

Effect of Torrefaction on Properties of Pellets Produced from Woody Biomass

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ABSTRACT: Torrefaction has been recognized as a promising strategy to improve handling and storage properties of wood-based pellets, thus producing a uniform-quality commodity with high energy density and hydrophobicity. In this work, pellets produced from spruce stem wood, bark, and forest residues were torrefied in a bench-scale tubular reactor at 225 and 275 °C with two residence times (30 and 60 min). The effects of torrefaction on general properties, grindability, mechanical properties, hydrophobicity, and microstructure of the studied pellets were investigated. The increase of torrefaction severity reduced mass yields, but the heating values and the fixed carbon content of the torrefied pellets considerably increased. The grindability of raw pellets was substantially improved after torrefaction treatment. The energy required for grinding torrefied pellets is less than 50% of the energy needed for grinding the untreated pellets. In comparison to untreated pellets, the particles from ground torrefied pellets have clearly smaller sizes in a narrower size range. The increase of torrefaction severity improved hydrophobicity of the pellets, which have high resistance to water uptake and maintain their integrity after immersion testing. Upon torrefaction treatment, the durability and tensile strength of the pellets slightly decreased. Scanning electron microscopy analysis results show that particles from wood pellets torrefied at 275 °C lost their fibrous structure with an evident decrease of length/diameter ratios compared to untreated wood pellets. The particles from ground torrefied pellets are more uniform in terms of shape and size. Torrefaction can considerably improve grindability and uniformity of wood-based pellets and make them more acceptable in pulverized fuel applications.

1. INTRODUCTION

Renewable energy sources are important to meet the ever increasing energy demand of modern society and replace the use of fossil fuels while minimizing greenhouse gas emissions and global warming.¹ Among the renewable energy sources, bioenergy is an attractive option and has great potential for further development. Via thermochemical and biological conversion routes, various energy products can be produced from biomass materials in the form of solid, liquid, and gas, which can be used directly or upgraded for different end-use applications.

Woody biomasses are among the most abundant biomass materials that are currently widely used for production of renewable bioenergy and high-value chemicals and materials.² However, further utilization of woody biomass on a large scale has been restrained as a result of drawbacks of them as solid fuel. Our previous work shows that woody biomasses generally have a high moisture content, low bulk and energy density, and poor grindability.³ In addition, different parts from one tree have different physicochemical properties that need to be considered for further logistics and utilization.³ With heterogeneous properties, conversion behaviors (i.e., gasification) of woody biomasses are different with the formation of various intermediates (i.e., char) and final products that have different reactivities.⁴ These limitations greatly affect behavior and efficiency of woody biomass conversion into energy. In addition, as a result of these limitations, delivery,

storage, and handling of the woody biomasses are complex and challenging compared to coal. This is more evident for large-scale utilization of woody biomasses to replace or co-fire with coal in existing power plants. With low bulk and energy density, transport capacity of conveyors and reclaimers for woody biomasses must be multifold larger than that for coal, to ensure sufficient feedstock supply and smooth operation of boilers.⁵ Moreover, the fibrous nature of woody biomass makes it difficult to grind and pulverize to obtain the desired particle size distribution.^{6,7} Even after grinding, woody biomass particles with fibrous appearance can be cohesive and readily stick together, causing plugging and clogging of feeding systems. Wood particles with heterogeneous size, shape, and surface roughness can also increase complexity of flow behaviors during conversion processes and cause a variety of flow-related challenges.⁸

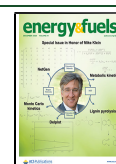
Torrefaction has been studied and tested as a pretreatment method to overcome these drawbacks of woody biomasses, thus producing an easily grindable, more spherical and uniform, and high-quality energy commodity for different

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applications. Torrefaction involves thermal treatment of biomass at 200–300 °C in an inert or oxygen-reduced atmosphere for a certain holding or residence time. During torrefaction, major constituents of biomass materials, primarily hemicellulose, are degraded as a result of depolymerization, demethoxylation, bond cleavage, and condensation reactions, driving out moisture and parts of the volatile compounds.^{1,9,10} Upon torrefaction, the heating value and hydrophobicity of biomass can be considerably improved as well as susceptibility to fungal and microbial degradation. As a result of decomposition of biopolymers, the torrefied biomass is more brittle and friable with significantly improved grindability compared to the raw biomass material.¹⁰ With an increase of torrefaction severity (i.e., torrefaction temperature and residence time), the woody biomass can be upgraded with respect to key qualities as solid fuel, such as heating value, grindability, and powder flowability.¹ However, the bulk density of the torrefied woody biomass is still low, which can be even lower than that of the raw biomass.⁹ It makes further transporting and logistics of torrefied woody biomass economically challenging. Pelletization subsequent to torrefaction has therefore been frequently suggested and studied to improve the energy density per unit volume of torrefied woody biomass to be stored and transported, to facilitate efficient logistics and usability throughout the value chain.^{5,11} Previous studies report that pelletization of torrefied woody biomass can be challenging and often associated with problems related to the pelletization process and resulting pellet quality.^{12–18} The moisture content of the biomass is important during the pelletization process as a result of its plasticizing effect and role to lower the glass transition temperature of cell wall polymers binding particles.¹⁶ Torrefaction treatment considerably removes the initial moisture content of the woody biomass, which can considerably reduce the binding affinity of torrefied materials as they are pelletized. Further, modification of cell wall polymers, loss of hemicellulose, and dehydroxylation of lignin at higher torrefaction severities result in reduced hydrogen bonding during densification.^{16,17} As a result of this, a conditioning step is often needed to increase the moisture content to realize pelletization of torrefied woody biomass.¹⁷ To pelletize woody biomass with sufficient mechanical strength, bond formation among particles/grains is critical, which can be due to mechanical interlocking, solid bridges, and intermolecular forces.^{15,16} However, the particles from ground torrefied biomass are brittle and difficult to bind together through mechanical interlocking.¹⁶ Binding agents are therefore often needed for producing high-quality pellets from torrefied woody materials, which helps to increase adhesive, cohesive, and interfacial forces between solid particles.^{14,15} Moreover, as a result of low bonding forces and loss of natural binding characteristics of biomass after torrefaction, high pellet die temperature and high compression pressure are required to enable pelletization of the torrefied materials and ensure production of high-quality solid pellets. A high pelletization temperature favors thermal softening of lignin that can act as a natural binding agent to increase bond forces between particles.¹⁷ However, high pelletization temperature and compression force cause an increase of friction generated in the pellet press channel and increase energy required for pelletization. During a continuous pelletization process, the stiff and abrasive nature of torrefied biomass might reduce the shelf life of the pellet die, which needs frequent maintenance.^{17,18} In addition, milling of torrefied biomass often

generates a large amount of dust that negatively affects handling and processing of the material before pelletization, which also amplifies the risks of fire and explosion.⁶

Torrefaction subsequent to pelletization is a promising alternative to accelerate the application and commercialization of torrefaction within an existing bioenergy value chain. The wood pellet industry is a well-established modern bioenergy industry.⁶ Wood pellets are traded globally as a bioenergy commodity that is used in various scale applications. In the context of increasingly stringent regulations, direct combustion and co-firing of wood pellets have been recognized as effective measures to replace coal in large-scale power plants.¹⁸ In addition, there is great interest in converting woody biomass into syngas through gasification for generating power and producing fuels and chemicals. Pelletization considerably improves the quality of woody biomass as solid fuel and reduces shipping costs and complexity of handling and storage. However, utilization of wood pellets in existing coal-fired plants or entrained flow gasifiers is currently still rather limited as a result of the fibrous nature of wood, uneven characteristics, and capital costs needed for storing pellets and handling and processing them before the introduction to the reactors.^{19,20} Torrefaction subsequent to pelletization could overcome many of the drawbacks of the wood pellets and increase availability of wood pellets for different conversion routes. Moreover, direct torrefaction of wood pellets can be a final step integrated in the existing plants to avoid challenges that might be encountered during pelletization of torrefied wood.⁵ Nowadays, wood pellets are mainly produced from wood chips, sawdust, and planer shavings. There is continuous interest to exploit more raw materials, i.e., bark and forest residues, for power and fuel production. In comparison to wood chips and sawdust, these raw materials have high heterogeneity in terms of shape, size, and composition.^{3,4} Pelletization subsequent to torrefaction can be a promising way to improve and homogenize properties of these unconventional biomass materials for further smooth and efficient conversion. It will also be important to improve economics across full-value chains from the forest to the power plant. Nevertheless, there are only a few studies that have investigated torrefaction subsequent to pelletization. Most of these studies focus on pellets produced from virgin wood chips and sawdust.^{20–25} To the best of our knowledge, there are no studies that have been carried out to investigate torrefaction behaviors of pellets produced from bark and forest residues and analysis of properties of the torrefied pellets.

The objective of this work is to (1) experimentally investigate the torrefaction of wood, bark, and forest residue pellets under different process conditions (i.e., temperature and residence time) and (2) evaluate changes in properties of the torrefied pellets.

2. EXPERIMENTAL SECTION

2.1. Materials. In the present work, stem wood, bark, top, and branches from Norway spruce trees were collected for pellet production. The raw materials were dried at a low temperature (<40 °C) to a moisture content of 4–8% and stored for further pelletization. The pre-dried materials were first hammer-milled (Vertica hammer mill DFZK-1, Bühler AG, Uzwil, Switzerland) pass a 4 mm sieve. The milled forest residues, bark, and stem wood were conditioned to moisture contents of 12.4, 11.0, and 12.7%, respectively, before further pelletized using a Bühler DPCB pellet mill (capacity of about 500 kg h⁻¹, Bühler AG, Uzwil, Switzerland) with a

rotating die and press channel length (PCL) of 65 mm with an inner diameter of 8 mm.

2.2. Torrefaction Experiment. The torrefaction experiments were performed using a bench-scale tubular reactor. The reactor includes a tubular vessel, an electrical gas preheater with a temperature controller, a condensate collector, and a gas supply system. For one torrefaction experiment, about 150 g of pellets was charged into the vessel at room temperature and then sealed. Afterward, the sealed vessel was placed inside an electrically heated furnace and connected with the inert gas supply system and the condenser. The temperature in the furnace is monitored by three thermocouples located on the top, middle, and bottom of the furnace. The tubular vessel was continuously purged with 1 L min⁻¹ nitrogen to displace air before heating and generate an inert atmosphere, thereby avoiding possible oxidization and ignition of the sample inside. After purging with nitrogen at room temperature for 1 h, the pellets were heated without nitrogen purging at a heating rate of 10 °C/min to two different final temperatures, 225 and 275 °C. The holding time for one sample at each final temperature was 30 and 60 min, respectively. Both the final temperature and holding time are commonly chosen for torrefaction, in accordance with published work.^{3,20,21} After each torrefaction experiment, the reactor was purged with continuous nitrogen flow and cooled to room temperature. The cooled torrefied pellets were after each run discharged and weighed to calculate the solid yield. The solid product mass yield of one torrefaction experiment was calculated as the percentage of initially loaded pre-dried biomass sample, as follows:

$$\text{mass yield} = \left(\frac{m_{\text{dry basis torrefied solid}}}{m_{\text{dry basis feedstock}}} \right) \times 100 \quad (1)$$

The torrefied pellets were sealed in airtight plastic bags and stored at room temperature for further analyses.

2.3. Analysis of Pellet Properties. **2.3.1. Proximate and Ultimate Analyses.** The studied pellets were characterized before and after torrefaction. Proximate analysis (volatile matter, fixed carbon, and ash content) was performed by following procedures described in ASTM standards E872 and D1102. For each sample, the analysis was repeated 5 times to obtain a standard deviation of below 1%. The fixed carbon content of one sample was calculated as the difference between 100 and the sum of volatile matter and ash contents. The volatile matter, ash, and fixed carbon contents of raw and torrefied pellets were reported on a dry basis and are presented in Tables 1 and 2. The elemental composition of raw and torrefied

Table 1. Properties of the Studied Fuels (Calculated on a Dry Basis)

sample	wood pellet	bark pellet	forest residue pellet
moisture content (wt %, as received)	5.42	8.53	8.07
volatile matter content (wt %, db)	84.72	72.38	77.33
ash content (wt %, db)	0.22	3.53	2.62
fixed carbon content (wt %, db)	15.07	24.09	20.05

pellets was analyzed by employing an elemental analyzer (Eurovector EA 3000 CHNS-O elemental analyzer). The oxygen content is calculated by difference. For one analyzed sample, the elemental analysis was repeated 3–5 times and average values of these measurements are presented. The heating values of ground untreated and torrefied pellets were measured using a bomb calorimeter (IKA C2000 calorimeter) based on ASTM standard D5865-03. On the basis of the mass yields and higher heating values (HHVs), the energy density and energy yield for one sample torrefied at different conditions can be calculated as follows:

Table 2. Mass and Energy Yield of Torrefied Pellets (Dry Basis)

feedstock	mass yield (%)	HHV (MJ kg ⁻¹)	energy yield (%)	relative energy density
pre-dried stem wood pellet	100	19.9	100	1
225 °C for 30 min	91.3	20.1	92.22	1.01
225 °C for 60 min	89.1	20.5	91.57	1.03
275 °C for 30 min	77.4	21.5	83.57	1.08
275 °C for 60 min	68.8	22.7	78.18	1.14
pre-dried bark pellet	100	20.3	100	1
225 °C for 30 min	86.0	20.6	87.11	1.01
225 °C for 60 min	81.9	21.0	84.77	1.04
275 °C for 30 min	71.3	22.7	79.58	1.12
275 °C for 60 min	67.0	23.4	77.32	1.15
pre-dried forest pellet	100	20.7	100	1
225 °C for 30 min	86.3	21.1	88.34	1.02
225 °C for 60 min	83.2	21.4	86.28	1.04
275 °C for 30 min	70.8	23.2	79.47	1.12
275 °C for 60 min	65.4	23.4	74.20	1.14

$$\text{energy density} = \frac{\text{HHV}_{\text{torrefied}}}{\text{HHV}_{\text{untreated}}} \quad (2)$$

$$\text{energy yield} = \text{mass yield} \times \left(\frac{\text{HHV}_{\text{torrefied}}}{\text{HHV}_{\text{untreated}}} \right) \times 100 \quad (3)$$

2.3.2. Grindability. Grindability of raw and torrefied pellets was determined by measuring the energy consumption for grinding and the size distribution of the resulting particles. First, the raw and torrefied pellets were ground using a laboratory cutting mill (IKA MF 10.1). The cutting mill used in this study consisted of a cutting blade with a 0.5 mm sieve. Through a top-feed channel, about 50 g of each pellet sample was manually feed into the mill at an even rate. The cutting mill motor was equipped with a circuit breaker to avoid possible motor overloading. The instantaneous power consumption was monitored by a digital wattmeter (Paladin 256-TWKW from Cromptan Instruments), which was connected to a computer for recording every 2 s using a LabVIEW system (National Instruments Corp., Austin, TX, U.S.A.) during the grinding experiment. The ground powder passing the sieve was weighed (Mettler PM1200, Mettler-Toledo AG, Greifensee, Switzerland). The specific energy consumption required for grinding was calculated by integrating the area under the instantaneous power consumption curve (watt seconds) above the idle power baseline with respect to time required for grinding the given amount of sample. The energy consumption required for grinding one sample is expressed per unit mass for comparison purposes. Particle size distributions of ground raw and torrefied pellets were assessed by sieving the material with a vibrating sieving machine (Fritsch Analysette 3 Pro). First 100 g of each ground pellet sample was loaded in a stack of sieves arranged from the largest to smallest openings, which have sizes of 500, 300, 200, 100, and 63 μm. Each sample was sieved with a sieve shaking time of 10 min at 60 Hz. After sieving, the mass collected on each sieve was weighed and presented as a percentage of the initial sample mass.

2.3.3. Mechanical Property. Mechanical properties of the pellets are very important at many stages, from handling, transportation, to end use. Mechanical strength of the raw and torrefied pellets was tested by a pellet harness tester (Amandus Kahl, Germany). Following the procedures described in refs 26 and 27, one pellet was compressed perpendicular to the cylindrical axis direction until identification of failure of the sample. The tensile strength of one pellet was measured by the machine in kilograms by an equivalence between the elastic compression of a spring that moves a piston against the pellet side and the force equivalent mass. The determination of tensile strength is

Table 3. Proximate and Ultimate Analyses of Untreated and Torrefied Pellets (Dry Basis)

feedstock	VM (% db)	ash (% db)	FC (% db)	C (% daf)	H (% daf)	N (% daf)	S (% daf)	O (% daf)
pre-dried stem wood pellet	84.72	0.22	15.07	47.48	6.47	0.09	0.02	45.94
225 °C for 30 min	82.50	0.23	17.67	48.34	6.11	0.13	0.02	45.41
225 °C for 60 min	80.41	0.25	19.34	49.31	6.14	0.12	0.02	44.41
275 °C for 30 min	76.88	0.36	22.76	52.70	5.97	0.13	0.02	41.18
275 °C for 60 min	73.21	0.41	26.38	53.97	5.87	0.12	0.02	40.01
pre-dried bark pellet	72.38	3.53	24.09	48.71	6.12	0.52	0.03	44.62
225 °C for 30 min	69.25	3.67	27.08	50.32	5.65	0.56	0.04	43.42
225 °C for 60 min	67.12	3.98	28.90	52.08	5.51	0.61	0.04	41.77
275 °C for 30 min	61.25	4.18	34.57	55.81	5.18	0.67	0.05	38.29
275 °C for 60 min	59.43	4.35	36.22	57.50	5.00	0.65	0.04	36.82
pre-dried forest pellet	77.33	2.62	20.05	48.60	6.25	0.75	0.04	44.36
225 °C for 30 min	75.34	2.78	21.88	50.74	6.01	0.79	0.06	42.40
225 °C for 60 min	73.81	2.97	23.22	52.16	5.98	0.78	0.05	41.03
275 °C for 30 min	69.97	3.14	26.89	55.64	5.65	0.88	0.04	37.79
275 °C for 60 min	64.85	3.23	31.92	56.20	5.64	0.86	0.05	37.25

considerably dependent upon the observation and identification of the correct mode of failure. It was reported that ideal tensile failure occurs when the studied specimen (i.e., pellet or tablet) fractures into two parts along the loading axis.³¹ Upon identification of the failure, the applied force was recorded and used to calculate the tensile strength. The tensile strength σ_x was calculated by the following formula:

$$\sigma_x = 2m_g/\pi dl$$

where m_g is the force equivalent mass, g is the gravitational acceleration, d is the compact diameter (m), and l is the compact thickness (m).

Mechanical durability and particle density are to critical parameters indicating the physical quality of pelletized materials. In this work, the mechanical durability of each studied pellet was measured using a tumbler (Bioenergy TUMBLER 1000+, Austria) parametrized in accordance with ISO 17831-1. The results were expressed as the percentage of the pellet weight remaining after the test and the initial weight. For each pellet sample, the mechanical durability test was carried out at least 3 times, and an average value is presented. The particle density of a single pellet in the current study was determined by dividing the mass of each pellet by its volume. The diameter and length of an individual pellet were measured using a digital caliper to calculate the volume of it. Over 20 single pellets were randomly selected for pellet density measurement, and average values are presented.

2.3.4. Hydrophobicity. The water uptake tests on raw and torrefied pellets were studied by immersing them in water for a certain testing time (1–72 h) at room temperature. For each immersion test, a solid/water ratio of 1:10 was used and a given amount of pellets was immersed in water in a glass beaker. Upon reaching a certain immersion time, each pellet was gently removed, wiped with a dry cloth, and weighed for each defined time interval (1, 2, 5, 12, 24, 48, and 72 h). The weight difference before and after the immersion test was considered as the amount of water uptake.

2.3.5. Scanning Electron Microscopy (SEM) Analysis. After grinding, the particles from raw and torrefied pellets were examined using a scanning electron microscope (Zessia Ultra, 55 Limited Edition). The SEM images were taken for the ground samples with a particle size in the range of 0.3–0.5 mm and smaller than 0.063 mm. The sample particles were spread on a carbon tape sticking on a sample tab and loaded into SEM for scanning. For particles in the same size range, SEM was operated with the same parameters. It makes it feasible to compare the particles from different samples in terms of size, shape, and morphology.

3. RESULTS AND DISCUSSION

3.1. Torrefaction Mass and Energy Yields. The pellets produced from spruce wood, bark, and forest residues were

successfully torrefied at temperatures of 225 and 275 °C with residence times of 30 and 60 min, respectively. All torrefied pellets kept integrity after torrefaction. Already after torrefaction at 225 °C, the pellets lost their shiny appearance and surface smoothness. The spruce wood pellets became brownish after torrefied at 225 °C, and all pellets became darker as they were torrefied at 275 °C. The color changes with the torrefaction temperature of thermally treated wood materials have been observed and reported in previous studies.^{17–19,28} The previous study results indicated that the color change is linked to changes in and conversion of acid-soluble lignin compounds in the wood material upon heat treatment.²⁸ Table 2 shows the mass yield, energy yield, and HHV of the torrefied pellets as a function of the torrefaction temperature and residence time. The mass yield of the torrefied pellets decreases with an increase in the torrefaction temperature and holding time. The torrefaction temperature gave a more evident effect than the residence time on mass yields for all torrefaction experiments. Although the decreasing trend for an increasing torrefaction temperature regarding mass was similar for all studied pellets compared to spruce wood pellets, the bark and forest residue pellets are more sensitive to the change of the torrefaction temperature. The mass yields of bark and forest residue pellets decreased to 81–86% already at a torrefaction temperature of 225 °C. Such differences might be linked to the initial chemical composition or decomposition rate of the pellets during torrefaction. The main chemical compositions of the spruce wood, bark, and forest residue are considerably different, especially for the content of extractives. The chemical composition of bark is considerably different from stem wood, and extractives are the most abundant compound group for major tree species.^{10,29,30} During thermal decomposition of the wood materials, degradation of extractives can start already at a low temperature (i.e., 130 °C), and most of them will decompose below the temperature of 250 °C. As torrefied at 225 °C, the bark pellets have more extractives that decompose intensively, resulting in a low mass yield, as shown in Table 2. In addition, the inorganic elements in biomass (i.e., K) might also play a certain catalytic role to increase the degradation rate at a given temperature and affect the yield of solid from a torrefaction process. The bark and forest residues normally have a much higher content of potassium than that of stem wood, which promotes decomposition reactions and can partially explain lower solid

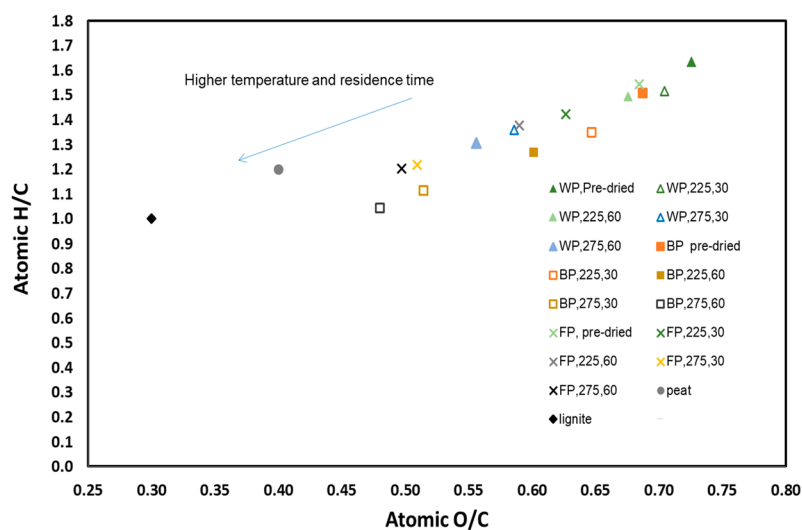


Figure 1. van Krevelen plot of atomic H/C versus atomic O/C for untreated and torrefied pellets.

yields from the bark and forest residue pellets upon torrefaction treatment.¹⁰ Table 2 shows the HHV of raw and torrefied pellets. It is interesting to see that the HHVs of torrefied bark and forest residue pellets are even slightly higher than that of torrefied wood pellets. It is partially related to the high content of extractives and lignin present in the bark and forest residue.¹⁰ The HHV of torrefied pellets increases along with the increase of the torrefaction temperature and residue time. A similar increasing HHV trend upon torrefaction treatment has been reported elsewhere by other researchers.^{20–24} As reported by Bahman et al., dried Douglas fir wood pellets have a HHV of 18.7 MJ/kg, which increased to 22.0 MJ/kg after torrefaction at 260 °C.²⁰ These results are rather comparable to the increase of the HHV of spruce wood pellets from 19.9 to 22.7 MJ/kg after torrefaction at 270 °C in the current work.

3.2. Properties of Torrefied Pellets. Table 3 shows the proximate and ultimate analyses of the raw and torrefied pellets. Torrefaction caused a decrease of the volatile matter content in the studied pellets, which is related to decomposition of chemical constituents, mainly hemicellulose. Similar changes in proximate analysis results were also observed in studies of torrefaction of pine pellets, Douglas fir wood pellets, and pellets produced from sawdusts.^{21–23} The ultimate analysis results showed that the carbon content of torrefied pellets increases compared to that of raw pellets. On the other hand, the hydrogen content of the torrefied samples slightly decreases upon the increase of torrefaction severity, while the oxygen content significantly decreases. Therefore, both O/C and H/C ratios of torrefied pellets decrease, which is mainly related to dehydration, decarboxylation, and depolymerization of the organic fraction during the torrefaction process.²⁴ As a result of decomposition of the organic fraction (i.e., hemicellulose) in biomass materials, the absolute content of carbon-rich lignin in the torrefied biomass increases. The lignin is also linked by various interunit bonds, including ether and carbon–carbon linkages. In addition, these bonds are mainly C–C bonds and have higher energy density in comparison to C–O and C–H bonds.^{1,12,13} Therefore, enrichment of lignin and C–C bonds in torrefied materials can partially explain the increase of HHV of torrefied pellets compared to raw pellets shown in Table 2. The van Krevelen

diagram in Figure 1 clearly shows a general decrease of O/C and H/C ratios of torrefied biomass. The relative decrease of O/C and H/C ratios indicates better combustion characteristics of torrefied pellets, because lower O/C and H/C ratios imply HHV and less thermodynamic loss related to the formation and heating of water vapor, respectively. The characteristics of bark and forest residue pellets torrefied at 275 °C for 60 min are close to those of peat,^{12,24} implying a positive improvement of quality of these materials as solid fuel.

3.3. Grindability. Figure 2 shows milling energy required for grinding untreated and torrefied pellets. There are

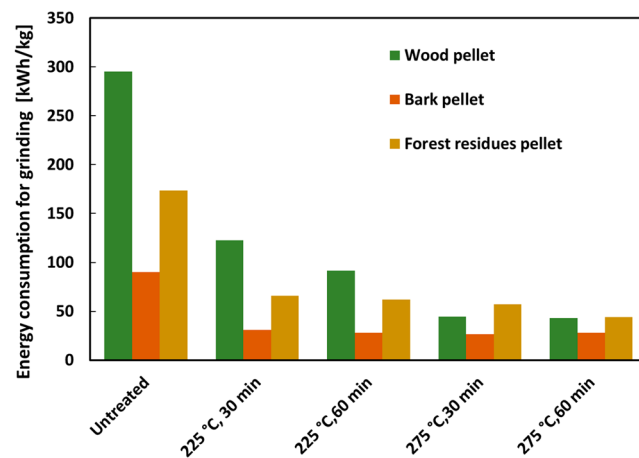


Figure 2. Energy required for grinding untreated and torrefied pellets.

considerable differences in energy consumption for grinding bark pellets, in comparison to wood and forest residue pellets. As shown in Figure 2, only 90 kWh/ton of electricity is needed for grinding untreated bark pellets, which is about half of what is required for grinding untreated wood and forest residue pellets. Grindability of woody biomasses has been studied and reported previously. However, most of the documented knowledge available concerns stem wood chips and pellets produced from stem wood chips, sawdust, and shavings.^{19,21,25} Very little has been published about the mechanical properties and grindability of pelletized bark and forest residues. The grinding of wood and wood-based materials is complicated and

affected by chemical, physical, mechanical, and fracture properties of these materials.²⁹ In comparison to stem wood, the bark has considerably different microstructures and chemical compositions as well. The bark normally consists of sclerenchymatous cells, which are thickened and hardened cells supporting tissues against microorganisms and mechanical damage.^{29,30} These cells are short and rounded and derived from lignification of parenchyma. In contrast, the cells in the stem wood are mostly axially oriented tracheids that are long (i.e., 2–4 mm) but with a small diameter (i.e., 20–30 μm) and have a high length/diameter ratio.³² Stem wood with such a cellular structure has a high strength and tenacity. In brief, the bark lacks a fibrous structure and is much less anisotropic compared to stem wood. Therefore, mechanical properties of bark are inferior to those of wood from a grindability point of view and have a low compression and shear strength. Although the studied bark and wood were milled and pressed through die channels during the pelletization process, the anisotropic and fibrous microstructure of stem wood is preserved. It makes grinding of wood pellets and further size reduction energy-intensive, and it is hard to obtain fine particles. The forest residues studied in the present work are mainly top and branches that contain bark, wood, and twigs. Therefore, the energy required for grinding forest residue pellets is between those used for grinding wood and bark pellets.

Figure 2 demonstrates a substantial decrease of milling energy required for grinding torrefied pellets. For wood pellets, the energy required for grinding the samples was considerably reduced as the pellets were torrefied at 225 °C; only about half of the energy is needed for grinding the untreated (raw) pellets. With an increase of torrefaction severity, the energy required for grinding decreases further from 102 and 91 kWh/ton to 44 and 42 kWh/ton after torrefaction at 275 °C for 30 and 60 min, respectively. For bark and forest residue pellets, the main grinding energy reduction was achieved after torrefaction at 225 °C, which only slightly decreased after torrefaction at 275 °C with 30 and 60 min of residence time. Only a few studies have reported on grinding of biomass pellets torrefied at various conditions. Torrefaction of wood pellets produced from southern yellow pine at 230 °C for 30 min showed a reduced grinding energy from 327.22 to 146.10 kWh kg⁻¹.¹² The same work demonstrates that, with a torrefaction at 290 °C, the grinding energy of pine pellets is reduced nearly 6-fold, from 327.22 to 50.76 kWh kg⁻¹.¹² For the spruce wood pellets studied in the current work, the energy needed for grinding is 295 kWh kg⁻¹, which is close to that reported by Manouchehrinejad et al.¹² After torrefied at 225 and 275 °C, the grinding energy reduced to 122.7 and 44.5 kWh kg⁻¹, which are quite comparable to those reported in ref 12. Similar to the current work, reduction in the grinding energy for torrefied pine wood pellets has been reported.^{21,22} The considerable improvement of grindability of torrefied pellets can probably to a large extent be explained by degradation of hemicellulose. Hemicellulose in nature binds lignin and cellulose fibrils and strengthens the cell wall. Degradation of hemicellulose increases brittleness of woody biomass and makes it easier to grind it into small particles.¹⁶ In addition, decomposition of wood biomass (i.e., extractives and hemicellulose) causes reduction of hydrogen-bonding sites and weakening of attraction forces between solid particles.¹⁷ Therefore, the grindability of torrefied pellets can be significantly improved, which can contribute to saving energy for grinding materials.²⁰

Grindability of pellets may also be referred to as resistance of them being ground to a certain size. Figure 3 shows the effect

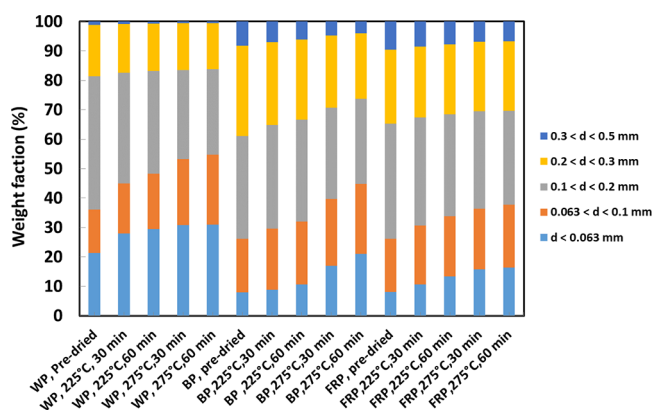


Figure 3. Particle size distributions for the untreated and torrefied wood, bark, and forest residue pellets.

of torrefaction on the size distribution of particles from ground untreated and torrefied pellets. Torrefaction treatment has evident influences on the percent of particles in the different size ranges, especially for the small particles with a size less than 0.1 mm. For the wood pellets, an obvious increase of small particles (8% compared to the reference sample) occurred already at a temperature of 225 °C with a residence time of 30 min. An increase of torrefaction severity resulted in a further increase of small particles. In comparison to the torrefaction residence time, the torrefaction temperature gave more evident effects on the increase of the fraction of small particles. A similar trend of increase of small particles was also reported for grinding and sieving pine pellets torrefied at elevated final temperatures.¹² On the other hand, the particle distribution of raw and torrefied bark and forest residue pellets is different from that of wood pellets. The ground raw bark and forest residue pellets contain much higher fractions of particles larger than 0.2 mm. For the ground bark pellets, the fraction of particles with a size in the range of $0.2 < d < 0.3$ mm decreases evidently after torrefaction treatment, as shown in Figure 3. Again, the fractions of small particles ($d < 0.1$ mm) in ground bark and forest residue pellets increase considerably with the increase of torrefaction severity. Figure 4 shows cumulative particle distribution curves of ground raw and torrefied pellets. The particle size distribution curves of ground wood, bark, and forest residue pellets evidently shift toward particles with smaller sizes. Torrefaction causes degradation of wood polymers and bonding forces between particles that formed during the pelletization process. It embrittles biomass pellets, with reduced mechanical strength, which can be much more easily ground, with the production of more fine particles.

3.4. Mechanical Properties. Mechanical properties and particle density are the main quality parameters indicating the quality of biomass pellets. Table 4 shows durability and compression strength of raw and torrefied pellets. The test results showed that raw bark pellets have slightly higher durability in comparison to raw wood and forest residue pellets. The durability of biomass pellets is dependent upon chemical and physical properties of the feedstock, pre-conditioning process (i.e., steam conditioning/preheating), and pelletization process conditions.^{31–33} Considering constant process variables used during pelletization, the durability differences of the raw wood, bark, and forest residue pellets are

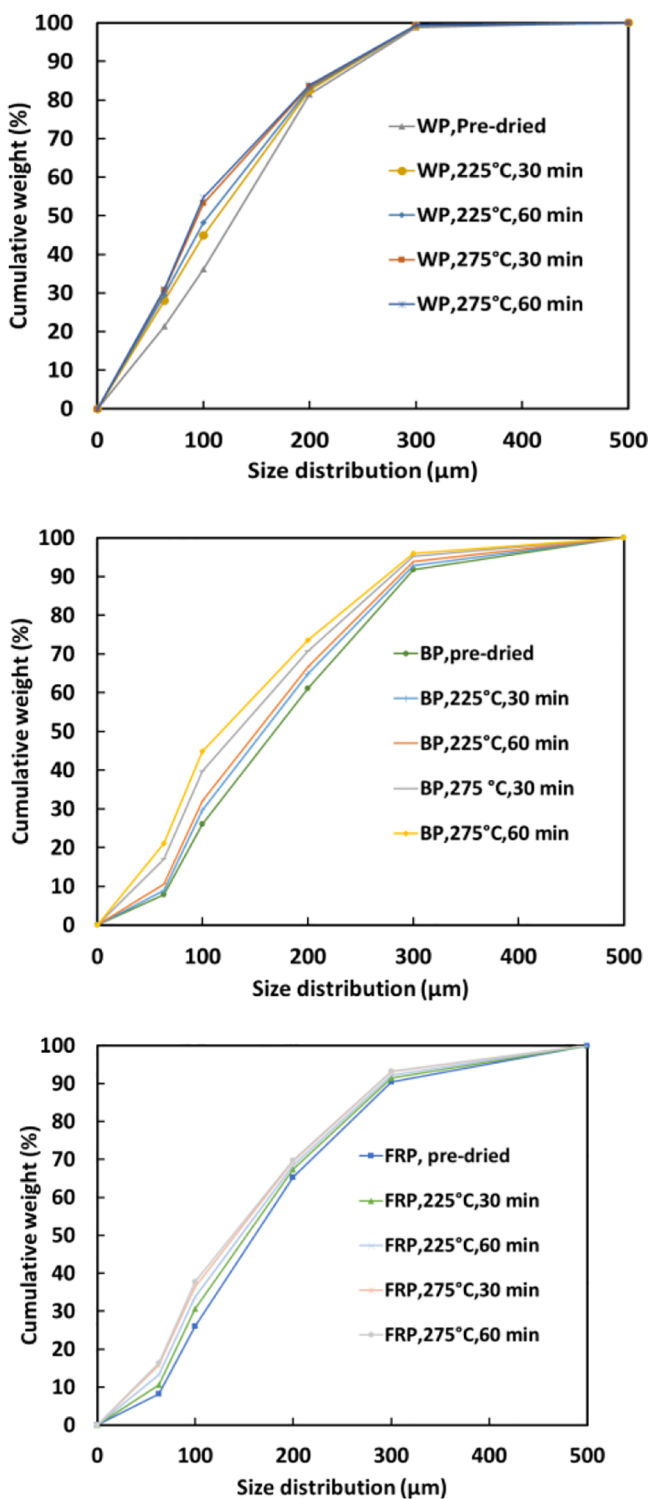


Figure 4. Cumulative particle distribution curves of ground untreated and torrefied pellets.

mainly related to different chemical properties of them. The bark often contains high contents of lignin and extractives (i.e., waxes, resins, and starches) that help to increase bonding of particles and overall pellet durability. Filbakk et al. found that pellets produced from Scot pine bark have higher durability than those produced from stem wood of the same trees.³⁴ In another work, Bradfield and Levi reported that blending of a certain amount of bark to stem wood from different species,

Table 4. Mechanical Properties and Density of Untreated and Torrefied Pellets (Dry Basis)

	wood pellet	bark pellet	forest residue pellet
Durability (%)			
untreated	98.24	98.55	98.37
225 °C for 30 min	98.19	97.93	97.94
225 °C for 60 min	97.85	97.08	97.26
275 °C for 30 min	97.08	96.08	96.00
275 °C for 60 min	96.86	95.45	93.24
Tensile Strength (MPa)			
untreated	2.52	1.93	1.58
225 °C for 30 min	2.41	1.70	1.34
225 °C for 60 min	2.39	1.69	1.38
275 °C for 30 min	2.08	1.54	1.32
275 °C for 60 min	1.99	1.44	1.31
Density (kg m ⁻³)			
untreated	1205	1197	1123
225 °C for 30 min	1156	1103	1101
225 °C for 60 min	1138	1110	1008
275 °C for 30 min	1123	1056	883
275 °C for 60 min	1079	1032	862

including red maple, red and white oak, and sweetgum, caused improvement of durability of produced pellets.³⁵ During the pelletization process, polymers, mainly lignin and extractives in woody biomass, will soften and help bind particles with their plasticizing effect and ability.³⁵ The results shown in Table 4, together with previous findings, support the theory that high contents of lignin and extractives can be advantages for the binding mechanism of particles and improving durability of pellets. After torrefaction treatment, the durability of the studied wood pellets slightly decreased. The downward trends for bark and forest residue pellet durability were similar to that of the wood pellet, and the durability drops were more evident. Durability of pellets is a measure of friability of pellets that normally reflects the amount of fines generated from pellets during handling, transportation, and storage processes. Durability is among the most important mechanical quality metric considered by industry. As shown in Table 4, the durability of the torrefied pellets was also fairly close and comparable to that of the raw material pellets. It is claimed in different standards that the pellet durability index should be higher than 95% for safe transportation and storage.^{35,36} Therefore, for the raw and torrefied pellets studied in the current work, they have sufficient stability under impact and abrasion to prevent dust and fine formation during handling and transport. Another important property of the biomass pellets is the strength that describes the force necessary to crush or damage (i.e., fragmentation and abrasion) a pellet. Table 4 shows that wood pellets have the highest tensile strength value of 2.52 MPa, whereas the forest residue pellets have the lowest tensile strength value of 1.58 MPa. The tensile strength of the studied pellets generally decreased after torrefaction treatment. Again, the torrefied wood pellets have superior tensile strength compared to the bark and forest residue pellets. The pellet strength (compression and tensile strength) decreased drastically at higher torrefaction temperatures, as also reported elsewhere in the literature.¹⁷ With a lower strength, the pellets will be more friable and less energy is required for comminution and grinding into particles. The durability and strength of the pellet mainly depend upon physical forces bonding particles together. Degradation of

hemicellulose and cellulose polymers was suggested as one of main causes for tensile strength loss of biomass pellets upon torrefaction treatment.³⁷ Decomposition of these polymers results in weakening, cleavage, and loss of secondary bonds between hemicellulose and cellulose and cleavage of covalent bonds between hemicellulose and lignin.³⁴ This disrupts and reduces the load-sharing capacity of the lignin–hemicellulose matrix, in which the cell fibrils are encrusted.³² In addition, water and volatiles release from pellets during decomposition of wood polymers. It leads to the formation of more voids in and between the particles in the pellet, reducing contacts, and adhesive forces between particles. A combination of the decomposition of wood polymers and a decrease of bonds between particles causes a decrease of the pellet strength, as reported in previous work.

3.5. Water Uptake. During transportation and storage, biomass pellets might be exposed to rain or high-humidity conditions. It will adversely affect the quality of biomass pellets from different aspects, especially durability and strength. In this work, the hydrophobic or water resistance of raw and torrefied pellets is tested by immersing them in water for certain holding times. During the tests, it was observed that swelling and disintegration of raw wood and forest residue pellets occurred within a short time after they were immersed in water. In contrast, the bark pellets retain a rather good form stability but also lost structural integrity after being immersed in water for about 4 h. The water uptake results of raw pellets are consistent with previous work. Similar poor stability of pellets produced from sawdust and logging residues were observed as they were immersed in water for 30 min at 20 °C.²⁹ The higher water resistance of bark pellets was explained by higher contents of hydrophobic extractives than the other studied pellets. In comparison to raw pellets, the torrefied pellets have low water uptake capacity and retained an intact structure after the testing period. An overall drop in water uptake of torrefied biomass pellets compared to untreated pellets has previously been reported.^{17,22,24} During torrefaction, decomposition of biomass (mainly hemicellulose) results in destruction of the hydroxyl (OH) group in the wood polymer and change of the structure. It leads to a reduction of hydrogen-bonding sites that can bond water, making it more difficult for torrefied material to adsorb water.^{12,22,24} Figure 5 shows the change of the moisture content of the torrefied pellets over the immersion period. In general, the pellets torrefied at a high temperature have a low water uptake capacity, which can be mainly due to more intensive degradation of wood polymers and reduction of hydrogen-bonding sites. In addition, a larger amount of volatile organic compounds and tar will be generated during decomposition of biomass at an increasing torrefaction temperature. These compounds might retain or condensate inside the microstructure of the pellets, obstructing the diffusion and passage of water through the solid and adsorption of water during the immersion consequently.²² As shown in Figure 5, all test pellet samples eventually reached saturation points, as reported in other similar studies.²² Although torrefied wood and forest residue pellets retained their initial shapes, swelling of them during immersion tests were observed after a certain immersion time. It indicates poorer water resistance of the torrefied wood and forest residue pellets compared to the bark pellets. Such differences between water uptake capacity of studied pellets can be partially related to the chemical compositions of them. For example, bark pellets contain more hydrophobic groups in cell

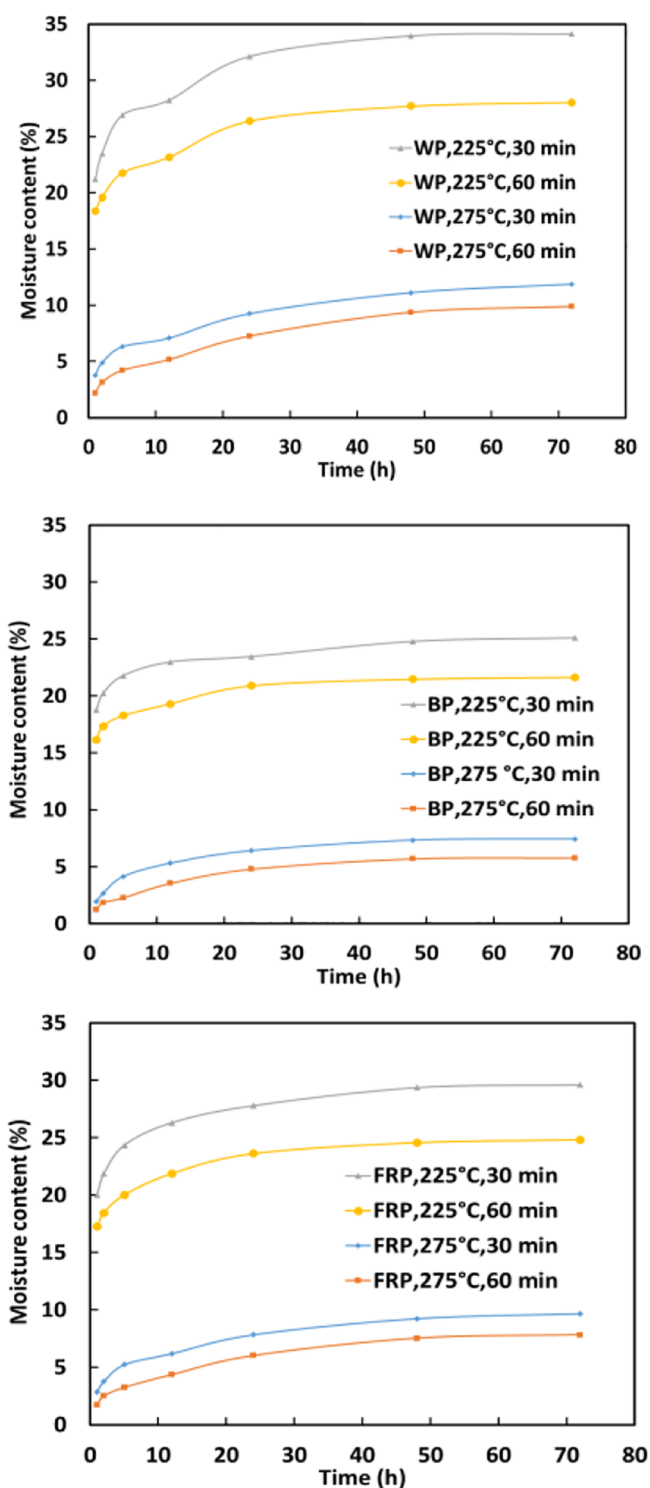


Figure 5. Moisture content of torrefied pellets after immersing in water for different durations: (a) wood pellets, (b) bark pellets, and (c) forest residue pellets.

wall polymers, which are preserved somehow after torrefaction treatment and limit water adsorption capacity of the torrefied bark pellets. Moreover, the chemical compositions of the wood, bark, and forest residue pellets are different and undergo different reaction paths, forming different intermediate and final products. In comparison to wood and forest residue pellets, more hydrophobic groups will form in bark pellets during the torrefaction process, which replace hydrophilic

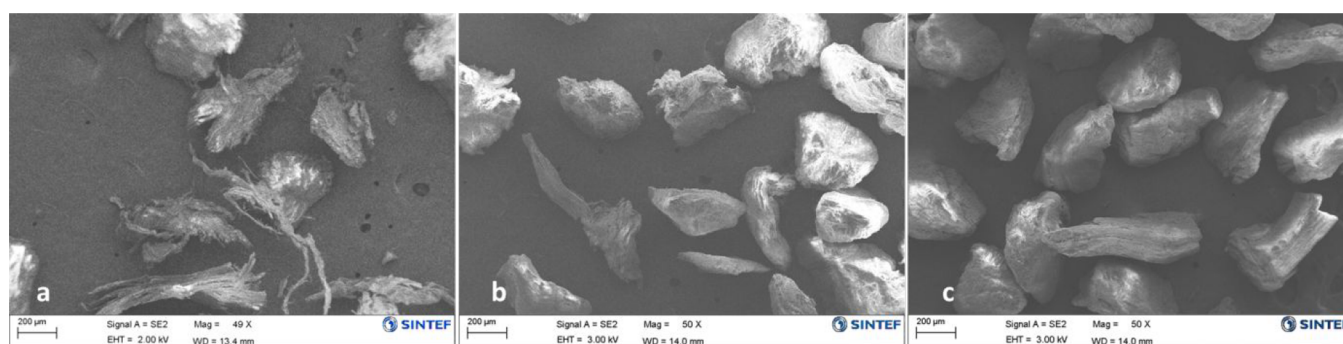


Figure 6. SEM images of the ground particles ($0.3 < d < 0.5$ mm) from wood pellets: (a) untreated, (b) torrefied at 225 °C for 60 min, and (c) torrefied at 275 °C for 60 min.

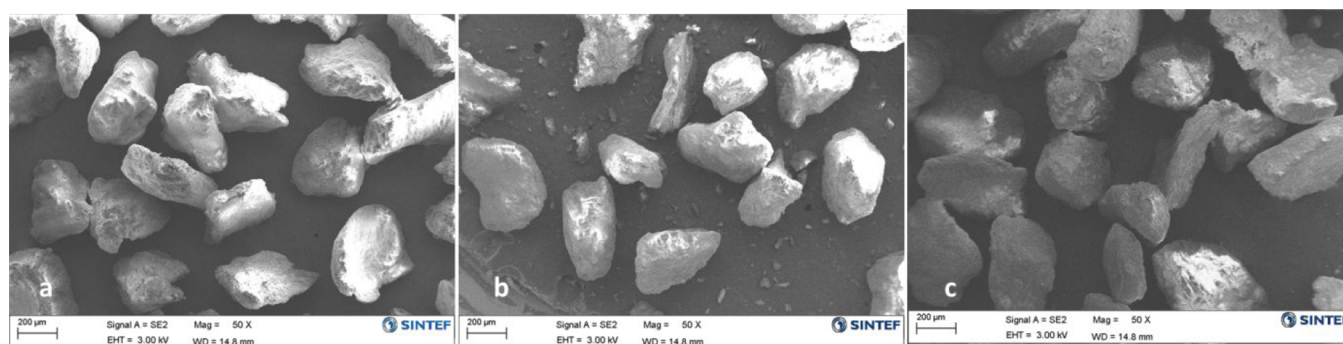


Figure 7. SEM images of the ground particles ($0.3 < d < 0.5$ mm) from bark pellets: (a) untreated, (b) torrefied at 225 °C for 60 min, and (c) torrefied at 275 °C for 60 min.

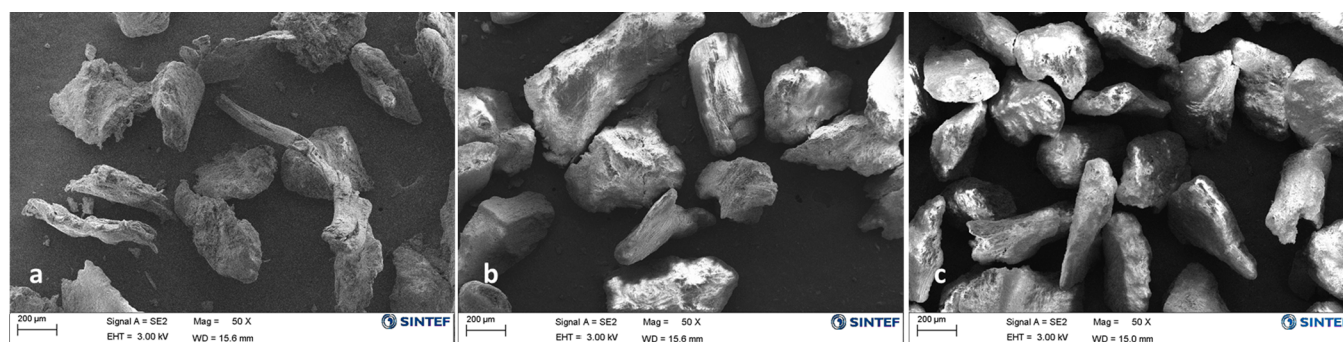


Figure 8. SEM images of the ground particles ($0.3 < d < 0.5$ mm) from forest residue pellets: (a) untreated, (b) torrefied at 225 °C for 60 min, and (c) torrefied at 275 °C for 60 min.

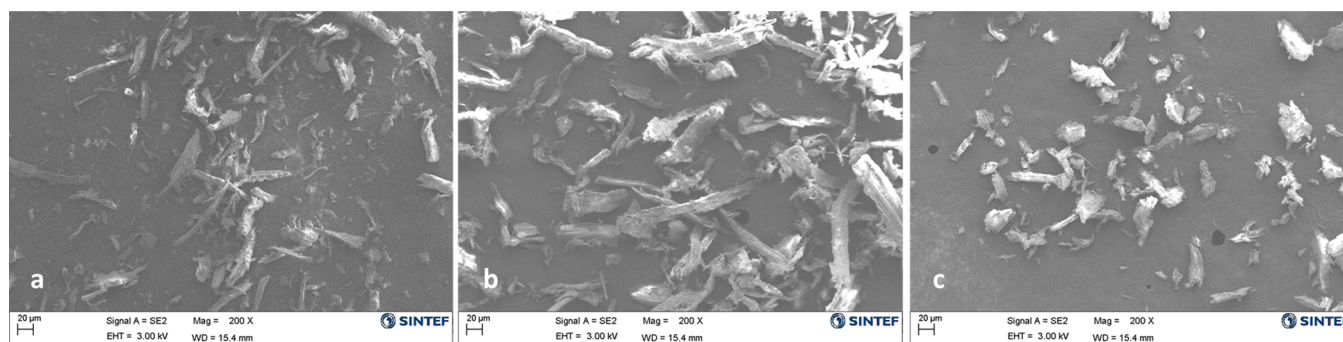


Figure 9. SEM images of the ground particles ($d < 0.063$ mm) from wood pellets: (a) untreated, (b) torrefied at 225 °C for 60 min, and (c) torrefied at 275 °C for 60 min.

groups initially being in the raw pellets and enhance water resistance of torrefied pellets.

3.6. SEM Analysis. The microstructure and morphology of ground raw and torrefied pellets were examined by a scanning

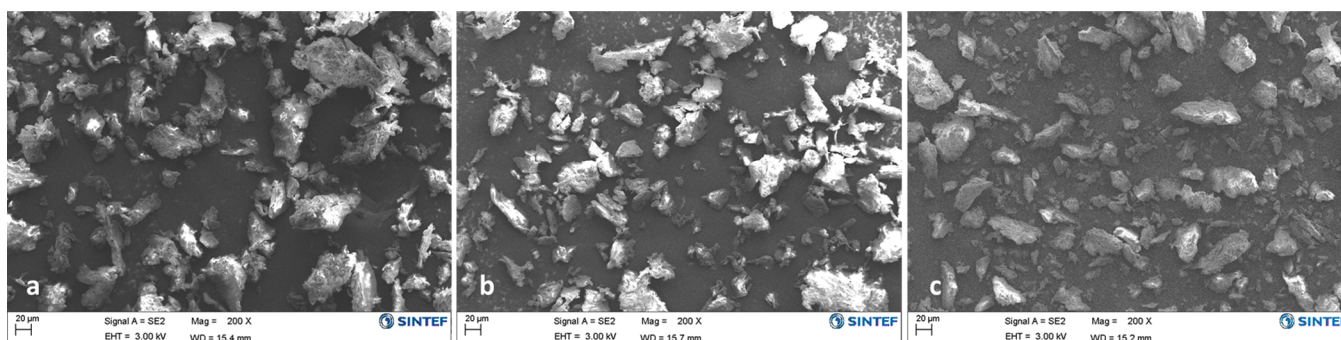


Figure 10. SEM images of the ground particles ($d < 0.063$ mm) from bark pellets: (a) untreated, (b) torrefied at 225 °C for 60 min, and (c) torrefied at 275 °C for 60 min.

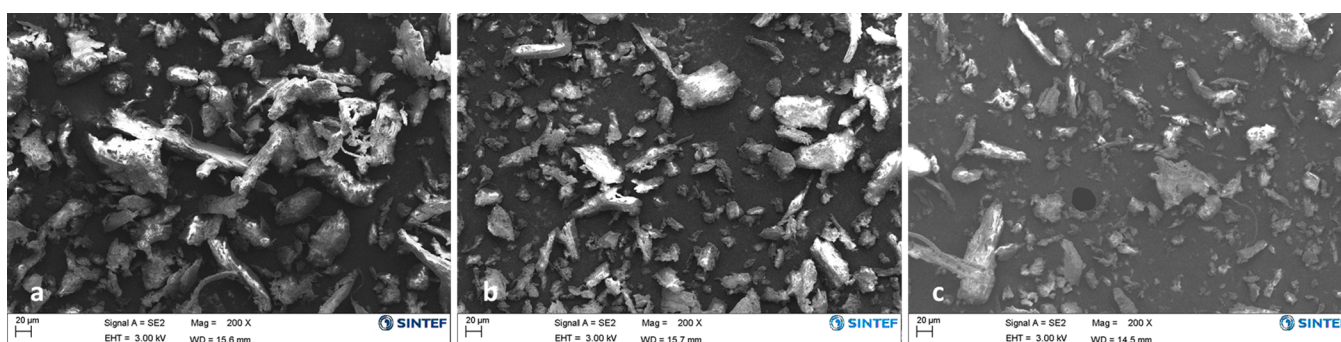


Figure 11. SEM images of the ground particles ($d < 0.063$ mm) from forest residue pellets: (a) untreated, (b) torrefied at 225 °C for 60 min, and (c) torrefied at 275 °C for 60 min.

electron microscope. Grindability test results showed a considerable change of the weight fraction of particles in the range of $0.3 < d < 0.5$ mm and $d < 0.063$ mm for the studied pellets upon torrefaction treatment. The analysis on particles with a size smaller than 0.063 mm is especially interesting, which provides supplementary information to grindability testing and is an indication of the flowing and conversion behavior of them.

Figures 6–8 show SEM images of ground untreated and torrefied wood, bark, and forest residue pellets in the size range of $0.3 < d < 0.5$ mm. Figure 6a clearly shows that the ground untreated wood pellet particles have a fibrous structure and some of them consist of highly fractured fiber bundles. The wood particles with such a fibrous structure are relatively resistant to breakage and are further ground down to particles with larger length/diameter ratios, as reported in other studies.^{38,39} However, the particles with loose and fibrous structures are barely found in panels b and c of Figure 6, which is partially related to decomposition and depolymerization of hemicellulose and cellulose during torrefaction of the wood pellets. In comparison to ground wood pellets, the ground bark and forest residue pellet particles are more compact and have an intact structure with low length/diameter ratios. Figures 9–11 show SEM images of particles ($d < 0.063$ mm) from ground untreated and torrefied wood, bark, and forest residue pellets. It is clear to see that particles shown in Figure 9a have large length/diameter ratios. Particles with a similar shape can be seen in Figure 9b, which are long in direction of the fiber axis but more elongated and splintery. However, for the particles from wood pellets after torrefaction at 275 °C for 60 min, they are rounder, with much smaller length/diameter ratios. Particles with such shapes and sizes might pass a sieve easier in comparison to the long fibrous particles from ground

untreated wood pellets.^{40,41} It can partially explain the evident increase of fine particles obtained from sieving tests, as shown in Figure 3. Figure 9 shows that particles from ground bark pellets are significantly different from those from ground wood pellets, which have low elongation ratios and are more powder-like.⁴² With the increase of torrefaction severity, more particles with a smaller size and more uniform shape are obtained. Different strength properties of wood and bark can partially explain different shapes and sizes of particles shown in Figures 8 and 9. The wood has very anisotropic strength properties, which results in wood breaking more easily in the direction of the fiber axis compared to across it.³⁸ Therefore, even after pelletization and grinding, a certain amount of wood particles still retained the fibrous structure with high length/diameter ratios. In comparison, the bark is much less anisotropic, and the primary breakage mechanism of bark is related to intercellular fracture. Therefore, less energy is required for grinding raw and torrefied bark pellets with the production of more smaller particles. Figure 10 shows that ground forest residue pellets contain both fibrous, flat-shaped particles and bulky particles, which might be from different parts of top and branches.

4. CONCLUSION

Torrefaction subsequent to pelletization is a promising strategy to improve the properties of wood, bark, and forest residue pellets. With increasing torrefaction severity, mass and energy yields decrease for all studied pellets, while HHV and energy density increased. Torrefied pellets have high hydrophobicity and remained intact, even after immersion in water for 72 h. The grindability of the torrefied pellets were significantly improved in comparison to that of untreated pellets. For the

wood and forest residue pellets torrefied at 225 °C, only approximately half of the grinding energy was needed in comparison to grinding the dried untreated pellets. Sieving of ground pellets showed that the fraction of fine particles ($d < 0.1$ mm) increased evidently, which is accompanied by a reduction of particles with large sizes (0.2 mm $< d$). Upon torrefaction treatment, the durability and strength of the studied wood, bark, and forest residue pellets were decreased, which correlated well with the improvement of grindability of them. The SEM analyses revealed considerable changes of morphological structures of the studied pellets before and after torrefaction. The particles contained in ground wood pellets have a fibrous structure and large length/diameter ratios. On the other hand, the ground bark pellets contain more round particles with an intact structure. SEM analyses showed that particles from ground torrefied pellets are more uniform in terms of size and shape.

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Notes

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