1 Use of tube flow fractionation in wood powder characterisation

2 **Pasi Karinkanta*, Ossi Laitinen**

3 Faculty of Technology, Fibre and Particle Engineering, University of Oulu, Finland

4 * corresponding author

5 Faculty of Technology, Fibre and Particle Engineering, P. O. Box 4300, FIN-90014 University of Oulu, Finland

6 E-mail: pasi.karinkanta@hotmail.com

8 **Abstract**

9 The size and shape of powdered wood particles are important properties when considering their use in wood burning or
10 the production of biofuels, biocomposites and biochemicals. Different measurement techniques can be used to measure
11 particle size, but unfortunately these do not provide information about the shape of the wood particles. In this study, a
12 fast and reliable tube flow fractionation method (analysing time 3min; particle size range 1µm-2000µm; analysed
13 particles per sample 30,000-200,000) is used to separate water-diluted wood powders which varies by size and shape.
14 30 different milled wood powder samples are analysed (average particle size from 20µm to 300µm and average aspect
15 ratio from 3 to 10). The major benefit in tube flow fractionation method is that the concealment of the finest particles in
16 CCD imaging is avoided by applying the imaging unit after the fractionation of wood particles. Wood powder can
17 easily be separated into different fractions by leading the water-diluted sample flow into different beakers for certain
18 time periods. The transmittance signal after tube flow fractionating yields information about the particle size, and
19 optical imaging provides information about the morphology of the particles. It was found in this study that rotor impact
20 milling to a finer size range produces small but elongated particles. Additionally there were only small differences in
21 the particle size distributions between jet milled and oscillatory ball milled samples, but it was noticed that oscillatory
22 ball milled powders have a significantly lower aspect ratio than jet milled samples.

23 **Keywords:** Aspect ratio, morphology, particle size, tube flow fractionation, wood, wood powder characterisation.

24 **1. Introduction**

25 Fine ground wood, that is, wood powders or flours, are sustainable raw materials that can be used in the production of
26 bioenergy, biocomposites and chemicals that originate from cellulose, lignin and hemicelluloses. Knowledge is already
27 available about the mechanical processing of wood into powdered forms [1–9] and their use in different applications
28 involving burning [10,11], composite production [12–15] and the preparation of biochemicals and biofuels [16–19]. It is
29 recognised that the size and shape of fine ground wood particles influence the chemical, enzymatic and thermochemical
30 conversion of wood, but they also influence the mechanical properties of wood-based composites.

31 Particle size measurement by sieving or laser diffraction offers a way to obtain knowledge about the particle size
32 distribution. Particle diameter in these methods is evaluated from the laser diffraction pattern or from the screen
33 aperture, where particle size is typically characterised by one value. To obtain knowledge about the shape, various static
34 microscopic methods are typically utilised. The major drawback of these methods is that in order to obtain statistically
35 reliable results is very laborious. One of the most promising techniques of size and shape measurement is to take
36 hundreds or thousands of photographs of the free flowing powder with a charged coupled device (CCD) and to measure
37 particle shape by using image analysing software. However, the drawback of this imaging method is that the finest
38 particles in the investigated sample are easily hidden or concealed by larger ones during image capture. Another
39 common problem is that it is impossible to distinguish the shape of very fine particles when the particle size is close to
40 the resolution limit. But when particles cannot be imaged optically, other microscopic methods can be used, such as
41 electron microscopy. In the imaging of the finest particles with electron microscopy, as well as with other methods,
42 there is also a problem that the particles are easily hidden or concealed behind larger particles.

43 In order to solve above-mentioned difficulties, the tube flow fractionation method together with the use of an imaging
44 unit (CCD) has been found to be a suitable method for measuring the morphology of different wood fibres [20]. After
45 fractionation, the largest particles are separated from the finest, and the concealment of the finest particles can be
46 avoided during imaging. Tube flow fractionation can also be used to separate samples into different fractions by size
47 and shape [21,22], which can be further analysed and used, for example, in enzymatic hydrolysis. This makes it possible
48 to analyse the largest particles separately by using optical microscopy and the finest particles by using electron
49 microscopy. However, there is not yet published detailed knowledge of whether the tube flow fractionation works with
50 wood powders or how well it can be applied in their characterisation.

51 In this work, different sized and shaped wood powders originating from Norway spruce (*Picea abies*) wood are
52 investigated with the tube flow fractionation method. The fractionated samples are characterised by measuring the light
53 transmittance of a light-emitting diode (LED, $\lambda = 450$ nm) and the particle dimensions from photographs captured with

a CCD camera. In this research, it is proven that tube flow fractionating separates fine wood powders by their size and shape. Wood powder can easily be separated into different fractions by leading the water-diluted sample flow into different beakers for certain time periods. The transmittance signal after tube flow fractionating yields information about the particle size, and optical imaging provides information about the morphology of the particles.

2. Materials and methods

2.1. Raw materials and mechanical processing

The raw material was Norway spruce (*Picea abies*) sawdust produced at Seikun saha (Pori, Finland), which is located in Western coast of Finland (61°29'N, 021°48'E) and delivered to the University of Oulu. After delivery, the sawdust was stored in a freezer. After storage, the sawdust was dried in an oven at a temperature below 105 °C and was sieved with a vibrating screen using an aperture size of 4 mm. The particles that could not get past the screen were rejected from the sample.

Milling of the dried and screened sawdust was accomplished with an air classifier mill, where a rotor impact mill ZPS (Hosokawa Alpine, Germany) was integrated with an air classifier 100 ATP (Hosokawa Alpine, Germany) by using different operating parameters (see Table 1). For further information about operating parameters on particle properties, see [23]. Air classifier milling was used also as pregrinding for jet and oscillatory ball milling experiments. In pregrinding, the finest particles were removed from the milled sample with air classification using the air classifier 100 ATP alone. The milling of preground samples was accomplished with an oscillatory ball mill CryoMill (Retsch, Germany) and a fluidised bed jet mill 100 AFG (Hosokawa Alpine, Germany) which was also integrated with the classifier 100 ATP. For further information about the operating parameters on the product properties in these fine grinding mills, see [24,25].

Table 1. Morphology characteristics of rotor impact milled wood powders. The quantity $d_{LS,50}$ refers to the median particle size measured by using the laser diffraction method.

Milling/classification	Feed	Varied operating parameters	Sample naming
Air classifier mill	Dried and screened sawdust	Rotor speed, classifier speed and impact angle	RM ($d_{LS,50}$)
Air classifier mill and post-classification	Dried and screened sawdust	Rotor speed, classifier speed and impact angle	RM+AC ($d_{LS,50}$)
Jet mill integrated with air classifier	RM+AC	Grinding air pressure, classifier speed	JM ($d_{LS,50}$)
Oscillatory ball mill	RM+AC	Milling time	OBM ($d_{LS,50}$)

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80 2.2. Preparation of diluted wood powder suspensions

81 One gram of the produced wood powder was diluted with deionised water in order to form 0.2% particle concentration.
82 Before dilution, the wood powder was dispersed by mixing 1 cm³ of a dispersant per 100 mg of the wood (dry mass).
83 The dispersant was prepared by diluting 2.5 g of Sokalan CP 5 (BASF) with 500 cm³ of deionised water. The diluted
84 sample was stirred for at least 30 min with a magnetic stirrer and held for 6 min in an ultrasonic bath to ensure complete
85 dispersal. Diluted and dispersed samples were applied for particle size measurement and tube flow fractionation.

86 In general, the amount of shrinking and swelling which takes place is directly proportional to moisture content changes
87 in the wood. Wood pieces shrinks and swells the greatest amount in the tangential direction and about half as much in
88 the radial direction (about 5%), and about 0.1% to 0.2% in the longitudinal direction [26]. Even if swelling increase
89 slightly particle size of analysed wood particles, this is not very significant source of error in practice. Pre-treatment of
90 samples were always similar and run in liquid mode with both devices (Tube flow fractionation method versus Laser
91 diffraction method) in order to better comparison of methods.

92 2.3. Particle size distribution

93 Volumetric particle size distribution was measured according to standard ISO 13320 [27] with an LS 13320 analyser
94 (Beckmann Coulter). Laser diffraction-based volumetric particle sizes were marked as $d_{LS,XX}$, where ‘XX’ stands for the
95 percentages in the cumulative distribution.

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2.4. Tube flow fractionation

Fractionation equipment has been presented earlier (see [22]), and this study utilises the same equipment and fractionation conditions. Fig. 1 presents a schematic picture of the tube flow fractionation equipment. The material of the tube is Teflon, and counterpressure during fractionation is 200 kPa. The theory of the fractionation principle has been described in more detail in [28]. In briefly when the sample is injected into the tube, the particles are distributed randomly. As the flow begins and proceeds, the slight turbulence in the transition flow regime begins to move the particles randomly inside the flow. The probability of being captured by the faster middle flow is higher for particles with one long dimension (e.g. high aspect ratio) or multiple long dimensions (e.g. flake type particles or large sawdust) than for particles with short dimensions (e.g. fines or small sawdust). Thus, the large particles tend to concentrate at the front end of the flow and finally come out from the tube first.

Fig. 1.

A 5 cm³ sample at a consistency of 0.2 % was fractionated with deionised water for 100 seconds at an average flow rate of (7.3–8.5) cm³s⁻¹ in a long plastic tube (diameter 4 mm) to separate the particles according to their size. The temperature of the suspensions stayed between 20.9 °C and 23.0 °C during tube flow fractionation. The Reynolds number for the water flow was 2200–3100 in the experiments. The variables of interest during fractionation, including flow velocity, pressure, temperature, sample volume, and consistency, are all maintained precisely at a constant level, as each variable has a direct effect on the fractionation. When keep fractionation conditions constant, certain particle sizes always exit the flow tube at the same time and this ensures that repeatability and reliable of tube flow fractionation methods is very good. The variation (standard deviation) of the combined sample fractionation and optical image analysis has been determined to be 4.5% and based on 23 parallel sample analysis [22]. Tube flow fractionation technique is use also in commercial pulp analysator (Valmet MAP - Valmet Pulp Analyzer, Valmet Automation), which is the online measurement of micro-scale details of fiber properties, including fibrillation, vessel segments, flocs and other particles [29].

2.5. Transmittance signal

After the fractionation of the sample, the signal detection unit, RM3 (Valmet Automation, Finland) was activated (see Fig. 1) where LED ($\lambda = 450$ nm) was used as the light source. In total, 2200–2600 transmittance signal values were collected while the sample flowed through the RM3- LED and transmittance signal detection unit. The zero positions of

the transmittance values were corrected by reducing the average transmittance signal obtained for the water flow. The beginning and the end of the signal (see Fig. 2b) were also removed due to random disturbance at the beginning of the signal. The restricted signal was then normalised to 100%, and the time count was started from the beginning of the restricted signal (Fig. 2c).

Fig. 2.

2.6. Optical microscopy by CCD camera

The wood particles in the fractionated particle-water suspension were visualised with a high-definition CCD camera unit (Basler acA1400-30gm, Germany) which captured approximately 1000 individual pictures including between 30,000-200,000 individual particle analysed per sample point (see Fig. 1). Image analysis of individual particles was accomplished by applying a Metso IMG image analysis software (Valmet Automation, Finland) and practically image analysis measurement sensitivity is around 1 µm. Image analysis procedure can be divided in six various steps: (1) image formation, (2) image scanning, (3) feature detection (count, shape, size etc.), (4) feature analysis, (5) data processing and (6) data presentation. Image analysis procedure has been clarified in Appendix A. Fig. 3 presents three example pictures and the transmittance signal of a wood powder sample taken after tube flow fractionation. The photographs obtained with the CCD camera were used to evaluate the cumulative projected area based the projected area diameter

$$d_p = 2\sqrt{\frac{A}{\pi}} \quad (1)$$

and the aspect ratio

$$AR = a/b. \quad (2)$$

In equations (1) and (2), A is the projected area of the particle, a is the maximum Feret's diameter of the projected particle and b is

$$b = \frac{A}{\pi a}. \quad (3)$$

Fig. 3.

3. Results and discussion

3.1. Comparison of measurement techniques

The measurement process starts not with the actual measurement, but with the sampling. The sample is usually a small sub-volume of an entire process batch. In order to obtain relevant information about batch, the sample needs to be representative. Spinning Riffler device has been used in this research in order to have representative samples from wood particles. It is also essential that all samples are well dispersed (see Chapter 2.2.), as agglomerates may plug the flow channel or misrepresent the actual results.

Traditionally, particle characterization methods have been gravimetric. However, since mesh screening does not produce fractions with sharp size limits, gravimetric techniques only give approximate information about particle size. The flow cytometer (FCM) is one potential method, but main limitations of this methods is quite narrow size range (from approximately 0.2 μ m to 100 μ m) and plenty of samples analysed in this research had particle size over that limit. The optical imaging methods are very suitable for analysing of particles like wood powders, because wood particles are typically proper size and contrast between background and wood particles are evident (i.e. grey vs. black). In the execution of the measurement, enough particles must be measured to give good statistics; typically several thousand, so 30,000-200,000 individual particle has been analysed per sample point in this research. Many measurement techniques have international standard procedures that can be followed to ensure reproducible results. In addition determination of aspect ratio of wood particles are focused in this research and method based on laser scattering is not possible to analyse aspect ratio but only sphere-equivalent diameter (SED). Table 2 summarise some measurement techniques, which can be used for particle size characterisation.

Table 2. Some measurement techniques, which can be used for particle size characterisation. The output size is the given size property, for example circle- or sphere-equivalent diameter (CED, SED).

Technique	Size range	Output size	Principle
Sieve	>20 μ m	Upper and lower limit of size	Gravimetric
Flow cytometry	0.2-100 μ m	CED, SED	Optical scattering
Laser diffraction	40nm-2000 μ m	SED	Optical scattering
Static microscopy	>1 μ m	Length, area, volume, aspect ratio	Optical imaging

Tube flow fractionation + image analysis	1µm-2000µm (particle size), 10µm-2000µm (aspect ratio)	Length, area, volume, aspect ratio	Optical imaging
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3.2. Different sized wood powders

Fig. 4 shows particle sizes of various wood samples analysed with tube flow fractionation method (based on image analysis) and laser diffraction method (based on optical scattering). Particle size of wood powders increased quite logically with both methods (image analysis and laser diffraction). Nevertheless same particle size was measured with laser diffraction (for instance around 200µm), even if average aspect ratio varies from 7 to 10. That is because method based on laser scattering is not take account of aspect ratio of particles. It has to also noted that image analysis particle size based on planar surface of particle while laser diffraction based on sphere-equivalent diameter (SED).

Fig. 4.

Fig. 5 shows the particle size distributions of various rotor impact milled (RM) wood powders and rotor impact milled wood powder that has been classified after milling (RM+AC). In the case of the classified sample (RM+AC), two replicated measurements were done on a single sample. The particle size distribution of the RM samples with the median particle size over 200 µm was cut at the large end, which means that there were particles larger than 2 mm present in these samples. In Fig. 5, when comparing the non-classified sample having a $d_{LS,50}$ of 202.6 µm with the classified sample having a $d_{LS,50}$ between 194.3 µm and 202.8 µm, it can be seen that the width of the particle size distribution of the classified sample was narrower.

Fig. 5.

It has been noted that tube flow fractionation classifies particles primarily according to their hydrodynamic length [30,31], but particle width and thickness have a smaller but still statistically significant effect on fractionation time [21]. Fig. 6 shows the normalised LED transmittance signal after tube flow fractionation of the wood powder samples. In Fig. 6 can be seen single- or double-peaked signal profiles, depending on the median particle size of the wood powders. The signal peak occurring during the first 3 s is called the 1st peak, and the signal peak occurring after the first 3 s is the 2nd peak. Fig. 6 shows the cumulative projected area-based distributions as a function of d_p and the aspect ratio (AR), by considering three different cases for each sample: the whole sample, only the particles from Fig. 5 (particle group 1, PG1) which flow during first the 3 s and only the particles in Fig. 5 (particle group 2, PG2) which flow after 3 s. The cumulative area distributions of all other RM and RM+AC samples are shown in Appendix B (Figs. B.1-B.6).

194 According to Figs. 7, B.1-B.6, the cumulative projected area of PG1 is composed mainly of particles with a larger d_p
195 and AR than in the case of a whole sample or PG2. On the other hand, the projected area of PG2 is composed of
196 particles with a smaller d_p and AR in comparison to the whole sample (Figs. 7, B.1-B.6). Between the RM samples,
197 there are substantial differences in d_p and AR values when considering only PG1 or the whole sample, but there are
198 only small differences when considering only PG2 (Figs 7 and B.1-B.6). The differences in the cumulative area
199 distribution between the whole sample and PG2 diminish as $d_{LS,50}$ decreases in the case of the RM samples (Figs. 7 and
200 B.1-B.6). For the RM samples, PG2 comprises a much larger area in the transmittance signal when $d_{LS,50}$ decreases (Fig.
201 6). This can be interpreted so that the whole sample is more like PG2 as the $d_{LS,50}$ decreases. Therefore, it is very logical
202 that the particle size and shape differences between the whole sample and PG2 diminish as $d_{LS,50}$ decreases in the case
203 of the RM samples. In the case of the classified sample (RM+AC), the removal of fine particles makes the signal area of
204 PG2 even smaller in comparison to the RM samples with larger $d_{LS,50}$ (Figs. 7, B.1-B.6). This can be interpreted so that
205 in the case of RM+AC, the whole sample is more like PG1 than in the case of the RM samples. The reason for this is
206 that in the case of RM+AC, the finest particles have been removed after milling. Therefore, it is logical that the
207 classified sample has the foremost difference in the cumulative area distributions between the whole sample and the
208 PG2 (Fig. 7, B.1-B.6). According to the results (Figs 7, B.1-B.6), tube flow fractionation classifies fine wood particles
209 by size and shape. The transmittance signal of fractionated samples (Fig. 6) provides insight about the particle size of
210 wood powders, where the presence of the 1st peak indicates the presence of large particles, and the presence of the 2nd
211 indicate the presence of fine particles. By considering the particle size distributions of the RM+AC sample, which has
212 no 2nd signal peak (Fig. 6), and the 'RM (23.4 μm)' sample, which has a negligible 1st peak (Fig. 6) in Fig. 5, the large
213 particles could be arbitrarily considered as the particles with a d_{LS} over 50 μm and the fine particles as those with a d_{LS}
214 less than 50 μm . Median particle sizes ($d_{LS,50}$) could not be evaluated directly from the transmittance signal due the
215 differences in the transmittance signals between the RM and RM+AC samples with a similar $d_{LS,50}$.

216 **Fig. 6.**

217 **Fig. 7.**

218 From Figs. 7, B.1-B.6 it can be seen that the particles with an aspect ratio of 1 comprise approximately 10% (AR10) or
219 an even larger projected area in the case of the whole sample and PG2. In Fig. 8, aspect ratios of the particle are shown
220 as a function of d_p , where samples are grouped into four (I–IV) different groups. According to Fig. 8, the smallest
221 particles with a d_p lower than 10.7 μm all have aspect ratios of 1. It is very unlikely that wood particles form a perfect

circle shape during mechanical processing, and therefore, it is suggested that particles having an aspect ratio of 1 are too small for reliable shape recognition. When considering the particle size ranges of $d_{LS,50} > 200 \mu\text{m}$ (group II in Fig. 8) and $d_{LS,50}$ between $47\mu\text{m}$ and $52 \mu\text{m}$ (group III in Fig. 8), it can be seen that milling to a finer particle size range can produce elongated particles ($AR > 10$) in the particle size range $d_p > 80 \mu\text{m}$. Milling to an even finer particle size range, $d_{LS,50} < 35 \mu\text{m}$ (group IV in Fig. 8), can produce elongated particles ($AR > 10$) with d_p between $65 \mu\text{m}$ and $80 \mu\text{m}$ (Fig. 8). This means that rotor impact milling to a finer size range produces smaller elongated particles. In Fig. 8, the shapes of the RM samples in the particle size range $d_p \leq 20 \mu\text{m}$ are very similar. One probable reason for this is that the shapes of the particles cannot be distinguished from others in this particle size range. The morphology of the classified sample (RM+AC) is different from that of the RM samples, especially when $d_p > 20 \mu\text{m}$ (group I in Fig. 8).

Fig. 8.

3.2. Comparison of wood powders having different morphology

Jet milling and oscillatory ball milling can be used to produce wood powders having a similar $d_{LS,50}$ but different shapes and relative degree of cellulose crystallinity [25]. Therefore, both jet milled (JM) and oscillatory ball milled (OBM) samples are taken under consideration where in both cases, the feed material has been RM+AC. Fig. 9 shows the differential volume-based particle size distributions of jet milled and oscillatory ball milled samples. When comparing jet milled and oscillatory ball milled samples, it can be seen that there are very small differences in the particle size distributions between samples that have $d_{LS,50}$ close to each other.

Fig. 9.

Fig. 10 shows the transmittance signal of fractionated JM and OBM samples. As in the case of rotor impact milled wood (Fig. 6), there can again be distinguished single- and double-peaked profiles independent from the used milling technique. It can also be seen that the area covered by the transmittance signal increases after 3 s, while the $d_{LS,50}$ decreases (Fig. 10). There are only very small or negligible differences in the signals between milling techniques (Figs. 6 and 10). The largest difference in the signals between milling techniques seems to be the time where the 2nd signal peak occurs in the finest samples; in the jet milled and rotor impact milled samples, the 2nd peak can be found right before 8 s (Figs. 6 and 10), but for the oscillatory ball milled samples, it is right after 8 s (Fig. 10).

Fig. 10.

Fig. 11 shows the cumulative projected area distributions as a function of d_P and the aspect ratio for jet and oscillatory ball milled samples with $d_{LS,50}$ close to each other. Appendix B (Figs B.7-B.15) shows the cumulative projected area distributions as a function of d_P and AR for all investigated jet and oscillatory ball milled samples. The cumulative area distributions of the JM and OBM samples (Figs. 11, B.7-B.15) show similar kinds of trends as a function of $d_{LS,50}$, as was discussed earlier in the case of rotor impact milled wood powders (see Section 3.1). In Fig. 12, when comparing jet milled and oscillatory ball milled samples with a $d_{LS,50}$ close to each other, it can be seen that oscillatory ball milled powders have a significantly lower aspect ratio. This can be observed from the CCD photos shown in Fig. 12, where the jet milled powders seem to retain an elongated shape. This information cannot be interpreted from the differential particle size distribution (Fig. 9) or from the signals (Fig. 10). When comparing the cumulative projected area distributions as a function of d_P between samples OBM (20.3 μm) and JM (19.9 μm), the oscillatory ball milled PG1 seem to have a significantly larger d_P (Fig. 11). It is suggested that this is due to the rounder or more flake-shaped particles in the OBM (20.3 μm) PG1, which has a larger projected area in comparison to the elongated particles in JM (19.9 μm) PG1 (Fig. 11).

Fig. 11.

According to Fig. 12, aspect ratio as a function of d_P follows a different trend for JM samples than for OBM samples, even when their $d_{LS,50}$ are close to each other. Therefore, it seems that jet milling favours the production of small and elongated particles, whereas oscillatory ball milling favours the production of rounder particles (Fig. 12). All of the OBM samples can be found in groups I–III, and the samples with $d_{LS,50} > 44 \mu\text{m}$ are located very close to the border of group I.

Fig. 12.

3.3. Separation of wood powder to different size fractions

With the imaging technique used, the differences in the shapes between wood powders were distinguishable when $d_P > 20 \mu\text{m}$. Therefore, it seems that particles with $d_P \leq 20 \mu\text{m}$ would need measurement techniques, such as electron microscopy, that can distinguish the shapes of these very small particles. According to Fig. 13, these particles do not make a significant contribution in PG1 and have the highest signal peak after 10 s. Therefore, the largest particles including undispersed or agglomerated particles, which are found in PG1, can be fractionated by tube flow fractionating without affecting the particles with $d_P \leq 20 \mu\text{m}$ according to image analysis. In addition the largest particles can also be

275 further investigated without the presence of the particles having $d_p \leq 20 \mu\text{m}$ after fractionation. Tube flow fractionation
 276 offers a simply and fast way to separate the largest particles from the smallest, where the morphology of the largest
 277 particles can be distinguished from CCD photos. The finest particles, which are too small to be distinguished by optical
 278 microscopy, can be physically removed from the flow to their own fraction.

279 **Fig. 13.**

280 **4. Conclusions**

281 Tube flow fractionation classifies fine wood particles by size and shape. Transmittance measurement after tube flow
 282 fractionation can be used to provide insight about the particle size distribution of wood powders. An optical imaging
 283 device connected to the tube flow fractionation equipment provide an excellent and non-laborious method to measure
 284 the morphology of visually recognisable fine wood particles. Tube flow fractionation prior to optical imaging helps to
 285 avoid concealment of the finest particles behind the larger particles. Tube flow fractionation can also be used to separate
 286 wood particles into different particle size classes for further analyses and use.

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356

357 **Fig. 1.** Schematic picture of the fractionation equipment. In the figure ‘FR1’, ‘FR2’, ‘FR3’ and ‘FR4’ are containers
 358 where different fractions of the fractionated sample are poured in.

359

360 **Fig. 2.** Signal treatment procedure a) the original LED transmittance signal of one RM sample after tube flow
 361 fractionation, b) the signal after zero position correction and restriction and c) the normalised signal where the time
 362 starts counting from the beginning of the restricted signal.

363

364 **Fig. 3.** Light-emitting diode (LED) signal transmittance signal as a function of time and three CCD photos of
 365 fractionated wood powder sample taken after certain time periods.

366

367 **Fig. 4.** Particle sizes of various wood samples analysed with tube flow fractionation method (based on image analysis)
 368 and laser diffraction method (based on optical scattering) including standard deviation of both methods. Average aspect
 369 ratio results based on image analysis.

370

371 **Fig. 5.** Volumetric differential particle size distributions of rotor impact milled powders.

Fig. 6. Normalised LED signal transmittance curves as a function of time for fractionated rotor impact milled wood powder samples. In the figure, PG1 stands for particle group 1, which flows before 3 s, and PG2 for particle group 2, which flows after 3 s.

Fig. 7. CCD photos and cumulative area distributions of fractionated rotor impact milled wood powder samples. The cumulative area distribution which considers only PG1 (see Fig. 5) is marked with ‘•’; PG2 (see Fig. 5) is marked with ‘•’, and the whole sample is marked with ‘•’.

Fig. 8. Aspect ratio as a function of projected area diameter d_p for rotor impact milled wood powder samples. Black dashed lines divide samples into groups I–IV.

Fig. 9. Volumetric differential particle size distributions of a) jet milled (JM) and b) oscillatory ball milled (OBM) wood samples, where in both, RM+AC has been used as the feed material.

Fig. 10. Normalised LED signal transmittance curves as a function of time for fractionated a) jet milled (JM) and b) oscillatory ball milled (OBM) wood samples where, in both, RM+AC has been used as the feed material.

Fig. 11. CCD photos and cumulative area distributions of fractionated jet milled and oscillatory ball milled wood

powder samples. Cumulative area distribution that considers only the PG1 (see Fig. 9) is marked with ‘•’, PG2 (see Fig.

9) is marked with ‘•’ and the whole sample is marked with ‘•’.

Fig. 12. Aspect ratio as a function of projected area diameters for a) jet milled and b) oscillatory ball milled wood powder samples. Black dashed lines divide samples into groups I–IV, which are same as in Fig. 7.

Fig. 13. Relative area of the particles having $d_p \leq 20 \mu\text{m}$ as a function of time after fractionation.

398

399 **Fig. A.1.** Schematic description of the image analysis procedure.

400

401 **Fig. B.1.** Cumulative area distributions of different RM+AC and RM samples as a function of d_p . Particle group 1 is
 402 marked with '■', particle group 2 is marked with '●' and the whole sample is marked with '▲'.

403

404 **Fig. B.2.** Cumulative area distributions of different RM+AC and RM samples as a function of AR. Particle group 1 is
 405 marked with '■', particle group 2 is marked with '●' and the whole sample is marked with '▲'.

406

407 **Fig. B.3.** Cumulative area distributions of different RM samples as a function of d_p . Particle group 1 is marked with '■',
 408 particle group 2 is marked with '●' and the whole sample is marked with '▲'.

409

410 **Fig. B.4.** Cumulative area distributions of different RM samples as a function of AR. Particle group 1 is marked with
 411 ‘■’, particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

412

413 **Fig. B.5.** Cumulative area distributions of different RM samples as a function of d_p . Particle group 1 is marked with ‘■’,
 414 particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

415

416 **Fig. B.6.** Cumulative area distributions of different RM samples as a function of AR. Particle group 1 is marked with

417 ‘■’, particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

418

419 **Fig. B.7.** Cumulative area distributions of different JM samples as a function of d_p . Particle group 1 is marked with ‘■’,
 420 particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

421

422 **Fig. B.8.** Cumulative area distributions of different JM samples as a function of AR. Particle group 1 is marked with

423 ‘■’, particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

424

425 **Fig. B.9.** Cumulative area distributions of different JM samples as a function of d_p . Particle group 1 is marked with '■',
 426 particle group 2 is marked with '●' and the whole sample is marked with '▲'.

427

428 **Fig. B.10.** Cumulative area distributions of different JM samples as a function of AR. Particle group 1 is marked with
 429 '■', particle group 2 is marked with '●' and the whole sample is marked with '▲'.

430

431 **Fig. B.11.** Cumulative area distributions of different OBM samples as a function of d_p . Particle group 1 is marked with
 432 ‘■’, particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

433

434 **Fig. B.12.** Cumulative area distributions of different OBM samples as a function of AR. Particle group 1 is marked with
 435 ‘■’, particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

436

437

438

Fig. B.13. Cumulative area distributions of different OBM samples as a function of d_p . Particle group 1 is marked with ‘■’, particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

439

440 **Fig. B.14.** Cumulative area distributions of different OBM samples as a function of AR. Particle group 1 is marked with

441 ‘■’, particle group 2 is marked with ‘●’ and the whole sample is marked with ‘▲’.

442

443 **Fig. B.15.** Cumulative area distributions of different OBM samples as a function of d_p and AR. Particle group 1 is
 444 marked with '■', particle group 2 is marked with '●' and the whole sample is marked with '▲'.

445