.2.1. Indoor Test

Indoor water vapor adsorption tests were performed for various normal and torrefied wood pellets (pellet sample numbers 1–12 in Table 1) in three thermo-hygrostat rooms at the Forestry and Forest Products Research Institute. Initially, three sets of commercial grass-scale plates, on which 6–10 specimens were placed, were prepared. Each plate was then placed in a thermo-hygrostat room with a temperature of 20 °C and 45% relative humidity (RH) and kept until the weight reached equilibrium (approximately 2 weeks). The plates were then moved to another room at a temperature of 20 °C and 65% RH, and then to another room at a temperature of 20 °C and 85% RH. Moisture content changes were calculated based on the weight change of each pellet in relation to the oven-dried

(105 °C) weight of the pellet measured at the end of the test. The oven-dried method used to determine the moisture content was performed according to JIS Z 7302-3 [33]. The moisture content was finally calculated by averaging the values calculated for the three sets of plates.

2.2.2. Outdoor Test

For the outdoor test, the changes in the shape and moisture content of the normal and torrefied pellets (pellet sample numbers 13–16 in Table 1) were investigated during weathering. The test location was a weathering test field at the Forestry and Forest Products Research Institute (latitude, 36°06 N; longitude, 140°06 E). A commercially available laundry mesh bag (400 × 300 mm) containing 500 g of pellets was placed on a stainless-steel mesh stand (355 × 270 × 42 mm) installed 1 m above the ground. The flame of the standby binder clips tightened the edge of each mesh bag. Figure 1 shows an overview of the outdoor weathering test. The exposure of the pellets was conducted both with and without a roof. The period was either from April 9 to 22 May 2015 (average temperature, 16.4 °C; maximum temperature, 28.8 °C; minimum temperature, 1.5 °C; average RH, 72.5%; minimum RH, 10.0%; total precipitation, 157.5 mm [34]) or from 16 July 2015 to 2 February 2016 (average temperature, 15.4 °C; maximum temperature, 36.1 °C; minimum temperature, 6.3 °C; average RH, 77.1%; minimum RH, 13.0%; total precipitation, 830 mm [34]). The pellet weight was measured, and the shape was photographed approximately every 2–4 weeks. The moisture content was estimated by calculating the weight change of each bag and the initial moisture content of the subsample of the pellets using the oven-dried method at 105 °C according to JIS Z 7302-3. After exposure, the diameters of 50 randomly extracted pellets were measured using a digimatic caliper (CD-20PSX, Mitutoyo, Japan) according to ISO 17829:2015, while the pellet lengths were not measured.

Figure 1. Overview of outdoor weathering test.

3. Results and Discussion

3.1. Indoor Test

The changes in moisture content during the exposure of pellets of each type to the three RH conditions are shown in Figure 2, and the standard deviation value for equilibrium moisture content at each RH condition is shown in Table 2. In all samples, the moisture content increased with increasing RH until reaching close to equilibrium. The moisture content at equilibrium was lower for torrefied pellets than for normal pellets under all RH conditions. The initial moisture content of the normal cedar pellets (pellet number 3) was lower than some of the torrefied cedar pellets, such as pellets number 5 and 6; however, the equilibrium value at RH 85% was greater than that of the torrefied pellets; it reached almost the same level as that of the other normal pellets. The increase in moisture content at the initial exposure stage was slow for the pre-torrefied pellets, which are expressed as solid symbols in Figure 2. After torrefaction, wood chips become brittle and are easier to comminute into smaller particles [17,19,35,36]. Therefore, torrefied pellets consisting of smaller particles may impede the permeation of water. Further, the post-torrefied pellets (open symbols) exhibited lower moisture content than the pre-torrefied pellets. This result is supported by the results obtained by Ghiasi et al. [25], who impregnated torrefied pellets with water and examined the time-dependent change in mass reduction when they were dried. The post-torrefied pellets exhibited a small amount of mass reduction, indicating that the amount of remaining water was also small. Figure 3 shows the relationship between the pellets' mass yield during torrefaction and final moisture content at 85% RH. Regression values were calculated according to the literature [26]. Although the regression values were not high, the equilibrium moisture content tended to decrease as the mass yield decreased. Previous studies have shown cases in which torrefied biomass moisture content decreases in relation to rising heat treatment temperature [23]. Further, there tends to be a decrease in the number of hydrogen bonds as torrefaction proceeds [25,37], indicating that the decrease in water uptake is related to increasing torrefaction severity (mass reduction). Further studies are needed to evaluate additional factors other than mass yield affecting the water resistance of torrefied pellets.

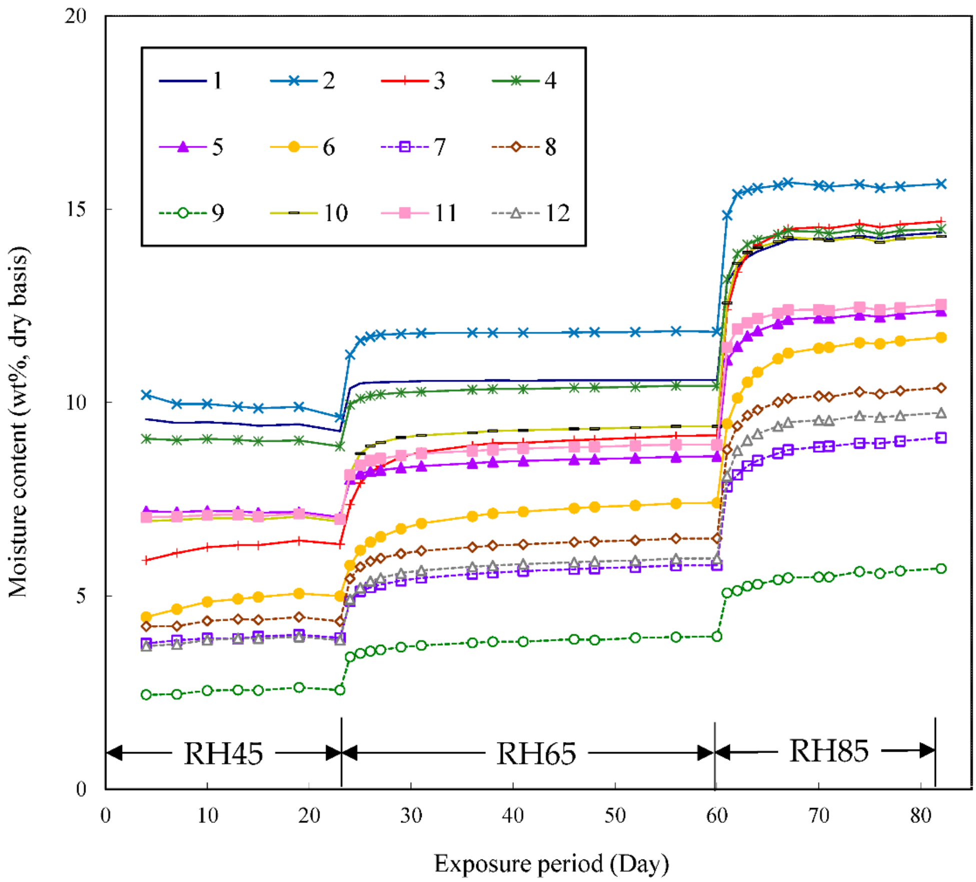


Figure 2. Changes in the moisture content of each pellet type during exposure to three stages of relative humidity (RH) conditions. Each item in the legend corresponds to a pellet sample number shown in Table 1. Solid, open, and the other symbols represent pre-torrefied, post-torrefied, and normal pellets, respectively.

Table 2. Standard deviation value for equilibrium moisture content at each relative humidity (RH) condition.

RH	Pellet Sample Number											
(%)	1	2	3	4	5	6	7	8	9	10	11	12
45	0.113	0.064	0.141	0.043	0.031	0.118	0.041	0.109	1.066	0.042	0.099	0.032
65	0.064	0.217	0.175	0.016	0.245	0.378	0.284	0.127	1.313	0.072	0.191	0.165
85	0.102	0.155	0.117	0.086	0.076	0.262	0.087	0.012	1.248	0.087	0.146	0.213

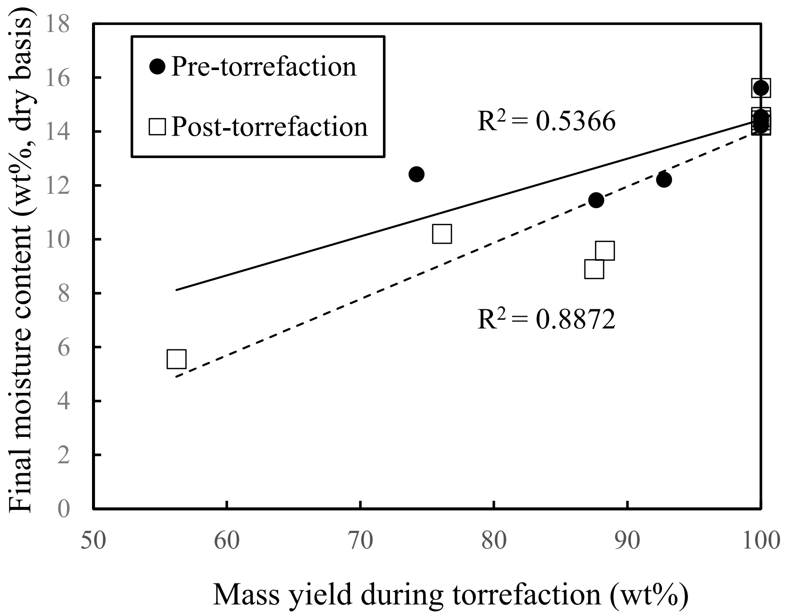


Figure 3. Relationship between the mass yield during torrefaction and the final moisture content of the pellets at 85% RH. Mass yield of 100% refers to normal pellets.

3.2. Outdoor Test

The pellet moisture contents and diameters before and after the outdoor weathering test are shown in Table 3. Under a roof, the moisture content measured at the end of the period was comparable to the results of the indoor test. The equilibrium moisture content of the pellets used in this study depended on the ambient temperature and humidity conditions. At the same time, the moisture content was higher without the roof than with it. On a dry basis, the average equilibrium moisture content of the wood in the test area was ~14% [38], indicating that such high moisture contents for pellets with the no-roof condition were affected by precipitation rather than humidity. In the no-roof condition, the moisture content of the normal pellets significantly increased to over 100% on a dry basis, whereas the increase in the torrefied pellets' moisture content was much lower. The final moisture contents to initial moisture contents were 4.3–8.3 times for pre-torrefied and 1.9–2.8 times for post-torrefied, indicating that the ratio of the increase in moisture content for the pre-torrefied pellets tended to be more than double that for the post-torrefied pellets. The maximum moisture content for the pre-torrefied pellets was 39.9%, which is not within the value of the ISO/TS 17225-8 [39], although, theoretically, it could enable combustion. The final moisture content was ~10% for the post-torrefied pellets; this indicates that the increase in moisture content was much lower for these pellets than for those of the other group.

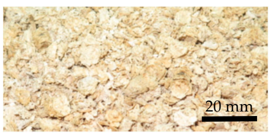
Table 3. Moisture content and pellet diameter during the weathering test.

Weathering	Pellet Type		Period *	Moisture Content (wt%, Dry Basis)		Diameter (mm)	
				Initial	Final	Initial	Final
With roof	13	Normal	1	7.2	14.2	6.06	6.14
	14	Pre-torrefied	1	5.3	14.9	6.04	6.15
	14	Pre-torrefied	2	4.8	9.7	—	—
	15	Post-torrefied	2	4.3	7.7	5.81	5.83
	16	Post-torrefied	2	4.4	6.4	—	—
Without roof	13	Normal	1	7.2	102.4	6.06	n.a. **
	14	Pre-torrefied	1	5.3	22.7	—	—
	14	Pre-torrefied	2	4.8	39.9	6.04	6.54
	15	Post-torrefied	2	4.3	12.0	5.81	5.91
	16	Post-torrefied	2	4.4	8.4	—	—

* 1: From 9 April to 22 May 2015. 2: From 16 July 2015 to 2 February 2016. ** Not available for determination because of lost shape.

Observation of the pellet shape after weathering revealed that the normal pellets had almost entirely lost their shape, while the pre-torrefied pellets showed partial swelling and collapse (Table 4). In contrast, no visual disintegration was observed in post-torrefied pellets. The average diameter of the post-torrefied pellets after weathering was only 5.9 mm, indicating limited swelling. Kubojima et al. [27] measured the dimensional change in the transverse direction when various pellets were swollen in water. They showed that the changes in post-torrefied pellets were small, supporting a result similar to that obtained in this study. Ghiasi et al. [25] have also noted that post-torrefied pellets retain their outer layer, preventing water from accessing their insides. This consideration can be applied to the results of this study, along with the assumption that the tarry material thermally generated during torrefaction bonded between the pellet particles, enhancing the suppression of water penetration. Therefore, the results demonstrate that the torrefaction of pellets can minimize dimensional change and water uptake. However, post-torrefied pellets have the disadvantage of a lower energy density compared with pre-torrefied pellets because of weight loss during torrefaction [18,30]. Further, torrefied materials tend to be less biodegradable [23,31,40]. Both the high energy density and water resistance of torrefied pellets could lead to advantages for storage in small-scale usage. Thus, pre-torrefied pellets could be suitable for indoor storage because of their higher energy density, while post-torrefied pellets are likely more suitable for outdoor storage because of their superior water resistance. Such uses might include daily outdoor cooking for individuals/groups or long-term storage fuel for communities in the case of natural disasters, such as typhoons or earthquakes.

Table 4. Visual observation of the pellets before and after weathering without a roof.

Pellet	13 Normal, Period 1	14 Pre-Torrefied, Period 2	16 Post-Torrefied, Period 2
Before weathering			
After weathering			

Japan experiences relatively heavy rainfall and snowfall, as well as frequent natural disasters, such as earthquakes and typhoons. Torrefied wood pellets could be a practical wood-based bioenergy source in residential areas because of their suitability for long-term storage, high energy density, and small size, helping to establish a resilient society. As highlighted, the production cost of torrefied pellets is higher than that of normal wood pellets [7,9,12,16]. In addition, the experimental conditions and results presented in this communication are limited with regard to evaluating the important physical and chemical properties associated with water resistance. Pellet strength, including mechanical durability, is another important factor during storage, and the loss of mechanical durability and physical structure during liquid water digestion has been qualitatively evaluated [41]. Further studies are planned to investigate various related topics, including the physical and chemical changes in fuel properties and the economic value of the small-scale use of local woody biomass torrefaction.

4. Conclusions

Various torrefied pellets fabricated using different methods were exposed to hygroscopic indoor and outdoor conditions to examine their water resistance. The results revealed that, in comparison with normal pellets, the torrefied pellets had lower moisture content when exposed to hygroscopic conditions. Further, the post-torrefied pellets, prepared by pelletization followed by torrefaction, had a much lower moisture content than the pre-torrefied pellets, prepared by torrefaction followed by pelletization. The former exhibited almost no change in diameter after the outdoor weathering test. Previously, torrefied pellets have been predominantly used for industrial power generation. The differences in waterproof property and energy density by the different schemes could expand the application of torrefied pellets. For example, torrefied pellets could also be used on a small scale for consumers (personal or community) because they can be placed in long-term storage. Overall, torrefied pellets could contribute to the development of wood-based bioenergy resources for small-scale usage, functioning as upgraded waterproof energy carriers with high energy density. However, further studies are needed to evaluate the physical and chemical property changes during storage, transportation, and usage and analyze their economic value.

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