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Research article

Characterization of micronized wood and energy-size relationship in wood comminution

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ABSTRACT

Mechanical milling demonstrates potential in the pretreatment arena to valorize lignocellulosic biomass because it eliminates chemicals and simplifies processing. The development of physical properties and energy consumption for generating micronized particles are key factors affecting potential use. This study investigated the effect of input moisture content, and feedstock particle size on developing physical characteristics of micronized particles and the resultant specific energy consumption in the milling process. Wood particles with different sizes were effectively comminuted to <20-μm within several minutes using a vibratory ring and puck mill. Moisture content was found to be a key factor influencing the development of particle morphology and crystallinity. Lower moisture content resulted in much rounder particles with lower crystallinity, while higher moisture content resulted in the micronized particles with larger aspect ratio and crystallinity. Crystallinity index and median aspect ratio of the micronized particles were linearly correlated. The particle size change during milling was highly correlated to the specific energy consumption of milling through the Rittinger's model ($0.91 < R^2 < 0.96$). Input moisture content and feed size were found to affect the energy intensity of grinding woody biomass. The Rittinger's constant was a good indicator of the material performance in this area. The results will provide a guidance for preferred milling conditions as well as designing scalable micronizing mills.

1. Introduction

The development of sustainable biofuels from woody biomass is becoming increasingly important for diversifying our transportation energy portfolio and mitigating the global greenhouse gas emission [1]. Both biochemical and thermochemical biorefinery processes have encountered challenges for commercial implementation, including the robust biomass recalcitrance and the need for an integrated feedstock supply. Depot-based technologies may be useful for transforming bulky biomass into an interchangeable feedstock for multiple applications; e.g. pretreated wood, or fermentable sugars [2]. Such an approach can potentially reduce risk and logistics costs of feedstock supply for centralized downstream processes. An appropriate pretreatment plays a key role for such depots by facilitating the potential distributed production of cellulosic sugars.

Mechanical deconstruction of biomass offers potential for streamlining processes and enabling supply chains that utilize existing forest products facilities, provide flexibility to operate at various scales, eliminate chemical input, and reduce or eliminating waste water treatment requirements [3,4]. Mechanical milling is a common method of altering physical properties of substrates (e.g., particle size, aspect ratio and cellulose crystallinity, etc.) and can significantly facilitate either biochemical or thermochemical conversions. Reduced particle size is beneficial to biomass conversion by improving particle reactivity and heat/mass transfer efficiency [3,5]. The aspect ratio of particles is a crucial factor for influencing biomass pyrolysis efficiency, though the relationship between particle shape and biochemical conversion efficiency remains elusive [6–8]. However, it is known that the cellulose crystallinity in lignocellulosic biomass influences the rate and extent of enzymatic digestion [3,9]. The physical properties of milled biomass can be controlled by milling conditions; e.g. the moisture content of feed material, and feedstock particle size. Kobayashi et al. [10] reported that wood powder pulverized with a vibratory mill at 3% moisture content had a reduced crystallinity of 14.3%, while wood powder ground from 11% moisture content resulted in a crystallinity of 37.7%. Even though the exact fracture mechanism of particles in the milling chamber is unknown, Gil et

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al. [11] have shown that the feedstock particle size influenced the particle morphology of final products in hammer milling. According to Gil et al., single particle breakage in an impact milling is related to the particle volume and contact area upon impact; both of which are determined by the particle size. Thus, a larger particle size increases the probability of structure fragmentation, leading to rapid particle size reduction. Despite a significant amount of information obtained on the effect of milling conditions on the physical properties of coarsely milled particles from several types of mills [4,12], a systematic understanding of the effect of milling conditions on producing fine particle fractions (i.e., partial size below 100 μm) and their ensuing physical properties is lacking. This finely milled biomass with an average particle size below 100 μm is termed here as micronized biomass. These micronized particles exhibiting a significant level of cell wall disintegration, have shown to be susceptible to enzymatic hydrolysis [13–15]. Such micronized biomass was also found to be easier thermochemical conversion (e.g. pyrolysis) with lower degradation temperature and lower thermal degradation activation energies, facilitating production of high-value furfurals, mainly due to their high reactivity [16]. Additionally, micronized wood is highly preferable for industrial application of low-cost natural fiber-based thermoplastic composites, as it provides high surface area for better reinforcement [17]. Conventional milling technology like hammer milling is incapable of providing sufficient size reduction to produce a micronized particle [18]. In contrast, micronized wood is commonly prepared using vibratory milling [19,20].

It is apparent that a considerable amount of energy is consumed during size reduction of lignocellulosic biomass [21,22]. A number of previous studies have attempted to establish the relationship between energy consumption and particle size reduction during biomass milling. For example, a power-law relation was proposed to correlate energy consumption with product particle size (e.g., screen size) or the biomass comminution ratios [23,24]. Inspired by the milling theories proposed to delineate the specific relationship for milling ores, Temmerman et al. characterized the energy-size relationship with the Rittinger's model for hammer milling wood chip and pellet. Among these studies, the Rittinger's model was recognized as the best approach to describe fine milling performance, as well as a good indicator for assessing a given comminution system [25–27]. However, predicting the energy consumption for micronized wood by mechanical milling with different operational conditions is still lacking. Therefore, modeling the energy consumption of fine wood milling with the size characteristics will be the first step to optimize the milling process for potential industrial application.

The main goal of this study is to delineate the physical property development of micronized particles under various milling conditions and correlate size reduction with energy consumption of the process. The experiments focus on exploring effects of moisture content and feed size for the starting material on changes in particle morphology, cellulose crystallinity, and specific energy consumption of the fine milling process. The knowledge gained in this study will aid in developing potential approaches and strategies for producing a micronized biomass substrate in an economical manner with mechanical pretreatment.

2. Materials and methods

2.1. Material

Clean, Douglas-fir (*Pseudotsuga menziesii*) wood chips were obtained locally (Vaagen Brothers Lumber Inc., Colville, WA). The as-received chips were passed through a vibrating screen with a 25.4-mm aperture and retained on a 4.75-mm screen, producing a chip with a geometric mean diameter of 10.36-mm. The screened wood chips were processed into particles by a hammer mill fitted with a 11.11-mm, 6.35-mm, and 3.18-mm, screen, respectively. These screened feedstock samples were labeled as Pxx (pass) with xx being the screen size (i.e., P25, P11, P6, and P3, respectively). The processed feedstock was subsequently stored in a temperature-and humidity-conditioned room with an equilibrium moisture content (EMC) of 15% and subsequently conditioned to different target EMCs values. After final conditioning, all material was stored in sealed plastic bags and moisture content was validated using gravimetric methods according to standard protocol [28].

2.2. Mechanical milling process

Fine milling experiments were carried out with a high-energy vibratory ring and puck mill (Standard Ring milling, motor power 1.1-kw, Rocklab Pty Ltd., New Zealand). The milling chamber had an inner diameter of 128-mm and height of 43-mm. The grinding media were a ring (inner diameter 78-mm, outside diameter, 100-mm, height 41-mm) and a puck (diameter 52-mm and height 41-mm). Both milling chamber and grinding media were made of tungsten carbide.

The milling experiments were designed to investigate different milling variables; these include milling time, initial moisture content, and size of feedstock. The resulting micronized wood powder was then characterized for particle size distribution, aspect ratio, and crystallinity. Milling times were varied from 2 min to 12 min with 2-minute interval. Four initial moisture contents (5, 10, 15, and 30% oven dried base) were tested, using P3 particles with charge quantities of 10-grams per grinding batch. For investigating effect of the feed size on milling performance, four different initial size levels (P25, P11, P6 and P3) were tested, using conditioned moisture content of 10% samples with charge quantities of 10-gram per milling batch.

2.3. Measurement of specific energy consumption

The milling energy consumption of the ring and puck mill was measured with a Fluke 1735 power logger (Fluke, USA). The active power, active energy, power factor, frequency, and time were acquired by computer. The specific grinding energy consumption was calculated using the following relation:

$$E_p = \int_0^t (P_t - P_0) d_t / m = \int_0^t \Delta P_t d_t / m$$

where: E_p is the specific net energy consumption (kJ/kg); P_t is the power consumed at time t; P_0 is the average power consumption under idle condition measured from an empty mill; and m is the mass charge in kg of wood to be pulverized.

2.4. Characterization of wood powder

2.4.1. Preparation of wood powder for analyses

The milled wood samples were prepared for particle size distribution and aspect ratio analyses as follows. Each sample was suspended in distilled water to achieve a 0.5% solids mass concentration. For the aspect ratio measurement, a diluted powder suspension was dispersed by adding 1-mL of a dispersant solution (0.1% w/w sodium dodecyl sulfonate) per 100-mg wood [29]. The suspended samples were stirred for 1 h with a magnetic stir rod, followed by sonication with 20% amplitude (Branson, Switzerland) for 2 min to ensure complete disintegration of particle agglomeration. The same procedure was following for measuring particle size distribution, except dispersant was not added.

2.4.2. Particle size distribution

The volumetric particle size distribution of the wood powders was analyzed using a laser scattering particle size analyzer (Mastersizer 3000 with Hydro LV wet sample dispersion, Malvern instrument, UK). The median size was used to represent the particles size for analysis. The dimensionless parameter Δ_d is used to represent the breadth of

the particle size distribution, where:

$$\Delta_d = \frac{d_{90} - d_{10}}{10\mu m}$$

and d_{90} and d_{10} are particle sizes corresponding to the 90% and 10% percentiles as assessed from the cumulative size distribution, respectively.

2.4.3. Aspect ratio analysis

The solid suspension of samples prepared as described above, was distributed on a silica wafer mounted on a metal stab. This process involved using a pipette to form a droplet with a volume of ca. 200-μL. After allowing the water to evaporate at ambient conditions, samples were sputtered with gold to prepare examination in a scanning electron microscopy (SEM). The measurement of aspect ratio was obtained from the analysis of SEM images using ImageJ (National Institutes of Health, USA) following the procedures described elsewhere [30,31]. For each sample, about 20 SEM images containing around 30,000 particles were analyzed. The aspect ratio of each particle was calculated as the ratio of the major and minor axis of the fit ellipse for the selected particle. The minimum aspect ratio value of 1 was restricted and describes a perfect and filled circle. The number based cumulative aspect ratio distribution was used to obtain the median aspect ratio AR_{50}, which was the value of 50% cumulative aspect ratio distribution. The width of aspect ratio distribution Δ_{AR} was defined as the difference between the aspect ratios corresponding to 90% and 10% values in the cumulative distribution.

2.5. Cellulose crystallinity

X-ray diffractograms of the milled wood particles were obtained using a powder x-ray diffractometer (Rigaku, Miniflex 600, Japan) with a Cu Kα ($\lambda = 0.154$ nm) radiation source generated at 40-kV and 15-mA. The instrument scanning range 2θ was from 10 to 40°. The relative degree of cellulose crystallinity, in terms of crystallinity index (CrI), was estimated using equation as described by Segal [32]:

$$CrI\ (\%) = [(I_{002} - I_{am})/I_{002} \quad 100$$

where I_{002} is the intensity of main peak, and is the intensity due to amorphous portion evaluated as the minimum intensity between the main and secondary peaks.

2.6. Scanning electron microscopy (SEM)

The morphology of prepared samples as described above was analyzed by scanning electron microscopy. Images of samples were acquired typically at 20-kV accelerating voltage using FEI Quanta 200F, field emission gun with high vacuum ETD detectors (FEI Company, Hillsboro, Oregon, USA). Prior to imaging, the suspended wet sample was air dried and sputtered with gold for good conductivity.

3. Results and discussion

3.1. Effect of milling conditions on characteristics of particle morphology

Mechanically fragmenting the lignocellulosic biomass into the micronized size range has been recognized as an essential step to enhance the substrate digestibility of mechanically pretreated substrates by others [13,33]. In this study, effectively micronized softwood was achieved using ring and puck mill under various milling conditions. The change in median particle size d_{50} as a function of milling time for feedstock with various initial moisture contents is presented in Fig. 1A. At the early stage of milling (e.g., at the 2-minute point), an elevated moisture content facilitates more rapid particle size reduction (Fig. 1A), which is likely a result of the higher specific energy input for milling higher moisture content samples (e.g. MC30%). A previous study reported that a mechanical refining process with a higher energy input caused more severe breakage of wood fiber with shorter length and width [34]. Additionally, Karinkanta et al., demonstrated that smaller particles were obtained at conditions with higher impact energy input when Norway spruce was pulverized with an oscillatory ball mill [29]. We also observe that d_{50} approached around 20 μm at 6-minute milling time under all tested moisture content conditions, while the changes in d_{50} were slight for additional milling time to 12 min. These systematic changes in particle size with respect to milling time are in agreement with the literature reports, which claimed that particle size could not infinitely decrease by prolonging milling time after attaining a critical size [22]. The width of particle size distribution Δ_d is increased as a function of d_{50} for samples with different initial moisture contents (Fig. 1B). The strong linear decrease in Δ_d with respect to d_{50} suggests that the mechanical milling resulted in effective particle size reduction of micronized wood, yielding more homogeneous particles.

The median particle size d_{50} of micronized wood produced from various feedstock sizes is depicted in Fig. 1C. For different feed size samples, we successfully comminuted the particles from the millimeter to micrometer range in 12-min or less, using a highly versatile and energy-efficient ring and puck milling system. Micronizing woody biomass from a chip size (i.e., P25 in Fig. 1C) can thus be effectively achieved in relatively a short milling time compared to the ball milling times of several hours or days reported by others [14,35,36]. At an early stage of the milling process, we notice that feedstock with various initial sizes were milled to similar size around 100-μm. This behavior was likely a result of a higher contact area upon impact for larger-sized samples during milling process. A previous study on impact milling of lignocellulosic biomass indicated that larger particles experienced a higher breakage probability, since the internal failure probability increases with volume as well as with the particle's contact area upon impact [11]. We also observe that a rapid decrease of particles reaching ca. 20 μm for various initial sizes at 6-minute milling time point, followed by a slight change of particle size after further milling to 12 min. The similar tendency of particle size evolution from different input sizes suggests that initial size has a negligible effect on development of micronized wood particle size, even though bigger size sample breaks up faster at early stage of milling process. The relationship between the width Δ_d and mean d_{50} of the particle size distribution for samples with different feed sizes is presented in Fig. 1D. The results also demonstrate effective size reduction and generation of homogeneous particles where the Δ_d decreases linearly with d_{50} (with high coefficient $R^2 = 0.98$).

The influence of milling conditions on changes to the shape of micronized particles, as represented by median aspect ratio AR_{50} and width distribution of aspect ratio Δ_{AR} are shown in Fig. 2. The initial moisture content differentiated the shape development of micronized wood (Fig. 2A). We observe that AR_{50} decreased gradually with the increase of milling time for those samples with initial moisture content of 5%, 10% and 15%, whereas less change in aspect ratio for samples with initial moisture content of 30% is noticed. These observations indicate that lower moisture content contributes to much rounder particles, and higher moisture content results in particles with higher aspect ratio. The linear decrease of aspect ratio distribution width Δ_{AR} with respect to mean AR_{50} indicates that micronized particles possessed much more uniform shape after mechanical milling. The median aspect ratio AR_{50} of micronized wood milled from various feedstock sizes is depicted in Fig. 1C and D. We notice that the feed size has negligible influence on AR_{50} to milling time relation for micronized wood produced by this type of milling (Fig. 2C). All the samples evolved to similar particle shape during milling process, regardless of their initial size. In addition, the linear relation between the breadth Δ_{AR} and mean AR_{50} of the particle aspect ratio distribution also suggests homogeneous particle shape of micronized wood regardless of the feed sizes (Fig. 2D).

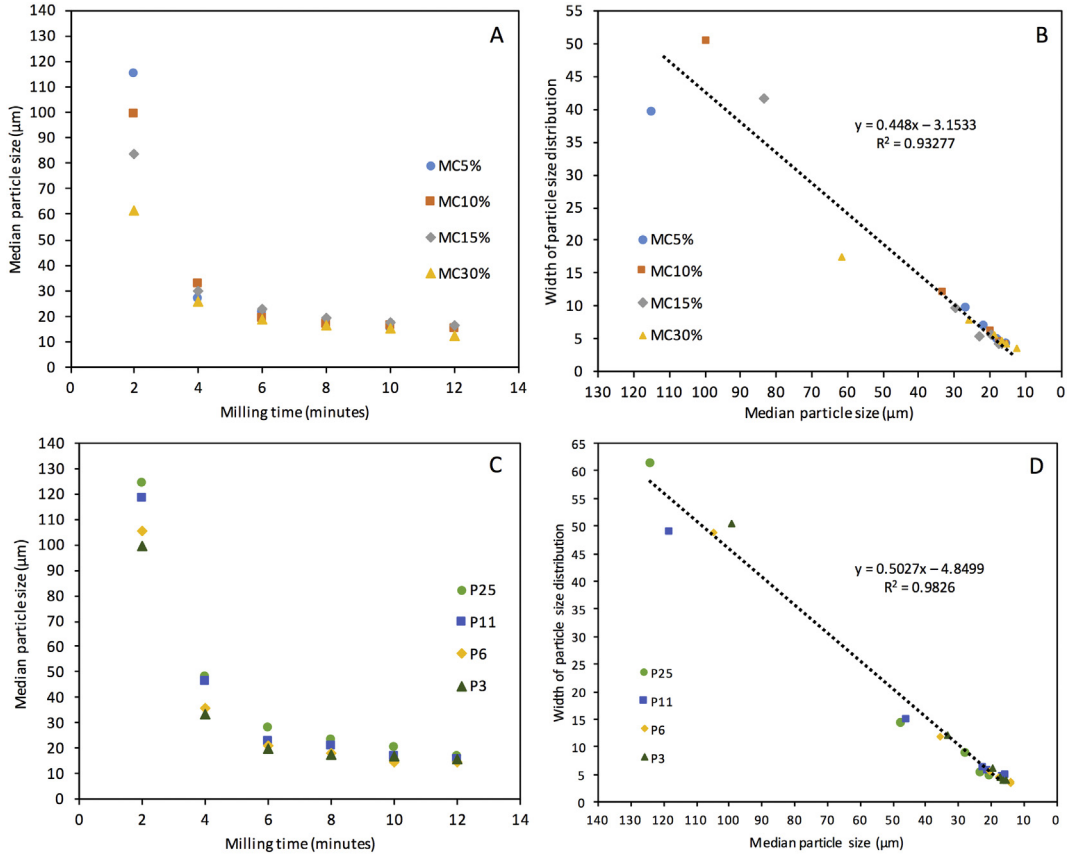

Fig. 1. Effect of milling conditions on the median particle size and width of particle size distribution. (A) median particle size changes of samples with different moisture contents; (B) width of particle size distribution of samples with different moisture contents; (C) median particle size changes of samples with different feed sizes; (D) width of particle size distribution of samples with different feed sizes.

3.2. Effect of milling conditions on cellulose crystallinity

Besides observing the morphology changes of micronized wood during the milling process, changes in cellulose crystallinity were also characterized. The crystallinity index (CrI) changes of micronized wood produced from various initial moisture contents are delineated in Fig. 3A. Overall, CrI decreases significantly with the increase of milling time for those tested samples, suggesting effective disruption of crystalline structure during ring and puck milling process. This is in agreement with other reports on crystallinity reduction of lignocellulosic biomass subjected to mechanical actions (e.g., impacting, and shearing, etc.) with various milling techniques [3,4]. More precisely, the CrI decreased rapidly to around 10% for micronized wood produced with relative low moisture contents (e.g., MC5% and MC10%), suggesting significant amorphization of crystalline cellulose. Nevertheless, the CrI of micronized samples that were milled with a relatively higher initial moisture content (e.g., MC30%) only decreased to 40% even after the longest milling time of 12-min, suggesting less changes in crystalline structure during milling process. These phenomena indicate that the initial moisture content significantly influenced the development of cellulose crystallinity. Previous research on milling pretreatment of sugarcane bagasse and wheat straw using dry ball and wet disk milling indicated that dry milling resulted in significant amorphozation of this herbaceous biomass feedstock, whereas wet milling only slightly reduced the crystallinity [37]. Hoeger et al., also reported that mechanical fibrillation of softwood pulping fibers in wet state with stone grinding contributed to only 15–25% crystallinity reduction with fibrillation time of 6–12 h [33]. The CrI changes of micronized wood produced from various initial sizes are depicted in Fig. 3B. There is gradual decrease of CrI to around 10% with respect to milling time for samples milled from different feed sizes. The changes in CrI show similar tendency for different feed size samples, suggesting negligible effect of feed size on crystallinity development of micronized wood. These results are consistent with previously reported observations that various woody and herbaceous biomass feedstock with different initial sizes were effectively milled to similar crystallinity using a tandem-ring mill [20].

3.3. Relations of structural characteristics of micronized wood

Mechanical milling conditions can generate micronized wood with distinctive particle morphology and crystallinity as discussed above. It is also interesting and essential to investigate the relationships among these structural characteristics of micronized wood for better understanding of sequential particle evolution during milling process. Thus, the changes of median aspect ratio AR_{50} as a function of median size d_{50} for those obtained samples under various milling conditions are shown in Fig. 4. It is observed that AR_{50} decreased slightly with the decline of d_{50} for samples with initial moisture content of MC30%, whereas those from other samples showed drastic decrease. Such phenomena may suggest that the development of micronized wood is mainly related to fracture modes that are determined by initial moisture contents during milling process. It is interpreted that the fiber fragmentation mode is responsible for the development of micronized wood with high moisture content input (e.g., MC30%). That means the fiber fracture parallel to the fiber length occurred more frequently than it did perpendicular to the wood grain, with higher initial wood moisture content (i.e., 30%). This fracture behavior resulted in a decrease in particle size with a negligible change in aspect ratio. As elucidated by previous research on mechanical pulping, refining along the fiber length resulted in fibers peeling rather than fracturing across the fiber length [38,39]. During the milling process, the lower moisture content samples

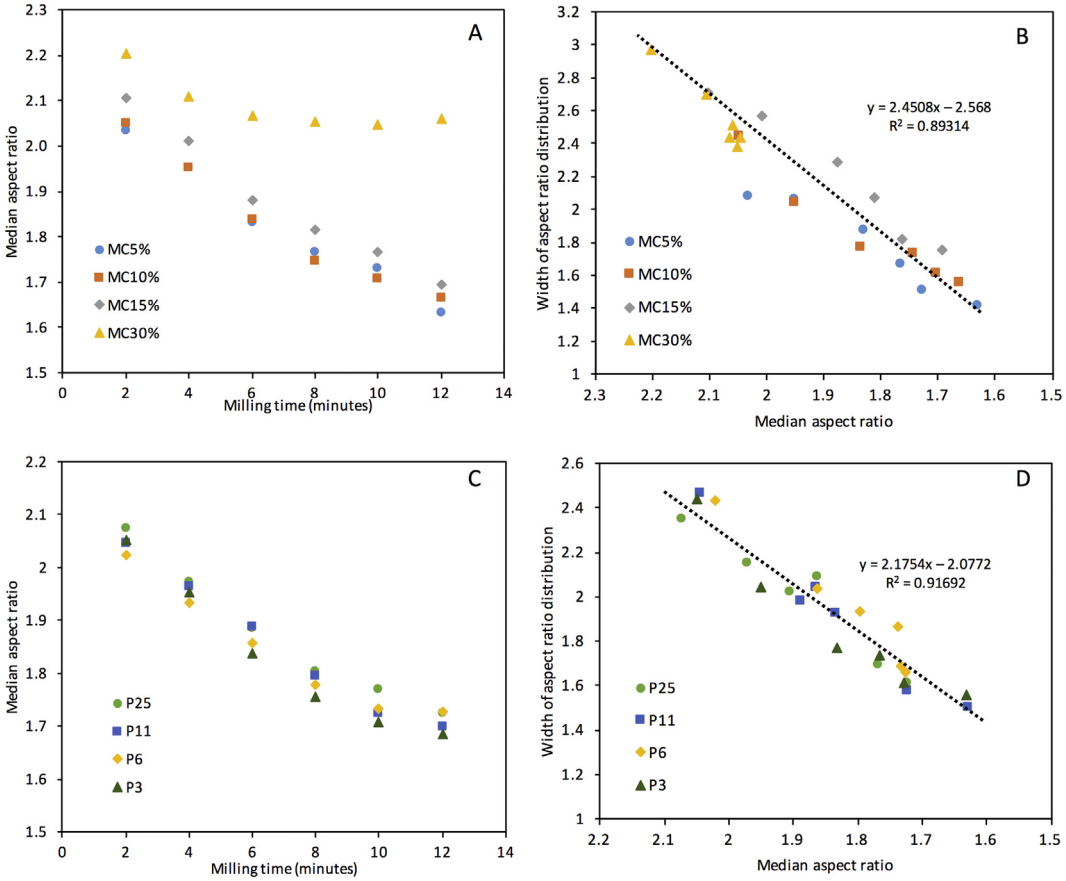

Fig. 2. Effect of milling conditions on the median aspect ratio and the width of particle aspect ratio distribution. (A) median aspect ratio changes of samples with different moisture contents; (B) width of aspect ratio distribution of samples with different moisture contents; (C) median aspect ratio changes of samples with different feed sizes; (D) width of aspect ratio distribution of samples with different feed sizes;

irrespective of their sizes might undergo chipping and abrasion, leading to shorter fibers along with particle size reduction, collectively.

On the other hand, the strength of wood fibers perpendicular to the cellulose fibrils is significantly higher when wood is below the fiber saturation point; ca. 30%. This behavior results from increased hydrogen bonding between cellulose chains that would otherwise be bridged by water molecules [40]. With relatively lower moisture contents, wood fibers possessed higher stiffness. Fracture would thus propagate in the direction perpendicular to the fiber length more frequently at low moisture content, reducing aspect ratio along with particle size. In addition, the increased probability of fracture perpendicular to the fiber length was also coincident with the disruption of crystalline structure, resulting in lower cellulose crystallinity for lower moisture content samples.

The difference of particle aspect ratios for milled wood at various moisture contents is also visually evident. SEM images of micronized particles produced at various moisture contents and a milling time of 12-min are shown in Fig. 5. Fibrous particles were obtained from MC30% sample, while relatively low moisture content samples resulted in globular and spherical particles.

The relations between cellulose crystallinity and particle morphology are depicted in Fig. 6. The CrI decreases significantly with the decrease of median particle size d_{50} for all milling conditions (Fig. 2.6A). Nevertheless, there is less change of CrI for samples milled with

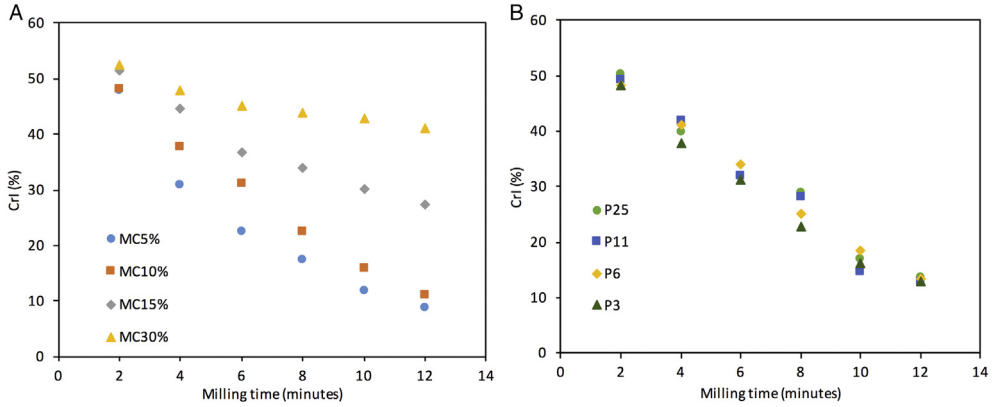

Fig. 3. Effect of changes in initial moisture content (A) and feed size (B) on development of crystallinity index (CrI) of micronized wood as a function of milling time.

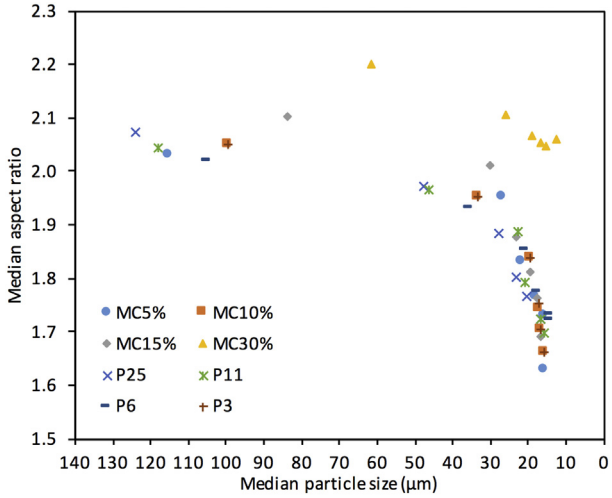

Fig. 4. Relation of median aspect ratio and median particle size of micronized wood from various milling conditions.

relatively higher moisture content (e.g., MC30%) compared to all others. This decreased change in crystallinity is noted despite the fact that the resulting particles were smaller than those produced at the lower moisture contents. This observation is in agreement with previous reports, which claimed that the moisture content of lignocellulosic substrates was a primary factor affecting the decrease in crystallinity during ball milling [10,41].

Moisture content is an important factor influencing the glass transition of amorphous polymers in wood cell wall matrix, and the glass transition temperature (Tg) shifts to lower temperature range when moisture content increases [42,43]. With the moisture content around 30%, hemicellulose has a Tg around −22 °C, thus existing in its rubbery state with greatly reduced stiffness at ambient milling condition. The Tg of hemicellulose is about 30 °C at approximately 10% moisture content [42]. That means the amorphous polymers were in their glass state during ambient milling of low moisture content samples. Wood fiber structure is also known to have microfibril bundles embedded within the amorphous polymers matrix [44]. Thus, during the milling process, the fracture of wood fibers occurring in higher moisture content samples would primarily include the breakage of rubbery state matrix and splitting apart of microfibril bundles without causing extensive disorder within the crystalline structure of cellulose. However, for the lower moisture content samples, the impact milling disrupted the brittle cell wall matrix and crystalline cellulose skeleton. This led to a collective decrease of particle size and cellulose crystallinity, which is in agreement with the previously discussed evolution of particle aspect ratio. Fig. 6B illustrates the strong linear relationship between AR_{50} and CrI, regardless of the moisture content and initial size. The quality of the linear fit is characterized by high coefficients ($R^2 = 0.90$). Here, larger aspect ratio values correspond to higher CrI.

3.4. Analysis of energy requirements for size reduction

Besides the physical properties of micronized particles, specific energy consumption for producing such particles is another important factor for evaluating fine milling performance of woody biomass. Rittinger's model is an appropriate model to describe the fine milling performance, where the new surface area generated per unit mass of the particle is directly proportional to the specific energy consumption for size reduction [25–27]. The specific surface area is inversely proportional to the particle size. Therefore, the Rittinger's model can be expressed as follows: $E = C(\frac{1}{d_p} - \frac{1}{d_f})$, where C is a constant characteristic of the material, d_p and d_f are the characteristic particle size of milled product and feed [45]. The advantage of applying this model lies in characterizing the grinding performance of woody biomass with one parameter, which can be determined from model constant C. The collection of limited parameters such as specific energy consumption and particle size will benefit to scale-up processing of fine milling of woody biomass.

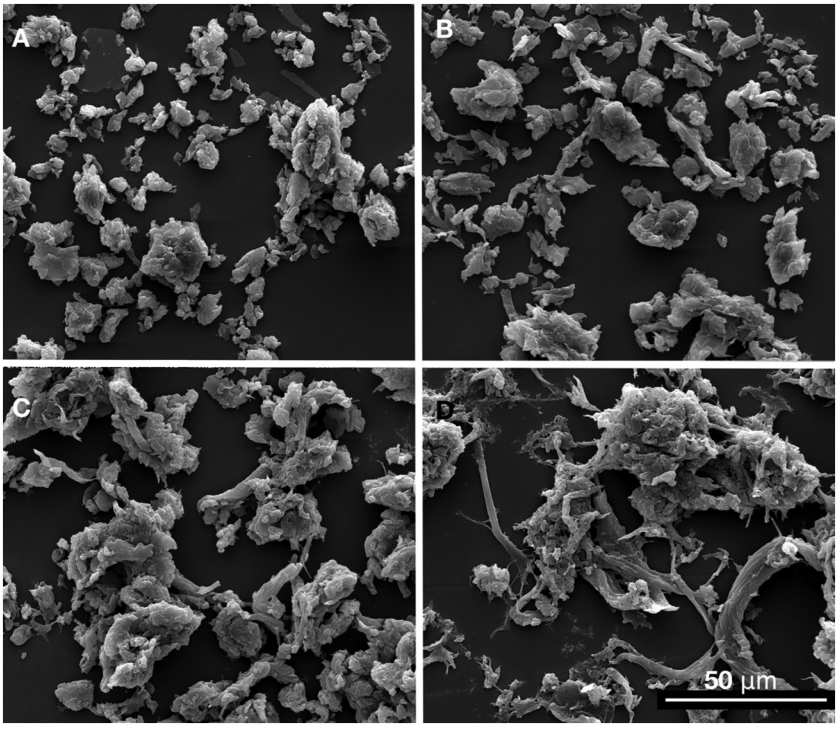

Fig. 5. Morphology difference of micronized wood with different initial moisture contents (A: MC5%, B: MC10%, C: MC15%, D: MC30%) during milling process; feed size: P3; loading: 10 g; milling time: 12 min.

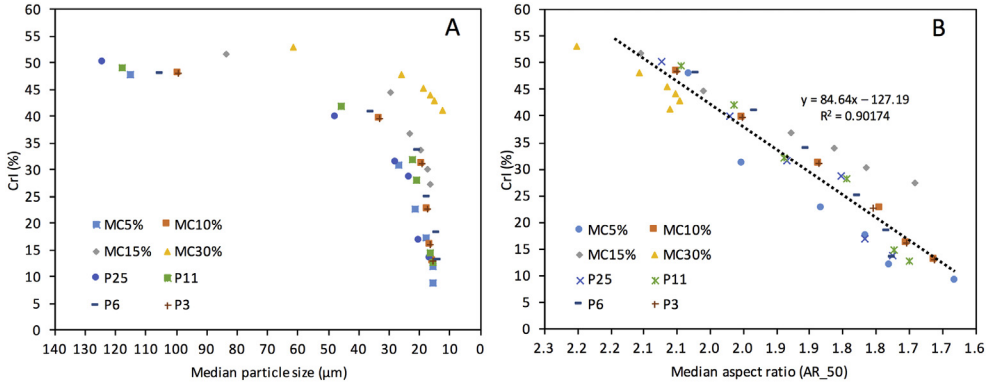

Fig. 6. Development of crystallinity index (CrI) as a function of (A) median particle size and (B) median aspect ratio of micronized wood from various milling conditions.

Linear relationships between specific energy consumption and particle size reduction parameter in Rittinger's model ($1/d_p - 1/d_f$) for samples with different initial moisture contents are shown in Fig. 7. The quality of the linear fit is characterized by high coefficients ($0.91 < R^2 < 0.95$). In previous study, Karinkanta et al. also reported linear relationship between the inverse value of d_{50} of finely ground wood powder and total available impact energy with oscillatory ball mill in accordance to Rittinger's model [29]. The model constant C can be a good indicator for grindability of woody biomass, and for evaluating the energy efficiency of milling process when similar materials are used as a probe [46]. As discussed above, moisture content was the key factor influencing the development of morphology and crystallinity of micronized wood, and to a large extent, the specific energy consumption. Increasing the moisture content induced increase of the specific energy consumption during milling process, as indicated by model constant values. This phenomenon was in agreement with previous research on energy consumption for milling various wood species with different moisture contents conducted using laboratory-scale hammer mill [46]. As indicated above, moisture content greatly affects the toughness of woody biomass. At low initial moisture content (e.g., MC 5%), the brittle polymer components fractured easily with low energy consumption. Increasing the moisture content resulted in the amorphous polymers becoming tougher due to the moisture acting as a plasticizer, thus requiring more energy consumption on fatigue to cause rupturing.

The specific energy consumption as a function of size reduction parameter ($1/d_p - 1/d_f$) in Rittinger's model for various initial size samples is shown in Fig. 8. The experimental results are also fitted well to Rittinger's model. The linear relations are identified with high coefficients ($0.91 < R^2 < 0.96$). The larger the feed size, the more energy was required for fine milling. Although ring and puck mill is capable of comminuting chip-sized feedstock to micrometer range within minutes, it is not economically viable from the energy perspective. Coarse milling of Douglas-fir with different fitted screen sizes as used in the current study through pilot-scale hammer mill had been carried out in our lab, and the results indicated that the energy consumption for milling wood chips was in the range of 59–141 kJ/kg depending on specific fitted screen size. Several researchers also reported that fine milling of woody biomass was much more energy consuming than coarse milling [4,21,46,47]. Therefore, multi-step comminution strategy with combination of coarse and fine milling seems much more economical than directly fine milling of large-sized feedstock. However, the multi-step comminution will inevitably increase the capital investments.

The difference in energy consumption for milling samples with various properties suggests that minimizing the grindability of woody biomass is necessary when using ring and puck mill for fine milling. Therefore, it is reasonable to standardize the milling variables using a reference standard (e.g. chemical composition, moisture content and size) to obtain the grindability of lab-scale test as reported in mineral and biomass industry [48–50]. In addition, our results show very competitive potential for industrial scale production of fine wood powders with such comminution technology. Disk milling predominately used in paper pulping area for wood fiber disintegration (i.e., size reduction) generally requires energy consumption of over 2 MJ/kg wood depending on wood species and the size reduction ratios [51]. Kobayashi et al. [10] have reported that the energy requirement for pulverizing

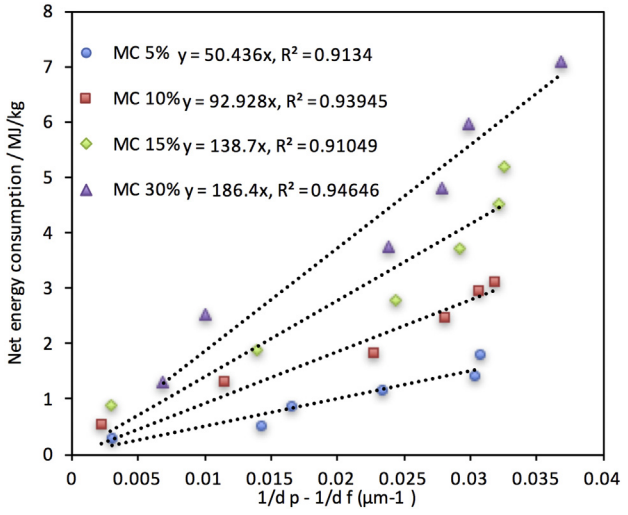

Fig. 7. Effect of moisture content on specific net energy consumption as a function of particle size reduction ($1/d_p - 1/d_f$).

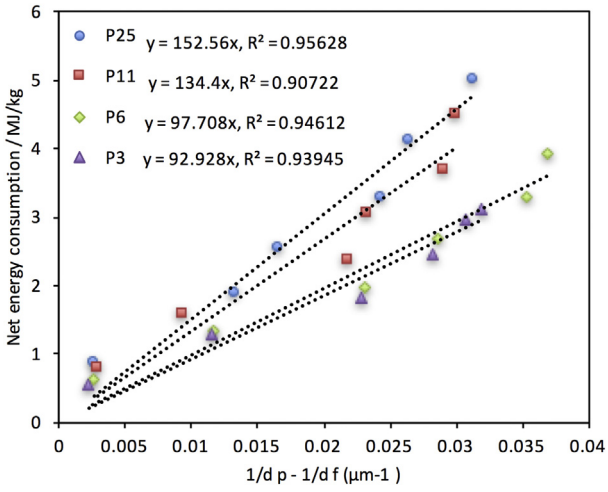

Fig. 8. Effect of feed size on specific net energy consumption as a function of particle size reduction ($1/d_p - 1/d_f$).

22 mm-size Norway spruce wood chips into powders with a size of 150 μm is 2.95 MJ/kg (including conveyer, compressor and powder sieve) using a continuous vibration mill. When dry spruce sawdust with size range around 1 mm was comminuted with an oscillatory ball mill, Karinkanta et al. [29] found that the energy consumption of 0.36 MJ/kg was needed to get wood particles with size around 100 μm and 1.4 MJ/kg was consumed to get wood powder with size around 20 μm. In this study, pulverization of Douglas-fir chips with moisture content of 5% and initial size about 1 mm (i.e., feed sample P3) down to size around 100 μm is accomplished with energy consumption of 0.28 MJ/kg, while size reduction to 20 μm is accomplished with energy consumption of 0.86 MJ/kg. Therefore, the energy consumption for wood particles pulverization provides a practical target for optimization of comminuting woody biomass into micron size range for industrial application using ring and puck mill.

4. Conclusions

We effectively micronized softwood Douglas-fir using a vibratory ring and puck mill within several minutes. The effect of substrate properties (i.e., moisture content and initial size) on development of physical properties of micronized particles and specific energy consumption for producing such particles were investigated. Results indicated that moisture content was the most important factor influencing milling behavior in terms of particle morphology and cellulose crystallinity. The lower moisture content samples resulted in much rounder particles with relatively smaller crystallinity than their higher moisture counterparts. The feed size had negligible influence on development of particle physical properties, but only affected the specific energy consumption. A multi-step milling strategy is recommended for a more economical approach for fine milling of woody biomass. The crystallinity and aspect ratio of particles underwent similar development tendency. There was a linear relationship between crystallinity index and median aspect ratio of milled particles.

Rittinger's model was ideal for describing the relationship between specific energy consumption, and particle size changes. The constant in this model was a good indicator of grindability of woody biomass with different properties. It is also recommended that standardizing milling test and parameters could benefit to evaluating the grinding performance of fibrous materials on a lab-scale, as such information would be useful for designing scale-up milling facilities of woody and herbaceous biomass feedstock.

List of symbols

d_{50}	Median particle size (μm)
Δ_d	The breadth of the particle size distribution (unitless)
AR_{50}	The value of 50% cumulative aspect ratio distribution (unitless)
Δ_{AR}	The difference between the aspect ratios corresponding to 90% and 10% values in the cumulative distribution
E_p	The specific net energy consumption (kJ/kg or MJ/kg)
t	The milling time (s)
P_t	The power consumed at time t (W)
P_0	The average power consumption under idle condition measured from an empty mill (W)
m	The mass charge in kg of wood (kg)

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