

Final report

Project details

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Project identification (program abbrev. and file)	AUWP – ForskEL 12325
Name of the programme which has funded the project	ForskEL
Project managing company/institution (name and address)	Technical University of Denmark – Department of Chemical and Biochemical Engineering (DTU Chemical Engineering)
Project partners	DTU – Chemical Engineering Ørsted Hofer A/S
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1.1 Short description of project objective and results

(Danish/ English 600-800 characters)

English

The project supports an efficient and fast conversion of pulverized coal-fired power plant boilers to operate on biomass fuels by investigating the key issues that can ensure an efficient and effective milling of biomass pellets in existing power plant coal mills. The study has highlighted that the industrial pelletization process has a larger impact on modifying the original wood particle shape than the pellet milling step and has been able to demonstrate the different grinding behavior of pellets with a different origin. Promising results have been found to predict the grinding behavior of wood pellets by laboratory testing in a roller mill equipped with a zigzag classifier. For the characterization of the milled particle morphology, traditional sieve analysis has shown to suffer from some limitations. Dynamic image analysis has shown the great importance of determining the length and shape of wood particles and hence has the potential to replace sieve analysis in the laboratory.

Danish

Projektet understøtter en effektiv og hurtig konvertering af pulveriserede kulfyrede kraftværkskedler til drift af biomassebrændstof ved at undersøge de vigtigste problemer, der kan sikre en effektiv fræsning af biomassepellets i eksisterende kulkraftsværkværker.

Undersøgelsen har fremhævet, at den industrielle pelleteringsproces har større indflydelse på ændring af den originale træpartikelform end pellet fræsningstrinnet og har været i stand til at demonstrere forskelligt slibeadfærd for pellets med en anden oprindelse.

Der er fundet lovende resultater at forudsige slibeadfærd for træpiller ved laboratorietest i en rullefræser udstyret med en zigzag klassificeringen.

Til karakterisering af den formalede partikelmorfologi har traditionel sigte-analyse vist sig at lide under nogle begrænsninger.

Dynamisk billedanalyse har vist den store betydning af bestemmelse af træpartiklernes længde og form og har derfor potentialet til at erstatte sigteanalyse i laboratoriet.

1.2 Executive summary

The utilization of sustainably produced wood pellets in the existing coal suspension-fired power plants can offer a cost-efficient option of mitigating greenhouse gas emissions. However, the fibrous and non-brittle structure of wood poses challenges with regard to the size reduction process of wood pellets in the existing coal mills. There is a lack of understanding of the pellet grinding behavior and morphology (i.e., size and shape) of milled pellet particles, and how the particle morphology can be related to the wood pellet processing history. New knowledge in this area has the potential to promote efficient and fast conversion of coal-fired power plants to the firing of milled pellets.

For this purpose, the AUWP project investigated the pellet grinding behavior in lab-, pilot- and industrial-scale mills. The grinding behavior was determined by measuring the specific grinding energy and analyzing the morphology (size and shape) of milled and internal pellet particles. New methods were introduced to characterize the grinding behavior of wood pellets by laboratory testing, and to investigate if it is possible to predict grinding results in power plant coal mills. The study used two industrial pellet qualities (I1 & I2), and two pellet types made from Austrian pine (softwood) and European beech (hardwood) stem wood. Pellets were characterized according to standardized methods.

The research has found that the industrial pellet production process has a larger impact on modifying the pre-densified wood particle shape than the pellet grinding process. The pellet grinding behavior can be related to the mill type and pellet processing history (including feedstock type and internal pellet particle size distribution). It was shown that grinding of raw and pelletized beech produces finer, rounder and less elongated particles, and requires a lower grinding energy than pine. The proposed laboratory roller mill-classifier system has the potential to assess the grinding properties of different pellet qualities. The comparison with grinding results from industrial mills showed that similar particle size reduction ratios can be achieved.

This project presents relevant experimental data that provide an understanding of the morphology changes occurring during the industrial pelletization process. It also has great value for power plant operators to maximize the pellet grinding capacity in the existing coal mills.

1.3 Project objectives

The objective of the project has been to support an efficient and fast conversion of pulverized coal-fired power plant boilers to operate on biomass fuels by investigating the key issues that can ensure an efficient and effective milling of biomass pellets in existing power plant coal mills.

In order to achieve the main project objective, the project also had the following underlying objectives:

- Investigating and testing new characterization methods focused on the morphology (size and shape) of grinded wood pellet particles.
- Investigating novel characterization methods to evaluate the grinding properties of wood pellets.
- Working in close collaboration with the industrial partners Ørsted and Hofer to run full-scale campaigns.
- Evaluating the influence of the different steps in the pellet production process on the wood particle characteristics, final pellet quality and grindability properties.
- Relating the properties of wood pellet particles grinded in a coal mill with the properties of the original wood pellets.
- Comparing milling measurements performed in the laboratory with measurements obtained onsite at the power plant to establish correlations.
- Evaluating the combustion performance of densified wood particles versus non densified wood particles for both softwood and hardwood

In general, the project evolved according to the original plan and as foreseen. However, a 6 months extension (until the 31st October 2019) was requested in order to run some additional measurements on full-scale samples from power plants, compare them with those analyzed in previous heating seasons and relate them with the results obtained in laboratory scale mills. This extension has also been used to prepare, submit, and arrange for publication two scientific manuscripts.

All the planned milestones are completed except M 2.2 and M 4.4 that are partially completed. In the following paragraphs, we describe how the project was implemented, the work done for each WP and the milestones completed.

WPO Project Management

The tasks included in WPO have been completed on time and according to the plan. The contract between the project participants was made and the kick-off meeting was held on 27th August 2015 at DTU-Risø Campus. There has been regular project meetings with all the project partners as well as monthly meetings with the PhD Student (Marvin Masche). Half-year reports have been prepared and submitted to EUDP.

WP1 Pellet production account and initial fuel characterization

The work done in this WP followed the initial plan and included:

- A review on the state of the art on biomass pellet production and the fundamental physicochemical mechanisms that control the quality of biomass pellets. The review also included the different existing characterization methods for raw biomass feedstock, pellet quality and milling behavior. This review on the state of the art resulted in an internal report that was made available and distributed among all the project partners. The most relevant parts of the report are included in the PhD thesis of Marvin Masche.

- Initial characterization of all the pellets used in the tests. This included I1 and I2 pellets from full-scale tests (WP2) and produced beech and pine pellets (WP 3 & 4). The characterization included, at least, proximate & ultimate analysis, net calorific value, pellet size measurements, durability, internal particle size distribution, internal particles morphology. The results of these characterization tests are included in each of the scientific papers published as a result of the AUWP project activities.

Milestones included in WP1:

- *M 1.1 Report on the state of the art in pellet production performed by large wood pellet producers – Completed*
- *M 1.2 Characterization of commercial pellets to be used in the experiments of other WPs, including an attempt to link the characteristics with the specifications of the given production process – Completed*

WP2 Full scale power plant measurements

As an important part of the project, two campaigns focusing on grinding and combustion test at Ørsted and HOFOR PF power plants were planned. However, only one full-scale campaign could be done. It was at Amagerværket (Copenhagen, HOFOR) in October/November 2016. The second campaign, that should have been at Ørsted was replaced by analysis, tests and results comparison performed on different quality, origin and wood dust samples obtained during the normal operation of the plant over the duration of the project.

The full-scale campaign at Amagerværket included methodical surveillance and operational data collection for two pellet qualities (I1 and I2) at different mill loads. An extensive data collection program was carried out, including sampling of pellets and sampling of dust after the coal mill. Data from onsite measuring equipment was also collected. The observation of flame stability and determination of the degree of fuel burnout was included to relate them to the characteristics of the fuel and to the operation conditions of the mill. The results from this campaign are reported in the scientific article *Masche, M. et al (2018) Wood pellet milling tests in a suspension-fired power plant*. The article presents the results regarding:

- Determination and evaluation of the best sampling method for comminuted pellets conveyed to the burners.
- Comparison of the morphology (size and shape) of material within pellets with that from pellets comminuted at different mill loads.
- Analysis of the influence of different pellet qualities on the milling process.
- Identification of the optimal conditions for comminuting wood pellets.

Milestones included in WP2:

- *M 2.1 Results of full-scale power plant measurement campaign 1 – Completed*
- *M 2.2 Results of full-scale power plant measurement campaign 2 – Not completed – This full-scale campaign at Ørsted was initially planned for Studstrupværket but the reconversion works in the plant to fully operate with wood pellets, together with the reorganization and entrance in the stock market of Ørsted (formerly Dong Energy) prevented the consortium from achieving this milestone. Instead, samples of different pellets and wood dust from Avedøreværket were analyzed to determine possible influences of wood origin (softwood or hardwood) and pellet qualities in the milling properties.*

WP3 Pellet characterization and milling properties study

The work in this WP was performed according to the initial plan and focused on:

- Comminution of biomass pellets into particles using different lab-scale milling principles (ball mill, disc mill, knife mill & roller mill) to relate them to the properties of the fuel (pellet quality and type of wood (softwood & hardwood)) and to the properties of the milled particles (size, shape and density). The results of this study are presented in a scientific paper currently under review *Masche, M. et al (2019) Grinding performance of wood pellets in laboratory-scale mills.*
- Comparison of the milling tests results at lab-scale with those from the full-scale campaign at Amagerværket for I1 and I2 pellets. The results of this study are presented in a scientific paper currently under review *Masche, M. et al (2019) An investigation of the grindability of wood pellets in a lab-scale roller mill with classifier.*
- Studying the changes in physical properties (particle size, shape, density) of Austrian pine (softwood) and European beech (hardwood), as they were mechanically processed from wood chips to pellets and then to milled pellets using semi-industrial mills and equipment. The results from this study are reported in a published scientific manuscript *Masche, M. et al (2019) From wood chips to pellets to milled pellets: the mechanical processing pathway of Austrian pine and European beech.*

Milestones included in WP 3:

The two milestones planned are completed and they are included in the scientific manuscripts published and in the PhD thesis presented and defended by Marvin Masche.

- *M 3.1 Recommended characterization method for biomass pellets to evaluate the grinding properties of these pellets – Completed*
- *M 3.2 Correlations between laboratory based measurements and onsite measurements at the power plants – Completed*

WP4 Dust particle size, morphology and combustion characteristics study

The studies included in this WP were accomplished following the initial plan and included:

- Development and verification of a method to determine the characteristics of biomass particles that will influence the combustion process. Different methods including sieving, 2D imaging (Camsizer), laser diffraction (Mastersizer), SEM microscopy and 3D X-ray computed tomography were tested and compared to fully characterize the wood particles. Conclusions on these comparisons are included in Chapter II of Marvin Masche's PhD Thesis.
- Analysis of all the samples from WP 1-3 using sieving and 2D imaging technique (Camsizer). These results are presented in the published scientific papers and in the PhD thesis.
- Single particle combustion studies on cube particles of 3 mm made of raw and pelletized beech and pine wood. The results of these studies are presented in the PhD thesis of Marvin Masche and are part of the Bachelor Thesis of Benjamin Clausen.

Milestones included in WP 4:

- *M 4.1 Characterization study based on morphology of grinded wood pellet particles completed and recommendations for the characterization of particulate material provided - Completed*
- *M 4.2 Particle characterization work finalized - Completed*
- *M 4.3 Combustion characterization work finalized - Completed*

- *M 4.4 New biomass pellet specifications based on optimal combustion properties – Partially completed* - Important conclusions were drawn and they are presented in the different scientific papers and the PhD Thesis. However, not a whole set of new biomass pellet specifications were established because additional tests and investigations are required.
- *M 4.5 Recommendations for the pellet production process in order to improve the pellet quality – Completed*

1.4 Project results and dissemination of results

The results of the AUWP project will be discussed in the following sections and are divided in four areas:

- Full scale power plant measurements
- Milling properties study for wood chips and pellets
- Dust particle size and morphology characterization
- Single particle combustion studies for densified versus raw wood cubes

1.4.1 Full scale power plant measurements results

The study assessed the large-scale milling behavior of industrial wood pellet qualities in vertical roller mills (VRMs) at the suspension-fired combined heat and power plant Amagerværket unit 1 (AMV1), located in Copenhagen (Denmark). AMV1 has a capacity of 80MW electricity and 250MW heat. Originally designed for coal, AMV1 was converted in 2010 to operate 100% on biomass, mainly wood pellets.

The large-scale pellet comminution in VRMs and subsequent particle separation in dynamic classifiers were tested to see if they are affected by the particle size distribution (PSD) of material within pellets, also known as the internal pellet PSD. The main objectives of the study were:

- To evaluate the sampling method for comminuted pellets conveyed to the burners.
- To compare the morphology (size and shape) of internal pellet particles with that from comminuted pellet particles at different mill loads.
- To analyze the influence of different pellet qualities on the milling process.
- To identify optimal conditions for comminuting wood pellets.

The sampling probe designed for the full-scale campaign (Figure 1) was proven to be well suited for the purpose. The sampling was done according to ISO 14488:2007, by using isoaxial and isokinetic sampling with cyclone to extract the fine comminuted pellet product via a vacuum with a mean air velocity of 30 m/s from inside the burner pipes. The samples were extracted from the burner pipe center, as it showed a more stable particle flow than along the pipe wall. Isokinetic wood dust sampling from vertical and symmetric mill outlet pipes is the preferred sampling method because sampling from horizontal pipes increased the risk of particle segregation. Sampling from asymmetric mill outlet pipes showed a more uneven wood dust distribution regarding fineness and mass flow of the comminuted particles.

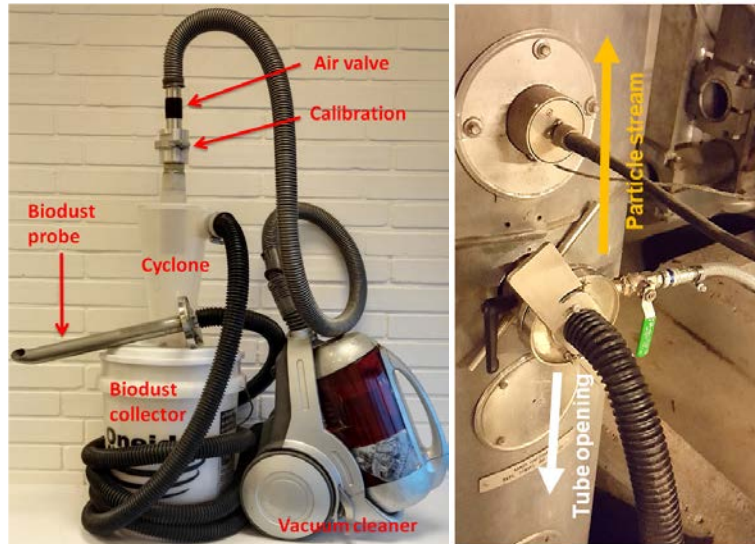


Figure 1 Designed isokinetic sampling device (cyclone vacuum) for the full-scale campaign at Amagerværket

The full-scale campaign also showed that internal particle size distribution of wood pellets affects the large-scale pellet milling behavior and the subsequent particle size classification. Pellets with finer internal particles lead to a finer comminuted product with lower specific grinding energy consumption (Figure 2). Comminuted pellet particles sampled from burner pipes were notably finer than internal pellet (feed) particles (Figure 2). At similar mill-classifier conditions, characteristic particle sizes of 0.50mm for comminuted I1 pellet particles (compared to 0.83mm for internal I1 pellet particles) and of 0.56mm for comminuted I2 pellets (compared to 1.09mm for internal I2 pellet particles), respectively, were obtained. This supports the hypothesis that the mills achieve a reduction of particle size compared with the internal pellet particles.

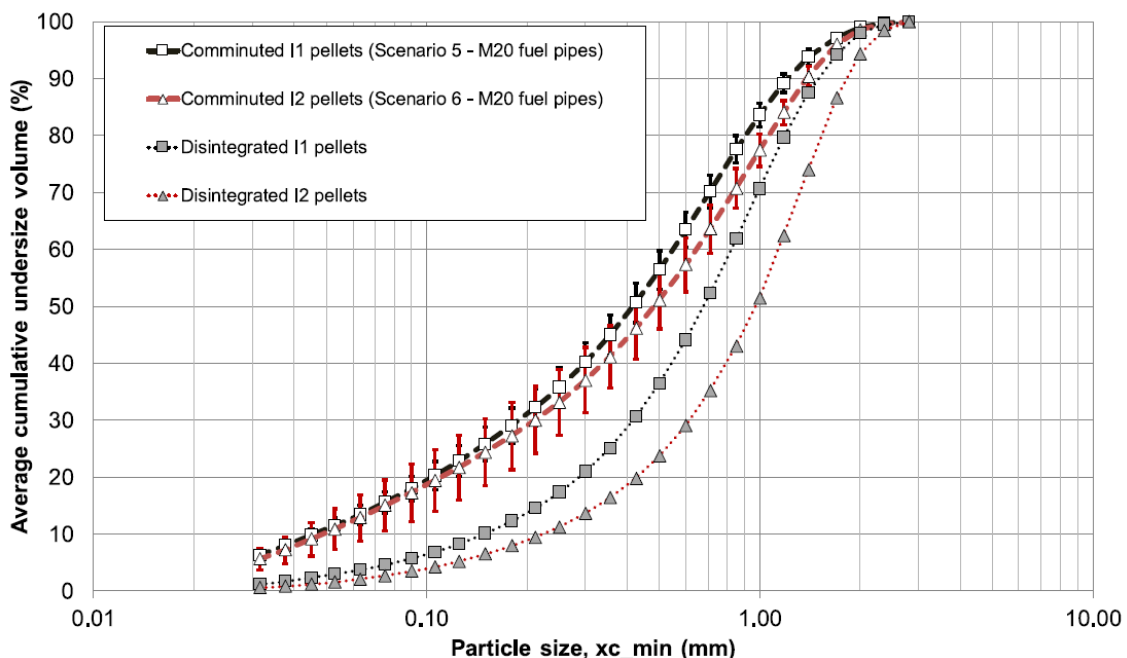


Figure 2 PSD comparison between disintegrated and comminuted pellets. Error bars represent one standard deviation within the different fuel pipes of the mill.

Another relevant result from the full-scale campaign is that roller mills do not affect the original elongated wood particle shape, regardless of the mill operating conditions (Figure 3).

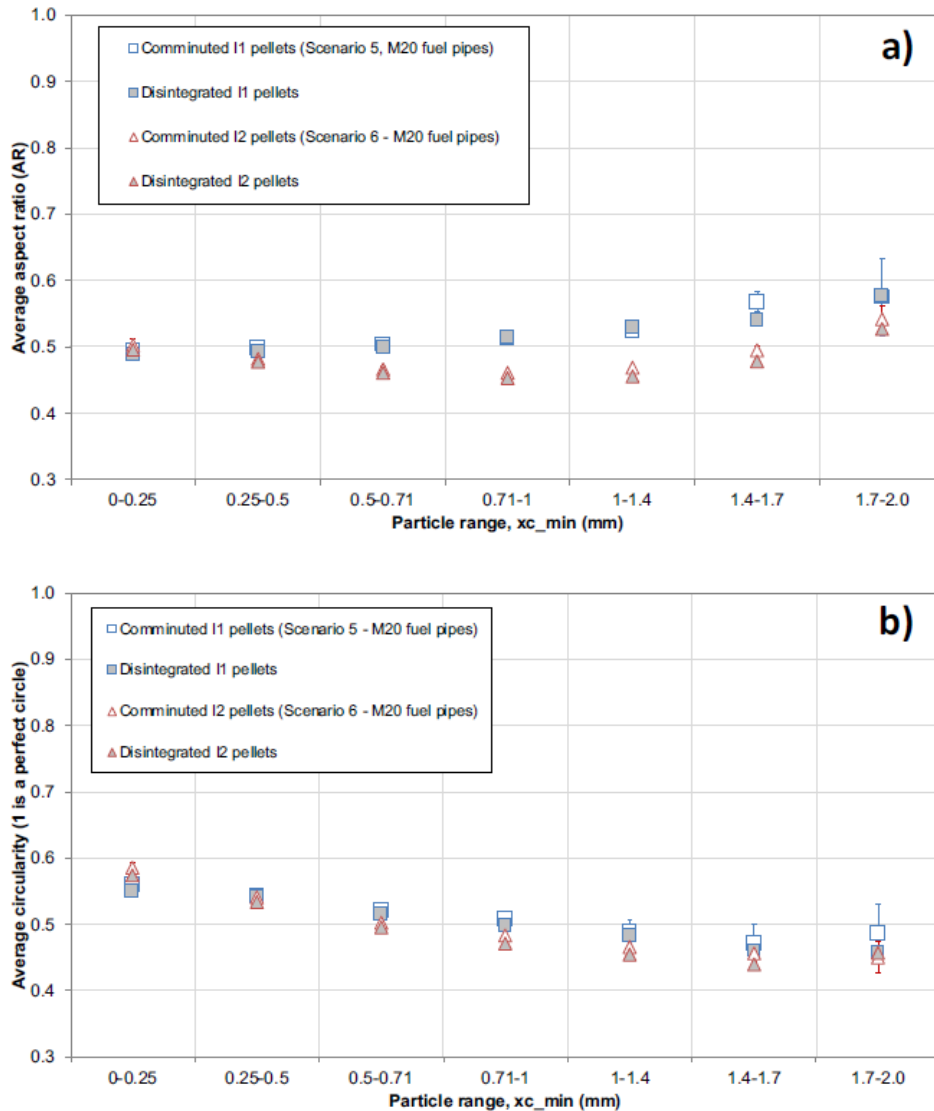


Figure 3 Average aspect ratio (a) and circularity (b) of disintegrated and comminuted I1 and I2 pellets. Error bars indicate one standard deviation within different fuel pipes of the mill.

The operation of the roller mill at higher loads and higher primary airflow rates has unfavorable effects on the mill power consumption, the differential mill pressure, and the classifier cut size. Only the specific energy consumption can be reduced, when the mill operates at higher loads.

The specific energy consumption for pellet comminution varies with the classifier cut size. At a constant mill load, an increased rotor speed and a reduced airflow rate lead to a smaller classifier cut size. However, this is at the expense of a higher energy consumption and a higher differential mill pressure.

1.4.2 Milling properties study for wood chips and pellets results

The full-scale campaign showed that the internal PSD of wood pellets plays an essential role in determining the PSD of comminuted pellet particles. Thus, a study was conducted to find out how the PSD and particle shape of internal pellet particles are obtained and, furthermore, which steps are crucial in determining the particle size and shape of material within pellets. The physical changes occurring during the mechanical processing of beech (hardwood) and pine trees (softwood) into chips, pellets, and milled pellets were assessed. All processing steps (Figure 4) were included in the study and carefully examined to provide knowledge of

how pelletization and comminution operations alter the physical properties of wood particles (size, shape, density).

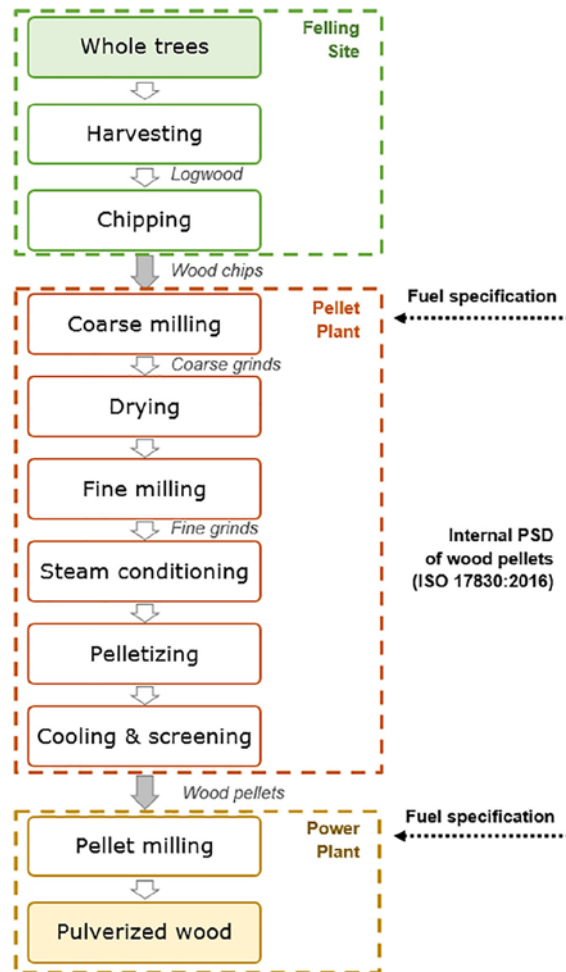


Figure 4 Major processing steps of converting whole trees to pellets for pulverized wood fired power plant boilers.

The study showed that the pelletizing process alters the shape and length of the wood particles more than the pellet milling step. There is a reduction in the particle length during pelletizing, which results in higher particle circularity and elongation (width-to length) ratio values (Figure 5). The average elongation ratio for beech particles increased from 0.38 (fine grinds) to 0.51 (disintegrated pellets) to 0.52 (milled pellets). In comparison, pine particles increased in elongation from 0.42 (fine grinds) to 0.48 (disintegrated pellets) to 0.49 (milled pellets).

It was also observed that milling beech produced more fines than pine in all milling steps using the exact same equipment with the same screen size (Figure 6). The comminution of beech and pine pellets in a hammer mill shifted the PSD to the left indicating a reduced width and length compared to internal pellet particles. This final milling step resulted in characteristic particle widths and lengths of 0.68 mm and 1.47 mm for milled beech pellets and 0.90 mm and 1.94 mm for milled pine pellets, respectively. Thus, the final milling step did not only disintegrate pellets into constituent internal particles but achieved some size reduction of the particles. Figure 6A shows that the size reduction in width was larger for pine pellet particles than for beech pellet particles. However, beech particles always show a finer PSD than pine particles, probably due to the different breakage mechanism of softwoods and hardwoods. Comminuting pellets produced a narrower (uniform) particle size

range compared to fine grinds and disintegrated pellets. Regarding the particles length (Figure 6B), on average, beech particles are shorter than those from pine.

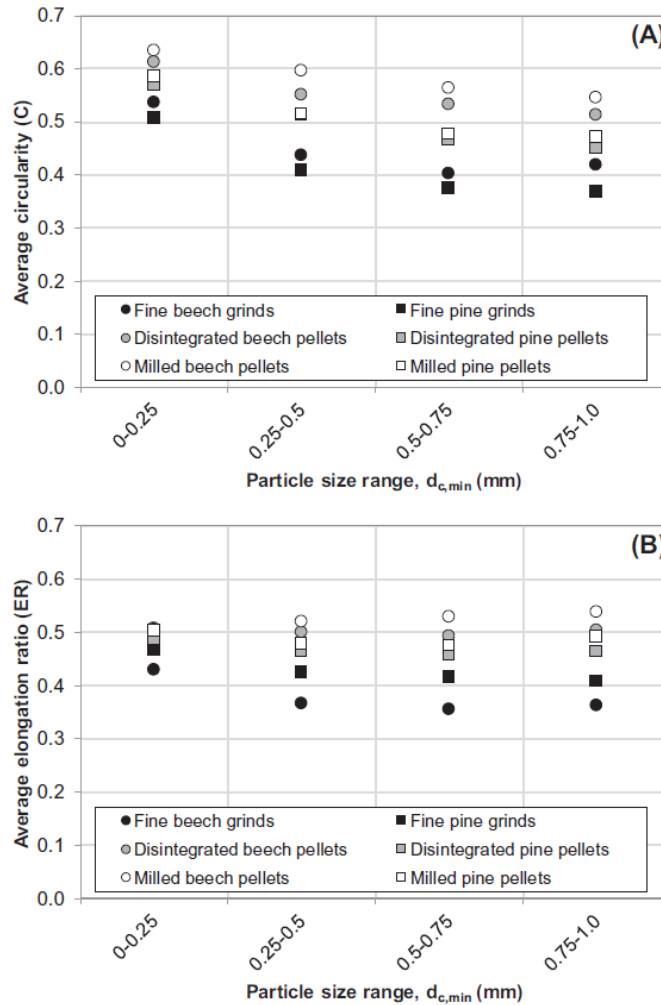


Figure 5 Average circularity (A) and elongation ratio (B) of fine grinds, disintegrated pellets, and milled pellets for beech and pine analyzed by Camsizer®X2. Values for particles wider than 1 mm were neglected due to the small number of particles analyzed.

Regarding energy consumption, for practical milling operations, beech wood requires less energy for milling than pine. However, pelletizing beech requires more energy than pine. This behavior is attributed to a lower extractives content in beech.

Table 1 Estimation of the process specific energy consumption (SEC) for milling and pelletizing prior to combustion of wood pellets based on the net calorific value of the oven dry matter (NCV_d)

Wood	NCV _d (MJ/kg, DW)	Pellet plant			Power plant SEC _{pellet milling} /NCV _d (%)	Total (%)
		SEC _{coarse milling} /NCV _d (%)	SEC _{fine milling} /NCV _d (%)	SEC _{pelletizing} /NCV _d (%)		
Beech	18.4	0.16	0.85	1.77	0.13	2.91
Pine	19.8	0.23	0.90	0.64	0.17	1.94

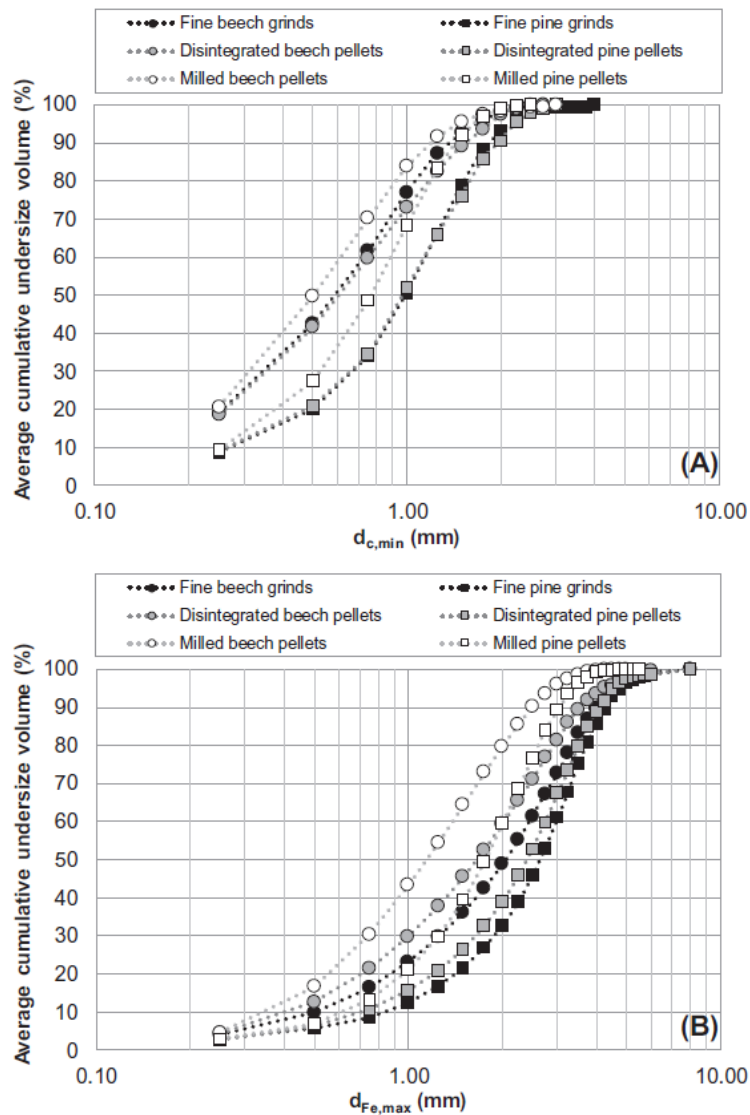


Figure 6 PSDs of milled pellets, disintegrated pellets, and fine grinds versus the particle width ($d_{c,min}$) and the particle length ($d_{Fe,max}$) determined by Camsizer®X2.

It is known that the PSD of the milled pellets affects the combustion efficiency, the unburned carbon levels in the ash and the combustion stability. The degree of particle size reduction achieved is affected by pellet properties, including chemical structure and mechanical properties (e.g., strength, durability, compressibility). One way to assess the pellet suitability for size reduction is to determine its grindability. The grindability is a measure of the pellet resistance to grinding, and as such reflects some physical pellet properties, such as strength, tenacity, and fracture.

Traditionally, existing mills are designed based on the grinding properties of coal. Several standard grindability tests have hence been developed for coal over the years: the Hardgrove Grindability Index (HGI) for ball, ring, and roller mills, the Bond Work Index (BWI) for tube and ball mills and the Hybrid Working Index (HWI) for planetary ball mills. All these tests may be used to estimate the grinding behavior of coal in industrial-scale mills. However, there is a lack of reliable standard grindability tests for lignocellulosic biomass. Recent studies [1,2] reported that the classical HGI method is insufficient for determining the grindability of non-thermally treated biomass. For this reason, in the AUWP project, a study was conducted to investigate the grindability characteristics of wood pellets of different properties in two laboratory-scale mills (disc mill and roller mill - Figure 7). The pellets used

were those from the full-scale campaign (I1 and I2) as well as those produced in the previous study (beech and pine). The objective was to address the lack of knowledge regarding wood pellet size reduction and to quantify and compare the influence of different working principles on the grindability properties of wood pellets.


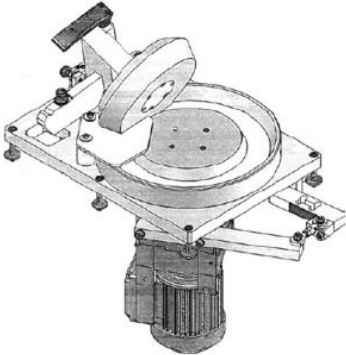
Mill	Disc mill	Roller mill
Setup		
Working principle	Compression and shearing	Compression and shearing
Motor	0.60 kW	0.12 kW
Feeder	Vibrating feeder	Dosing feeder
Wattmeter	Available	Available
Feed rate	500 ccm/min	500 ccm/min
#Repetitions	3	3

Figure 7 Laboratory mill setups and operating conditions for pellet grindability tests

In this study, the following results were obtained:

- The moisture content of wood pellets affects their durability and grindability characteristics in mills. Drying increases the pellets brittleness, which improves their grindability, resulting in energy savings and higher milled product fineness.
- Wood pellets exhibit a ductile fracture behavior during the diametral compressive strength test (Figure 8), which can provide a basic understanding of how pellets will fracture in a compression mill. The maximum compressive strength depends strongly on the internal particle size distribution of pellets (Figure 9), which hence seems to be decisive for the grinding energy effort in compression mills.

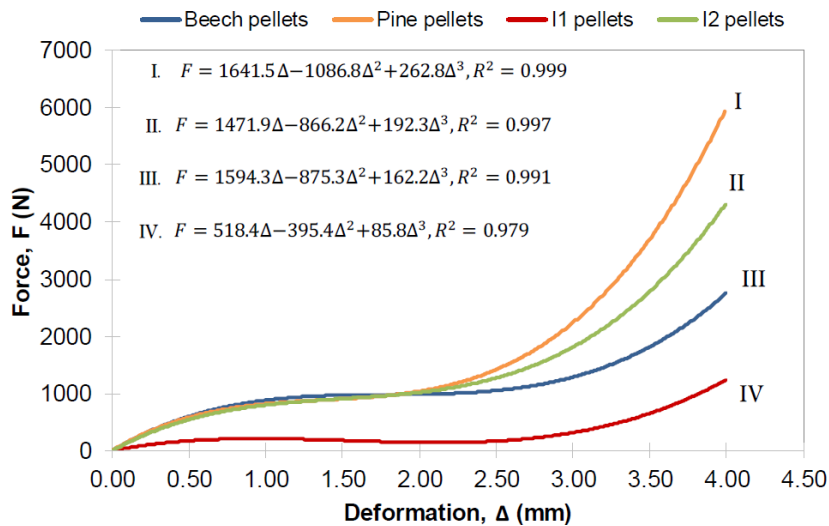


Figure 8 Typical force-deformation curve for each pellet type in diametral compression at a compression rate of 10 mm/min.

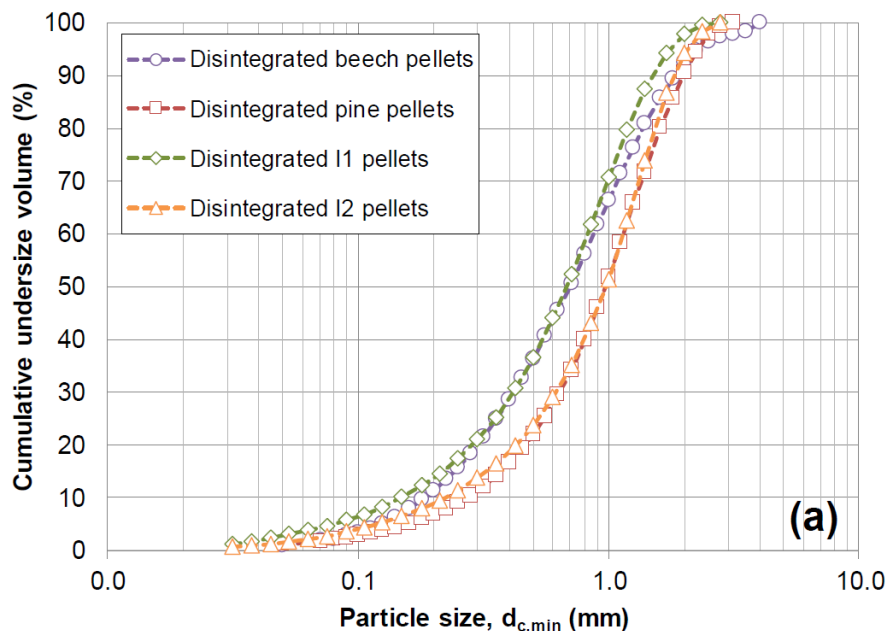


Figure 9 Internal PSD of the pellets used in the grinding study

- The shape of milled pellet particles depends mainly on the shape of the internal pellet particles, as grinding does not change the inherent wood particle shape significantly (Figure 10).
- The roller mill is less sensitive to the pellet moisture content and wood type. The size reduction degree during milling seems to be governed by the pellet properties (i.e., plant origin and internal particle size distribution). The disc mill produced a higher proportion of fine particles but required higher grinding energy than the roller mill.
- A simple continuous open-circuit lab-scale disc mill, like the one used in the present study, was suggested to determine the relative grindability of wood pellets in terms of grinding effort and milled product characteristics.

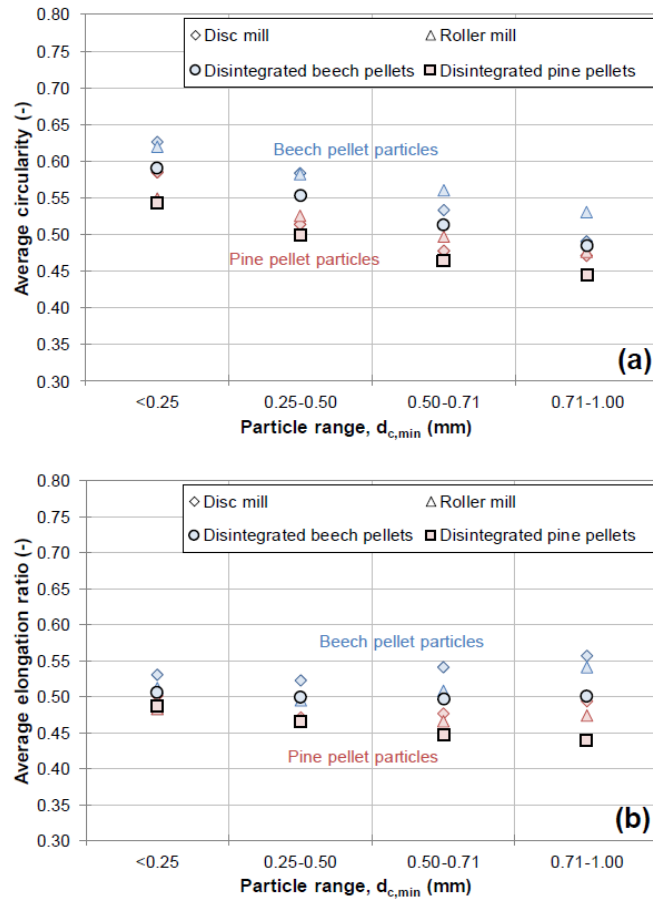


Figure 10 Average circularity (a) and elongation ratio (b) of beech and pine pellets milled in two different laboratory mills. Values for particles larger than 1.00 mm are neglected due to the small number of particles analyzed.

The results from the previous grindability study were complemented with those using the same roller mill but including a classifier (Figure 11) to separate the milled particles into two fractions: fine fraction (final product) and coarse fraction (it gets recirculated in the mill) in what it is called close-circuit milling.

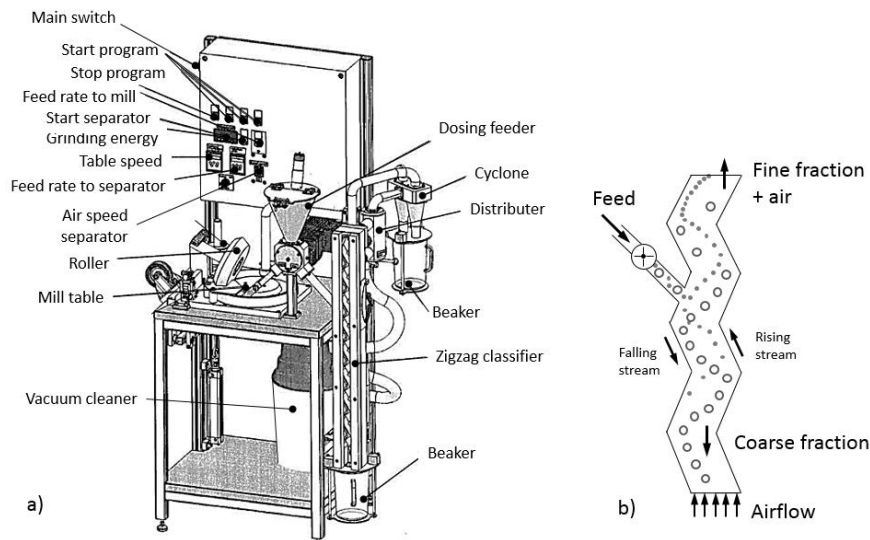


Figure 11 Schematic view of the roller mill (a) and the separation principle of the zigzag classifier (b).

The idea was to mimic the milling process in full-scale power plants. For this reason, the results from the lab-scale close-circuit milling were compared with those from the full-scale closed-circuit grinding obtained during the campaign at Amagerværket. The following conclusions were drawn from this experimental study using pine, beech, I1 and I2 pellets:

- To achieve a characteristic product particle size, beech pellets required the lowest grinding energy compared to the other three pellet samples, which have shown approximately similar grinding energies. Thus, utilizing beech pellets in the industrial roller mills has the potential to improve the grinding performance.
- The comparison of the grinding behavior of the two industrial pellet qualities in the lab-and industrial-scale mill showed that the lab-scale method requires lower grinding energies for obtaining similar size reduction ratios. However, the tendencies with respect to the effect of pellet type and size reduction ratio on grinding energy was reasonably similar. Hence, the application of the lab-scale mill can have great economic and practical motivation for power plant operators, as there is no need for costly pilot-or industrial-scale experiments.
- The comparison between open- and closed-circuit roller-milling showed that the closed-circuit operation achieves an actual size reduction effect on the internal pellet particles (i.e., higher and narrower product fineness) and lower grinding energy than the open-circuit operation.

1.4.3 Dust particle size and morphology characterization results

Different methods including sieving, 2D imaging (Camsizer), laser diffraction (Mastersizer), SEM microscopy and 3D X-ray computed tomography have been tested in the AUWP project for wood dust morphology characterization.

Sieve analysis (or mechanical screening) is the most common, simple, and cheap method to determine the PSD of powdered materials. It has been used for decades and it is also the standard method to determine the PSD of solid biofuels (e.g., sawdust). It classifies large sample quantities physically into size fractions using wire mesh sieves, which commonly have square holes. The characteristic sieve size, d_{sieving} (in mm), is then defined based on the square-hole sieves as the minimum aperture size through which each wood particle can pass. The individual size fractions are weighed. The sieve aperture size above and below the fraction governs the size range of each fraction. This information is converted into a mass-based cumulative (undersize) distribution related to the d_{sieving} . The obtained results with sieving analysis show that sieving data mainly describe the width of particles, especially for the large size fractions. However, the accuracy of particle size analysis by sieving is reduced with decreasing particle size. This could be explained by the increased tendency of particles clogging the openings of sieves with small mesh sizes due to an increased dust cohesiveness when decreasing the particle size.

Digital 2D image analysis, such as the Camsizer® system (Retsch Technology GmbH, Germany) have been recently used to characterize the morphology of biomass particles. The use of 2D images has the advantage that both size and shape information can be collected. Particles are individually detected as projected areas, digitalized, and the images processed. Based on the 2D particle image, a quantitative description of the size and shape can be given. The PSD can be represented by different particle size descriptors (e.g., Feret diameter). Compared to sieve analysis, the PSD is presented as a cumulative volume distribution.

Several tests were conducted to characterize the morphology of biomass particles using alternative methods including laser diffraction (Mastersizer), 3D X-ray computed tomography and scanning electron microscopy (SEM).

Measuring non-spherical biomass particles with laser diffraction was found to be inaccurate, as laser diffraction assumes that measured particles are spheres. Moreover, laser diffraction provides no shape information.

Visualizing biomass particles with SEM or static image analysis (e.g., optical microscope) has some drawbacks due to the time consuming analysis, particles overlapping, small sample size (low particle statistic), and the need for proper sample preparation (e.g., sputter-coating for SEM).

Particle size measurements using 3D X-ray computed tomography (Figure 12) allow the precise representation of all three particle dimensions, thus enabling calculations of the particle volume. However, the analysis of particles in bulk is very difficult, samples require proper preparation, and particle scans are very time-consuming.

Based on the results obtained, the main method for particle morphology characterization used in the present project has been the Digital 2D image analysis.



Figure 12 3D representation of wood particles by X-ray computed tomography.

The Rosin-Rammler-Bennet-Sperling (RRBS) model distribution (also called Rosin-Rammler or Weibull distribution), originally used to represent the sieve analysis results of crushed coal, has been found to fit well with the PSD of milled biomass particles. The RRBS model is a two-parameter distribution function expressed as follows:

$$R(d) = 100 - 100 \cdot e^{-\left(\frac{d}{d^*}\right)^n}$$

Where $R(d)$ is the cumulative (% , undersize) distribution of material finer than the particle size d , d^* is the characteristic particle size defined as the size at which 63.21 % of the PSD lies below, and n is the distribution parameter. A plot of $\ln[\ln[100/(100-R(d))]]$ against $\ln(d)$ on the double logarithmic scale will give a straight line of slope n , if the PSD fits the RRBS model equation. The d^* also characterizes the material fineness. The results from this project have proven that size distribution of comminuted wood pellets by sieve analysis and Camsizer® X2 (X-Jet mode) can be well fit with the Rosin-Rammler-Bennet-Sperling model.

The present project has also proven the applicability of Von Rittinger's comminution theory for wood pellets for different kinds of mill (roller mill, disc mill, hammer mill, knife mill). Von

Rittinger's comminution law analyzes the relationship between SGEC and particle size reduction according to:

$$SGEC = K_R \left(\frac{1}{d_p} - \frac{1}{d_f} \right)$$

where d_p is the 90th percentile passing size of the comminuted product and d_f is the 90th percentile passing size of the material within the pellet feed. The material characteristic parameter K_R (kWhmmt^{-1}) allows to characterize the pellet grindability by a single value.

During the determination of the internal PSD of pellets it was observed that beech pellets are difficult to disintegrate in hot water following the standard procedure ISO 17830:2016. There were agglomerates after the disintegration. Thus, it is recommended to perform the disintegration procedure twice to have individual particles and to avoid agglomerates.

1.4.4 Single particle combustion studies for densified vs raw wood cubes results

A study was carried out to investigate the combustion characteristics (devolatilization time, char conversion time, and swelling/shrinking) of raw and pelletized particles of beech and pine in a single particle combustion (SPC) reactor. The aim was to examine the influence of apparent density and pelletizing conditions on the combustion characteristics of wood. Both the raw and pelletized cubes had the same dimensions $3.2 \times 3.2 \times 3.2 \text{mm}$.

This is the first study to describe and compare the combustion behavior of raw and pelletized wood of different apparent densities at suspension-firing conditions. The following conclusions can be drawn from the experimental study:

- Increasing the pelletizing pressure has a stronger effect on the apparent wood density than increasing the pelletizing temperature (Figure 13). Pelletizing pine leads to a better pellet quality than beech indicated by stronger inter-particle bonds that lead to higher dimensional stability and higher apparent densities.

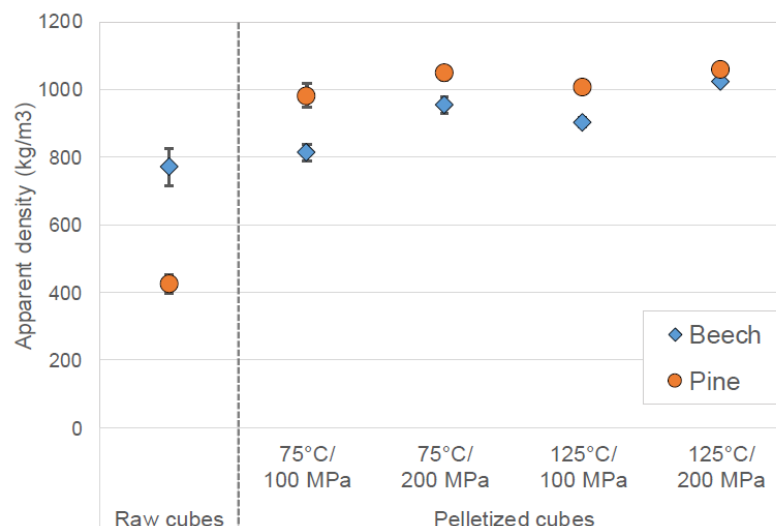


Figure 13 Apparent densities of raw wood cubes and wood cubes produced under different pelletizing conditions.

- The apparent density of raw and pelletized wood has a strong influence on the devolatilization times (Figure 14). The char combustion time is the most time-consuming step of the total particle conversion process.

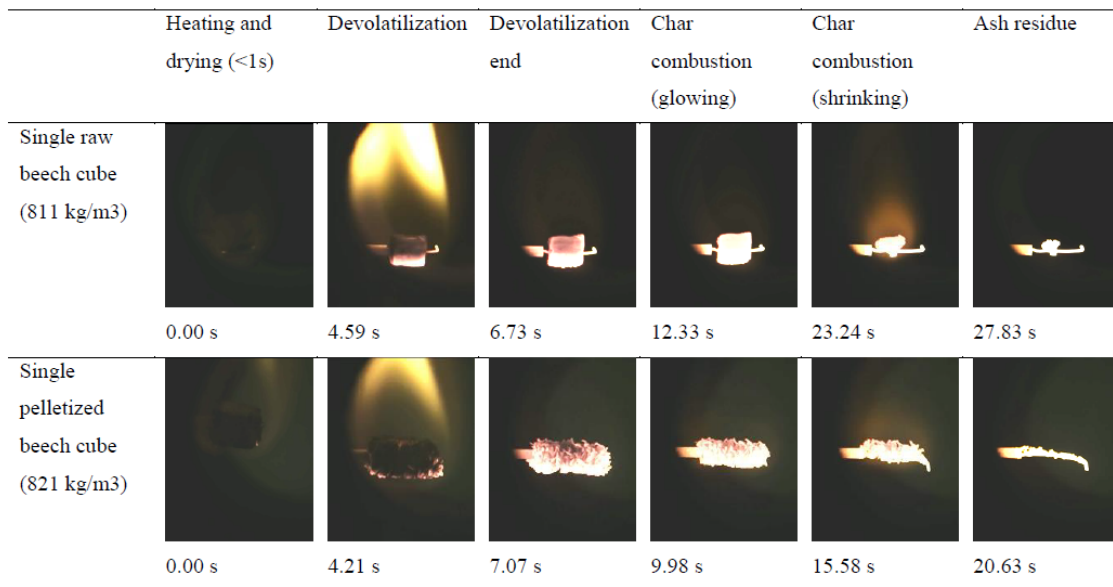


Figure 14 Total conversion process of a raw and pelletized beech cube in the SPC reactor (1262°C, 5.0 vol.% O₂, 1.5 m/s gas velocity), including their residence times.

- During devolatilization, raw wood shrinks (especially in tangential direction), while inter-particle bonds in pellets break, causing significant pellet swelling (especially in the press direction) (Figure 15).

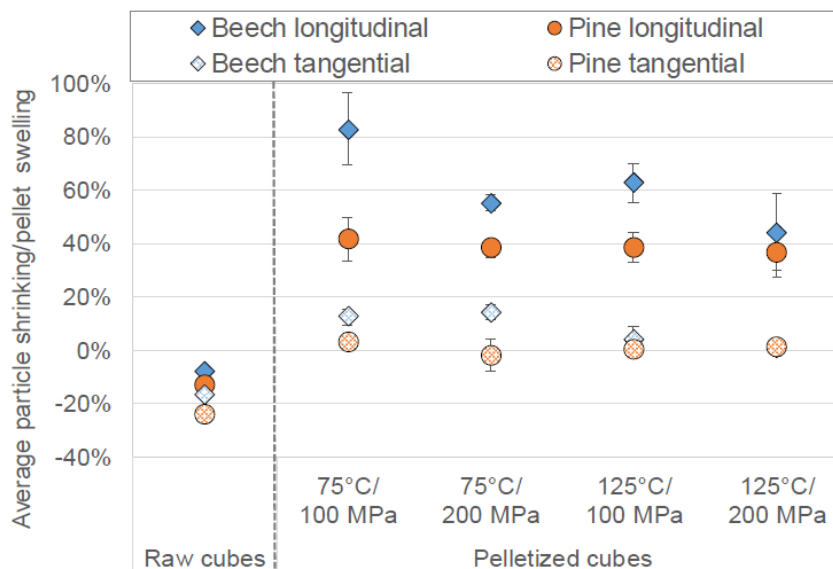


Figure 15 Average swelling of pine and beech pellets during devolatilization in the SPC reactor. Error bars indicate the first standard deviation from the mean.

- Wood pellets fragment during char combustion compared to raw wood. Pellet fragmentation was more pronounced for beech pellets indicating weaker interparticle

bonds in beech pellets. The fragmentation probably has some effect on the char combustion times.

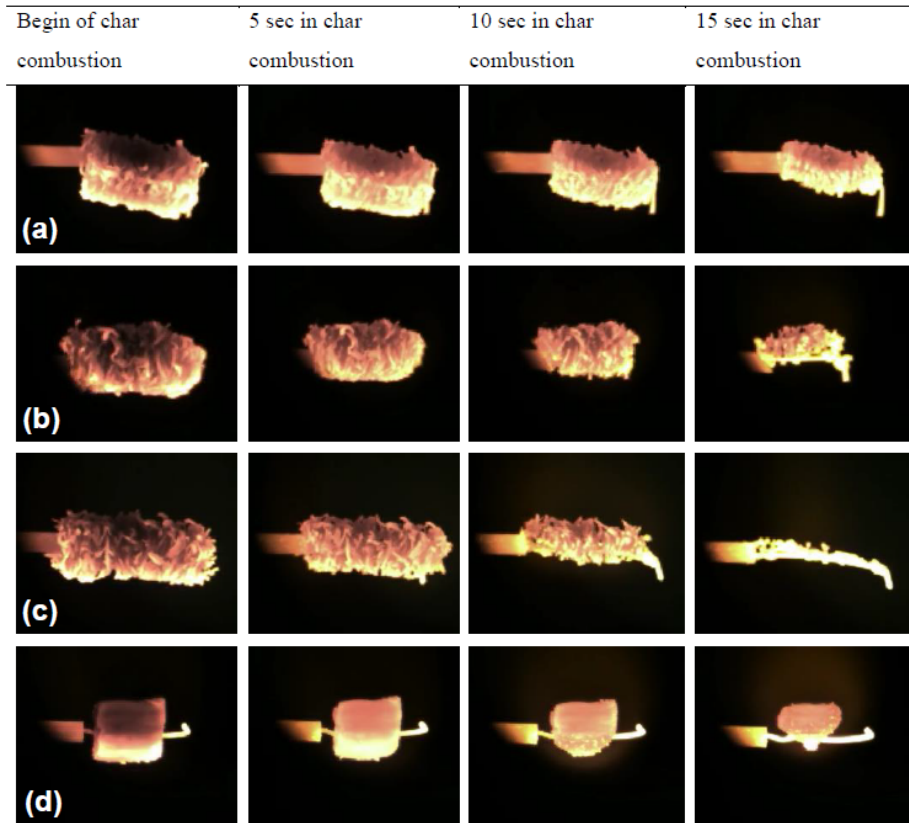


Figure 16 Char combustion process of (a) pelletized pine cube (1035 kg/m³), (b) pelletized beech cube (1030 kg/m³), (c) pelletized beech cube (821 kg/m³), and (d) raw beech cube (811 kg/m³).

1.4.5 Dissemination of the results

The results of the project have been published and widely disseminated via a PhD Thesis publicly available at DTU Orbit, a Bachelor thesis that can be sent under request, 4 conference contributions available at DTU Orbit and 4 scientific publications that will be available at DTU Orbit once the embargo periods of the journals expire. In addition, the results of the different studies have been presented during 2017 and 2018 in CHEC seminars at DTU Chemical Engineering, DTU Chemical Engineering Department Day and CHEC Annual Day.

- **PhD Thesis (1):** Masche, M. (2019) Milling and Physical Properties of Wood Pellets for Suspension-Fired Power Plants. PhD Thesis. Chemical and Biochemical Engineering Department. Technical University of Denmark (Denmark)
- **Bachelor Thesis (1):** Clausen, B.E. (2018) Combustion behavior of Raw and Pelletized Wood at Suspension-Fired Conditions. Bachelor Thesis. Chemical and Biochemical Engineering Department. Technical University of Denmark (Denmark)
- **Scientific publications (4):**
 Masche, M.; Puig-Arnabat, M.; Wadenbäck, J.; Clausen, S.; Jensen, P.A.; Ahrenfeldt, J.; Henriksen, U.B. (2018) Wood pellet milling tests in a suspension-fired power plant. *Fuel Process Technol*, 173:89–102.

Masche, M.; Puig-Arnavat, M.; Jensen, P.A.; Holm, J.K.; Clausen, S.; Ahrenfeldt, J.; Henriksen, U.B (2019) From wood chips to pellets to milled pellets: the mechanical processing pathway of Austrian pine and European beech. Powder Technology, 350: 134-145

Masche, M.; Puig-Arnavat, M.; Jensen, P.A.; Holm, J.K.; Clausen, S.; Ahrenfeldt, J.; Henriksen, U.B (2019) Grinding performance of wood pellets in laboratory-scale mills. Biomass & Bioenergy (under review)

Masche, M.; Puig-Arnavat, M.; Jensen, P.A.; Holm, J.K.; Clausen, S.; Ahrenfeldt, J.; Henriksen, U.B (2019) An investigation of the grindability of wood pellets in a lab-scale roller mill with classifier. Biomass & Bioenergy (under review)

- **Conference contributions (4):**

Masche, M.; Puig-Arnavat, M.; Holm, J.K.; Jensen, P.A.; Ahrenfeldt, J.; Clausen, S.; Henriksen, U.B. (2018) From wood chips to pellets to milled pellets: the mechanical processing pathway of Austrian pine and European beech. 2nd International Conference on Bioresource Technology for Bioenergy, 16-19 September 2018, Sitges, Spain

Masche, M.; Puig-Arnavat, M.; Holm, J.K.; Jensen, P.A.; Ahrenfeldt, J.; Clausen, S.; Henriksen, U.B. (2018) Combustion behavior of single particles of raw wood and pelletized wood. 8th Workshop on Co-firing Biomass with Coal, 11-13 September 2018 Copenhagen, Denmark - Best Poster Award

Masche, M.; Puig-Arnavat, M.; Ahrenfeldt, J.; Jensen, P.A.; Clausen, S.; Badenbäck, J.; Henriksen, U.B. (2018) Wood Pellet Milling Performance in a Suspension-Fired Power Plant. Poster presentation. 26th European Biomass Conference and Exhibition, 14-17th May 2018, Copenhagen, Denmark (Poster presentation)

Masche, M.; Puig-Arnavat, M.; Ahrenfeldt, J.; Henriksen, U.B.; Arendt Jensen, P.; Wadenbäck, J. (2017) Full-scale milling tests of wood pellets for combustion in a suspension-fired power plant boiler. Nordic Flame Days 2017. 10-11th October 2017, Stockholm, Sweden. (Oral presentation)

1.5 Utilization of project results

The objective of the project has been to support an efficient and fast conversion of pulverized coal-fired power plant boilers to operate on biomass fuels by investigating the key issues that can ensure an efficient and effective milling of biomass pellets in existing power plant coal mills. Conversion of a single generating unit of 350 MW from coal to biomass contributes a CO₂-neutral electricity production equivalent to the establishment of an offshore wind farm of 500 MW. Thus, the present project results contribute to realize the following Danish energy policy objectives:

- To facilitate a conversion from fossil fuels to biomass.
- Greater use of biomass.
- To reduce Danish net emissions of CO₂.
- To provide a reliable biomass based power supply with high electrical efficiency, high plant availability and high load adaption capability.
- To increase fuel flexibility to secure a sustainable and competitive supply of biomass pellets.

The project results support the implementation of a technology (biomass PF-firing) that within a reasonably short time scale can provide a large reduction of the Danish emissions of CO₂. The project results are being used by the two industrial partners Høfor and Ørsted to

improve the operation of their power plants. They have gained new understanding of the grindability of pellets in already existing (coal) mills and new knowledge of the milled particle morphology that helps them to ensure a more efficient grinding process and, as a result, a more efficient burning of the biomass particles. They have also learned that the pellet processing history has more influence on the morphology of comminuted pellet particles than the milling.

The methodology suggested to evaluate the pellet grindability, in lab-scale equipment, can be directly used by power plant operators to determine the behavior of wood pellets in industrial scale mills. In addition, the project has also suggested the best method to study the morphology of wood dust particles that can be applied by power plant operators.

The project has completed significant experimental research on the characterization of wood pellets and milled pellet particles with the aim to contribute to the limited experience and knowledge about the pellet grindability in lab-and industrial-scale mills. The project has been able to support the green transition from coal to biomass power plants by giving answers to the following questions:

- How can the milled wood pellet particle morphology be characterized?
- How can wood pellet grinding properties be tested? Can lab-scale mills predict the milling results obtained at power plants?
- Is it possible to relate the wood pellet properties to the pellet grinding characteristics in a mill?
- How do comminution and pelletization affect the physical properties (e.g., size, shape, and density) of wood particles?
- How does the wood species affect the grinding and combustion characteristics?
- How does pelletization influence the combustion process of wood particles?

The results obtained by the PhD student working in the project have been widely disseminated, included in four scientific manuscripts already published or under review by well-known scientific journals and in four oral presentations/posters in international conferences. The results and methodology developed within the project have also been included in two DTU courses ("Thermal gasification and sustainability" – DTU course 28271 and "Laboratory in Chemical and Biochemical Engineering" – DTU course 28231).

1.6 Project conclusion and perspective

Considerable insight has been gained with regard to the morphology changes of wood along its processing pathway from chips to pellets and to milled pellet particles. The study has highlighted that the industrial pelletization process has a larger impact on modifying the (original) pre-densified wood particle shape than the pellet milling step. Hardwood (beech) particles became rounder and less elongated along their processing pathway compared to softwood (pine) particles. This work has great value for power plant operators, as the milled pellet particle shapes obtained by roller or hammer mills are a result of the pellet processing history and origin. In view of these findings, this study represents an excellent initial step towards the understanding of the morphology changes occurring during the industrial pellet production process. This finding potentially allows pellet producers to control the shape (aerodynamic) properties of particles for suspension-firing.

The study has been able to demonstrate the different grinding behavior of pellets with a different origin. At the pellet plant, the milling of similarly sized hardwood (beech) and softwood (pine) chips has shown that pine produced coarser particles, led to a lower Von Rittinger's size reduction ratio, and required higher grinding energy. In the process of pellet formation, hence, pine pellets comprised coarser pre-densified particles compared to beech

pellets, which, milled to a specific target size, required less grinding energy than pine pellets. The pellet grinding behavior can thus be ascribed to the pellet processing history, including both feedstock origin and internal pellet particle size distribution. Therefore, pellets with similar internal pellet particle sizes and origin are likely to demonstrate a similar grinding behavior.

Promising results have been found to predict the grinding behavior of wood pellets with a different origin by laboratory testing. The proposed roller mill equipped with a zigzag classifier was aimed to simulate a continuous grinding operation similar to an industrial vertical roller mill. It was shown that the lab-scale mill is useful to assess the grinding properties (milled product fineness and grinding energy) of the different pellet qualities. Beech pellets required the lowest grinding energy to achieve a specific size reduction ratio than the other three pellet samples. The comparison with the grinding results obtained at the power plant showed similar trends with regard to the pellet type and size reduction ratio on grinding energy. Hence, the proposed lab-scale mill with classifier has a great potential to be applied as a standard method to predict the pellet grindability at industrial scale. Accurate prediction of the pellet grindability prior to the industrial-scale operation has economic and practical motivation for power plant operators, as there is no need for costly pilot-or industrial-scale experiments.

While some researchers believe that mills only break the pellets down into the pre-densified particle sizes, the study has demonstrated that mills not only break the weak interparticle bonds in pellets, but also achieve a size reduction of the particle length and width compared to the pre-densified (and disintegrated pellet) particles. The degree of particle size reduction achieved by the mill is thereby dependent upon the mill (i.e., type, conditions, circuit) and the material properties. Von Rittinger's comminution theory described well the relationship between size reduction ratio and grinding energy, suggesting that the milling of fibrous and non-brittle wood is likely dominated by the creation of new surface areas. Thus, Von Rittinger's comminution theory is a promising approach to evaluate the pellet breakage behavior during milling.

Considerable understanding regarding the grinding behavior of industrial wood pellets in the existing coal roller mills with classification equipment at the suspension fired power plant Amagerværket (Denmark) has been gained. Milling of pellets with coarser internal particle sizes resulted in higher specific grinding energy, differential mill pressure, and larger fraction of milled coarse particles. The study has underlined that the internal (disintegrated) particle size distribution is an important specification to determine the pellet grindability. Information about the pellet grindability has great value for power plant operators. Thus, utilizing pellets made of finer particles can help to maximize the mill capacity, reduce the risk of mill choking (and mill wear), and facilitate complete combustion of particles in the available boiler residence time. However, those pellets are typically more expensive to produce, resulting in additional fuel costs.

The study has also reviewed current pellet specifications and established new measurable parameters that might be incorporated to characterize the chemical, physical, and mechanical properties of wood pellets for industrial use. Among these are biopolymer analysis, number of pellets present in a given sample, and diametral compressive strength. The number of pellets had a large effect on the pellet durability. The diametral compressive strength test has shown that pellets had a non-linear (ductile) stress-strain behavior, which may predict how pellets will fracture in a compression mill (e.g., disc or roller mill).

The moisture content of pellets affects their grinding characteristics. Drying increased the pellet brittleness, milled product fineness, and reduced the grinding energy requirement. Thus, drying has the potential to maximize the mill capacity and improve the boiler efficiency. This finding highlights the importance of fuel drying, especially in biomass power plants that feature mill types with no integrated drying step, such as the disc or hammer

mill, where new drying concepts may be installed to enhance the milling performance and reduce mill wear. The additional energy input for drying may be reduced by utilizing the waste heat contained in flue gases.

The pellet disintegration method in hot water (ISO 17830:2016), developed to determine the internal pellet particle size distribution, has great practical utility for the evaluation of the size reduction ratio achieved by the mill, especially when the predensified particle size distribution is unknown. However, beech pellets were more difficult to disintegrate into individual particles than pine pellets. Higher attractive forces between the smaller beech particles may explain their tendency to form agglomerates. Adding hot water again to the dried disintegrated beech pellets resulted in a better separation of all particles. The study hence suggests to perform the procedure twice, if particle agglomerates are observed after the first disintegration.

For the characterization of the milled particle morphology, traditional sieve analysis has shown to suffer from some limitations. These include the determination of only one particle dimension (i.e., width) and lower accuracy to describe the width of fine particles due to clogging of smaller sieve openings. Dynamic image analysis has shown the great importance of determining the length and shape of wood particles and hence has the potential to replace sieve analysis in the laboratory.

Finally, the study has been able to investigate the influence of wood pelletization on the combustion behavior at suspension-firing conditions. The study has shown that the wood type and pelletizing conditions affect the pellet swelling during devolatilization and the pellet fragmentation during char combustion, as well as that the apparent pellet density influences the devolatilization time. The findings implicate that fragmentation has some effect on the char combustion times.

Annex

Annex I: Marvin Masche PhD thesis including the 4 scientific publications

Annex II: Benjamin Clausen Bachelor thesis

Annex III: Conference contributions

Annex IV: Internal report: "State of the art of biomass pellet production, biomass feedstock characteristics and pelletization conditions affecting the pellet quality"