Paper VI

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Predicting droplet size from black liquor spray characteristics

PASI MIIKKULAINEN, ARI KANKKUNEN, MIKA P. JÄRVINEN, AND CARL-JOHAN FOGELHOLM

ABSTRACT: Full-scale black liquor spraying experiments were carried out at three spraying temperatures and three mass flow rates with high solids content black liquor. We studied the spray disintegration mechanism, the length of the sheet, and estimation of drop size from the wavelength of a black liquor sheet. The flash-dominated disintegration mechanism produced a very short uniform sheet. We observed wave formation and perforation in non-flashing cases. An image-analysis-based algorithm to measure the sheet length was developed and tested. Fast Fourier transform and autocorrelation functions were tested to find the dominating wavelength of the spray to predict the final drop size.

Application: The ability to predict the final drop size from spray properties makes it possible to better control the location of the combustion processes and char-bed formation in kraft recovery boilers.

Black liquor drop size distribution and droplet shape, velocity, and trajectory play an important role in the control of combustion processes in kraft recovery boiler furnaces. These spray properties determine the combustion time and, in principle, the location where the reactions take place. Black liquor is commonly sprayed into a kraft recovery boiler furnace by a set of splashplate nozzles. Splashplate nozzles produce a thin liquid sheet, which is broken up by different mechanisms, resulting in a wide distribution of large, odd-shaped drops. The spraying practice defining the spray disintegration mechanism is, in addition to the air delivery system, the main tool for controlling the combustion process. In practice, temperature and pressure are used to adjust the spraying.

Black liquor is a highly viscous liquid with high dry solids content. The fundamental principle of the disintegration of a liquid consists of increasing the surface area of a sheet until it becomes unstable and disintegrates. Fraser and Eisenklam [1] defined three modes of sheet disintegration mechanisms, namely *rim, wave*, and *perforated-sheet* disintegration. Spielbauer and Aidun [2] found that the main types of disintegration mechanisms of black liquor sheets are wave and perforated-sheet disintegration. In both cases, ligaments are formed first and drops are formed from the ligaments.

In this paper, we focus on the disintegration of flat sheets formed on splashplate nozzles at a spraying temperature above the atmospheric boiling point. The flash-dominated disintegration, in addition to wave and perforated-sheet disintegration mechanisms, is probably the most common disintegration mechanism that takes place in the recovery boiler. It is also common for these disintegration mechanisms to appear simultaneously, which makes the spray analysis rather difficult [3, 4].

In the perforated-sheet disintegration, holes appear in the sheet. Disturbances puncture the sheet when the sheet is thin enough. The holes expand by surface tension and finally form long threads, which then disintegrate into drops [5, 6].

Wave disintegration phenomena are described by instability theories of the surface wave. It is assumed that the surface wave is a regular sinusoidal wave. In the experiments, irregular surface waves have been observed, which makes the analysis complicated [7]. The basic idea is that major wave disturbances caused by the surrounding gas disintegrate the sheet perpendicularly to the radial direction [5, 6].

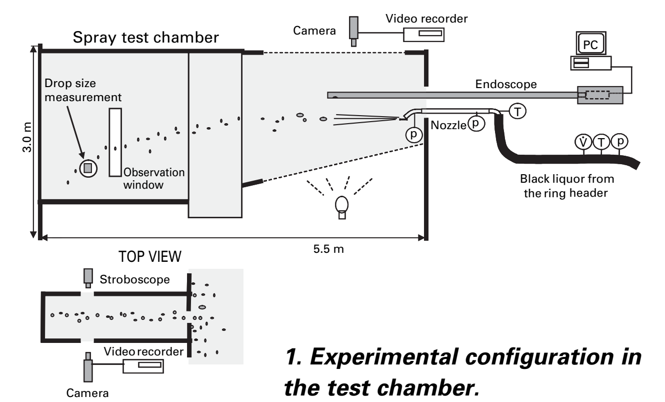
In the case of heavy flashing, a perforated sheet—or no sheet at all—is formed and the droplet formation mechanism is different compared to the non-flashing case. Atomization is accomplished by the rapid escape of gas exiting the nozzle. Large ligaments do not exist [4, 6, 8].

When the liquid sheet has disintegrated into ligaments, the internal forces of the liquid try to keep the ligaments in a stable form. Rayleigh [9] studied the disintegration of a cylindrical ligament into drops. He found that, when the wavelength of the disturbance exceeds the perimeter of a ligament, the surface tension can no longer hold the ligament together. He observed that the wavelength when a ligament breaks up was half the wavelength of the original disturbance. His model assumes that ligaments break up to equal-sized droplets. In this paper, we use this theory to calculate the size of the forming drops. The measured wavelength is used to determine the size of the forming ligament.

We have no method of measuring drop size distribution under in-furnace conditions. However, other spray properties, such as spray velocity, trajectory, and sheet length and disintegration mechanism can be measured by using a furnace endoscope. The measurements of the spray velocity and trajectory, using the furnace endoscope under in-furnace conditions, are presented by Miikkulainen *et al.* [3, 4, 10]. A large number of drop size measurements have been carried out under well-defined laboratory and spraying chamber conditions, wherein drop size distribution and other spray properties are determined [8, 11, 12]. Miikkulainen *et al.* [4] reported that the furnace environment has a negligible effect on the spray properties near the nozzle exit. This is the case for the sprays, where flashing dominates the disintegration process. If a long uniform liquid sheet is formed on the splashplate, the spray is probably sensitive to flue gas flows in a furnace, which may break up the sheet more rapidly. The heat radiation may increase the viscosity and hinder the breakup process. The objective of this paper is to study the correlation between the spray disintegration and drop size. It would be very worthwhile to be able to predict the drop size from the spray

CASE	ΔT_e, °C	$\dot{m}$, kg/s	u_s, m/s	l_S, mm	λ, mm	σ, mm	d_L, mm
1	18	4.3	14.8	362	26.1	9.8	3.9
2	18	5.2	13.3	429	32.3	11.1	4.6
3	18	6.0	13.0	447	36.7	13.5	5.2
4	17	4.3	12.1	364	32.3	13.3	4.8
5	17	5.2	12.2	398	31.1	12.6	4.9
6	17	6.1	12.0	487	*49.1*	*24.1*	*6.0*
7	14	4.3	9.9	606	44.1	14.4	4.7
8	14	5.1	9.6	624	45.4	17.9	5.3
9	14	6.1	9.7	557	40.4	15.3	5.7

I. Spraying parameters of the test cases and measured velocity (u_s), average length of the sheet at the spray centerline (l_S), wavelength (λ), standard deviation of the wave length (σ), and calculated diameter of the ligament (d_L).



1. Experimental configuration in the test chamber.

properties that can be measured under in-furnace conditions.

This paper presents a method and results of an image-analysis-based algorithm developed to analyze the black liquor spray break-up mechanism, length of a liquid sheet, and finally, the resulting drop size. The predicted drop size is compared to the measured mass median diameter obtained from the full-scale spraying experiments. The spraying experiments were carried out at a Finnish pulp mill, where high solids black liquor was sprayed into a spraying chamber. The spraying chamber was built next to a modern recovery boiler to achieve spraying conditions that were as authentic as possible. The measured mass median diameter represents the median of the total mass in particles, which is important in calculations and modeling work of combustion and particle trajectories. The predicted drop size represents one particle size, but does not give any information about the size distribution. Here, it is used to study the possibilities of predicting the drop size from other spray properties.

METHODS

Spraying experiments

The spraying experiments took place at a modern Finnish pulp mill where softwood black liquor was sprayed during the test period at a temperature of 130°C to 135°C, 14°C to 18°C above the atmospheric boiling point (excess temperature, ΔT_e). The liquor dry solids content was high; it varied between 76% and 77%. The viscosity of the black liquor was 122-160 mPas.

The three different mass flow rates ($\dot{m}$) of 4.3 kg/s, 5.2 kg/s, and 6.1 kg/s were used to observe the effect of the load on spray properties. The normal spraying practice of the recovery boiler was approximately 5.2 kg/s at 16°C above the atmospheric boiling point. Three different spraying temperatures were used to study the flashing phenomenon and its effect on the spray properties. The test cases are numbered 1 to 9 and are presented in **Table I**.

Black liquor spray properties and their effects on drop formation were studied in the horizontal spraying chamber that was built in the boiler room next to the operating recovery boiler. **Figure 1** shows a schematic of our experimental configuration. A 13 m black liquor hose led from the ring header of the boiler. The hose was well insulated. The liquor spray was illuminated beneath a plexiglass window. A full-scale commercial type nozzle was used in the tests. The nozzle exit area was partly reduced by the splashplate. The diameter of the nozzle pipe was 27 mm and the splashplate angle was 23°. To get a picture of the spray breakup mechanism, we positioned a black-and-white, high-shutter-speed CCD–camera on the roof of the spraying chamber. The main dimensions of the spraying chamber were 5.5 m x 3 m x 2 m. Process data, such as liquor volume flow rate, temperature, pressure, and dry solids content, were stored in the boiler control system database. A liquor sample was taken daily and analyzed. Temperature and pressure were also measured from the nozzle pipe. The atmospheric boiling point of the black liquor was approximately 120°C, depending mainly on the solids content [4].

We used the furnace endoscope to measure the velocity of the spray and to compare the spray properties in the test chamber and in the operating recovery boiler. This air-cooled endoscopic tube is approximately 3 m long, with a high-shutter-speed CCD-camera at one end and a prism for a right angle view at the other. We inserted the furnace endoscope into the furnace through the liquor gun hole, above the nozzle, so that the shooting distance from the endoscope lens to the liquor sheet was 30 cm. The experiments in the test chamber were similar. The velocity was measured by using the triple-exposure mode of the camera and an image-analysis system [4, 10].

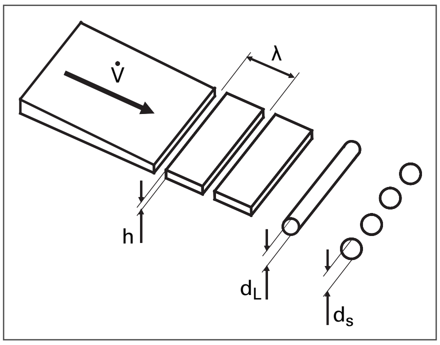
The drop size and shape were determined by a system using a video camera and image analysis. The video camera recorded the spray at a distance of 4 m from the nozzle. The drop size and shape were assumed to be final at that distance. The spray was lit by a stroboscope from the opposite side of the chamber so that the droplets could be detected without motion blur by a standard video camera (Fig. 1). The particles were detected by an image analysis system. Particles that were out of focus or in contact with the edge of the image were rejected. The volume of each accepted particle was calculated. Nonspherical particles were assumed to form spheres of equal volume [11].

Drop formation theory

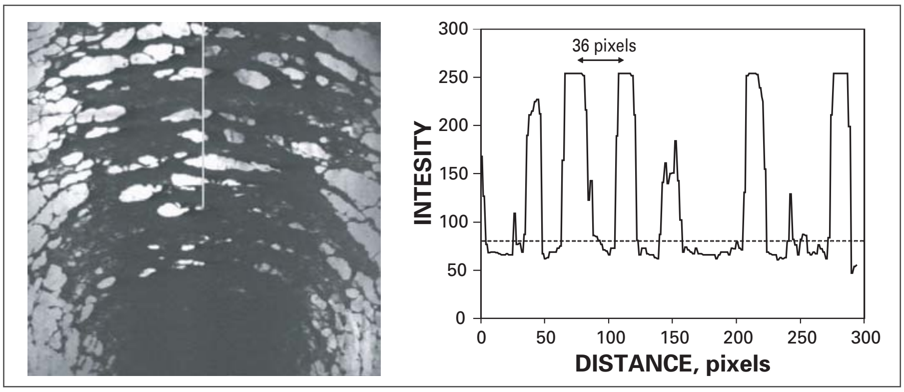
The assumption is that ligaments disintegrate into spherical drops (**Fig. 2**). The size of the forming ligament can be estimated by determining the wavelength of the disintegrating black liquor sheet. We can try to calculate the forming drop size when the wavelength and the thickness of the sheet are known. The thickness of the sheet should correlate directly to the final drop size [13].

The thickness of the sheet (h) was not measured; it had to be calculated using Eq. 1:

$$h = \frac{\dot{V}_{\mathrm{sec}}}{L_{\mathrm{sec}}\, u_{\mathrm{s}}} \tag{1}$$



2. Spray break-up model. Sheet breaks up to ligaments* (diameter, d_L)*. Ligaments break up to equal-sized* (d_s) *spherical droplets.



3. Example of a location of a given line and its intensity values.

where V_{sec} is the volume flow rate in a sector of one degree, L_{sec} is the width of the sector in a sheet breakup point, and u_s is the velocity of the sheet. We measured the mass flow rate distribution and the velocity of the spray at the spray centerline (u_s). The mass flow rate distribution was measured only for the base case $\dot{m}$ = 5.2 kg/s, ΔT_e = 17.4. We assumed the same proportion of the total mass flow rate into the sector of the centerline of the spray for all cases. The mass flow rate distribution probably changes when the pressure or temperature of the black liquor is changed.

The size of ligaments can be estimated from the wavelength of the sheet at the location of the break-up point, i.e., from the distance of holes in the sheet between forming ligaments. The size of the ligament can be calculated from Eq. 2:

$$d_L = 2\sqrt{\frac{h\lambda}{\pi}} \qquad (2)$$

where λ is the wavelength. Rayleigh's model [9] assumes that ligaments break up into equal sized drops when the area of a drop is greater than the area of the corresponding ligament. According to Rayleigh, the diameter of the droplet that forms from the ligament equals

$$d_s = ad_L \qquad (3)$$

where a = 1.89. Although the model is for fluids of low viscosity, the same idea that the collapse of a ligament produces equal sized droplets is used here to predict the drop size of black liquor. Predicted drop sizes are compared with the measured mass median drop sizes to find out the applicability of this approach with a highly viscous liquid sheet disintegrating, not only by wave-formation, but also by perforation and flashing.

Sheet length

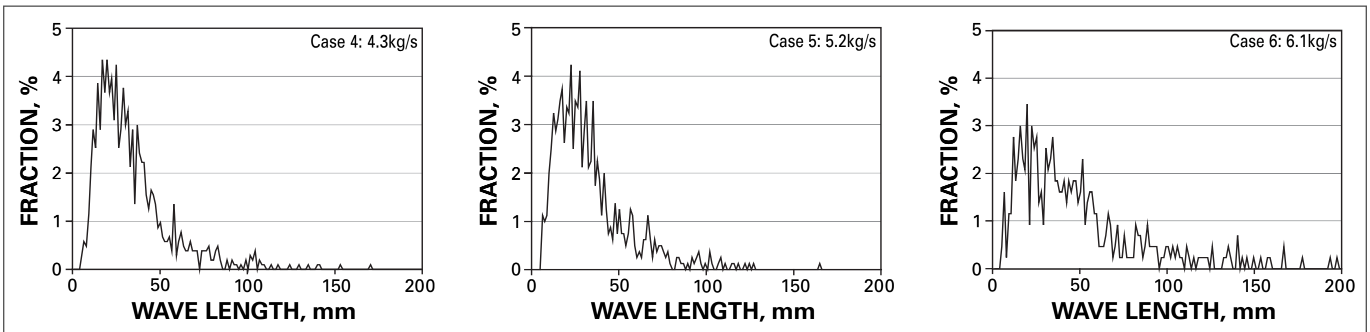
To determine the length of the sheet of black liquor spray, we developed an image-analysis-based algorithm. Using the intensity analysis functions of an image-analysis program, we collected intensity value data from frame sequences of 100 pictures (**Fig. 3**). Intensity values along a line from the nozzle exit to a specific angle from the spray centerline were plotted and the profile analyzed. Intensity values greater than 90 corresponded to holes in the sheet. We used 15 mm as the limit of the diameter of the first hole. Several limits for the first hole in a particular angle were tested; the result did not greatly change when the limit of the size was decreased. We also found 15 mm to be a reasonable value for the limit when ascertained by visual examination of video material. The intensity profiles were processed independently. When covering the range of 180 degrees at 1-degree intervals, 18,000 data points were received.

Wavelength determination

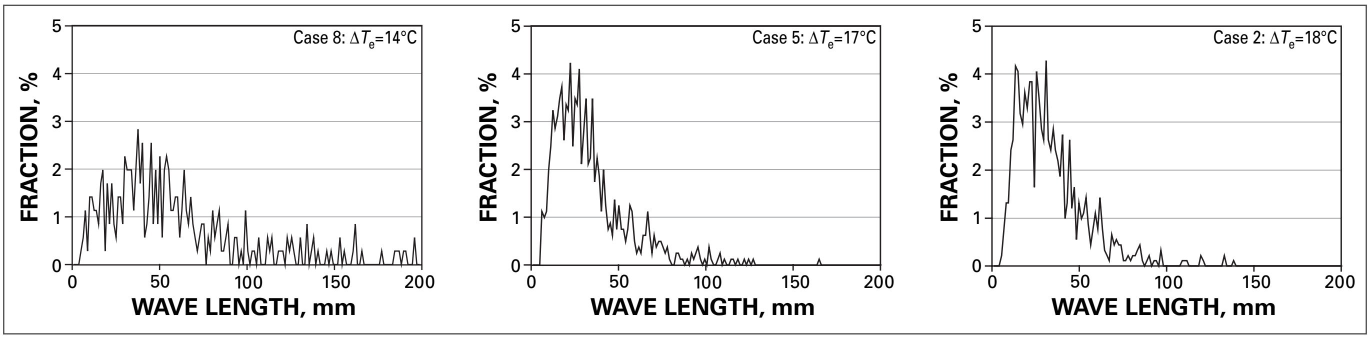
The focus was on the centerline of the spray, because of the existing drop size measurement data. In each test case, the distances between all the forming holes at the spray centerline were measured and the number fraction of each distance was counted (**Figs. 4** and **5**). The distances were measured roughly by determining the edges of the ligaments and holes and calculating the center of a hole.

If a dominant wavelength exists, it should be found by statistical or spectral methods such as an autocorrelation function or Fourier transformation. Intensity values along a given line were plotted and the profile analyzed to search for the frequency at which a possible wave formation took place (Fig. 3).

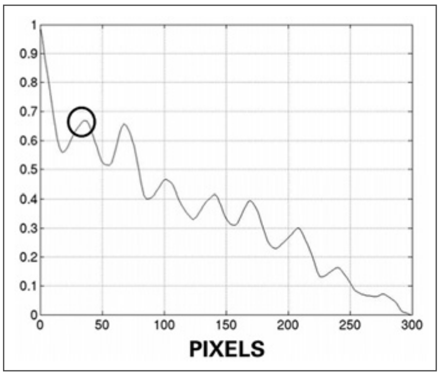
We used an autocorrelation function [14] to detect non-randomness in the data. When autocorrelation was used for



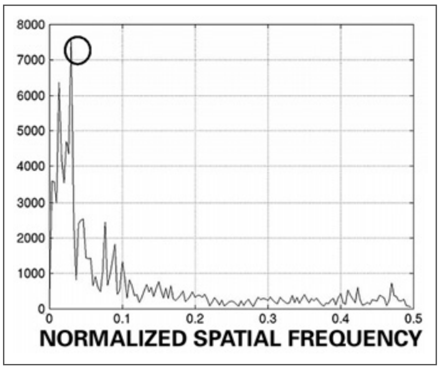
4. The effect of mass flow rate on wave distribution, ΔT_e = 17°C.



5. The effect of excess temperature on wave distribution, mass flow rate 5.2 kg/s.



6. Example of normalized autocorrelation function and the location of the first peak (circle), which is at 36 pixels.



7. FFT function and the location of the strongest peak (circle).

that purpose, only the first (lag 1) autocorrelation was of interest. The result of the autocorrelation is a series of peaks. If one dominant wavelength existed, the peaks after the first one were mainly considered as multiples of the first peak (**Fig. 6**). We did not study the intensity of the first lag, only that it existed. The standard deviation of the results were calculated instead. The idea was to systematically analyze 100 pictures from each spraying case to receive enough information about the suitability of the method. Through autocorrelation of the intensity data, we found the dominating wavelength, regardless of the disintegration mechanism of the sheet. Only the first peak (counted from the center of the autocorrelation) was used in the calculations, even though other peaks, which were not always the multiples of the first peak, existed. This means that the shortest dominating wavelength of each case was used in the drop size calculations.

Fast Fourier transform (FFT) is another common tool for determining the frequency in a noisy signal. When a test image, which contained black and white stripes, was used, FFT [15] gave a strong intensity peak. The result was easy to confirm from the original picture by measuring the distance of the center points of the white stripes. But when the same method was used for spray images, the existence of several wavelengths at the same time made the FFT difficult to use. The vertical resolution of the used images was 576 pixels. When looking for the wavelengths, only 250-330 pixels could be used because the nozzle or the solid black liquor sheet shown in the picture did not give any additional information about wavelengths. The FFT method appeared to be too inaccurate to analyze the exact wavelengths. When the location of the strongest peak of the frequency content was looked for and the result changed back to pixels, the result was the resolution divided by the location of the peak. If the wavelength is long in relation to the sample, the divisor is small (**Fig. 7**). Therefore, the resolution of the FFT method was only suitable for looking for the order of magnitude of the wavelength in the test cases.

RESULTS AND DISCUSSION

Figure 8 provides an example picture of each test case. The spraying parameters are also presented in Table I. Cases 1-6 in Fig. 8 represent the flash break-up mechanism. A very short liquid sheet is formed, which disintegrates very rapidly. The series 1-3 and 4-6 seem almost identical. Large ligaments perpendicular to the major velocity component can be observed, although the break-up mechanism can be categorized as flash breakup. When the spraying temperature is lowered by 3°C to 4°C (cases 7-9), the disintegration mechanism is totally different. A long uniform sheet appeared and broke up by perforation and wave formation. The scale in all the pictures is almost the same, but a larger area of the spray is shown in cases 7-9 as the sheet was much longer. We also saw a great difference in velocity (Table I). When flashing does not accelerate the flow, the velocity of the spray is 2-4 m/s lower.

Figures 4 and 5 show three wave distributions. They present the effect of mass flow rate and the effect of excess temperature on the wave distribution. The number distributions of "waves" are measured by counting all the distances between forming holes in the sheet at the spray centerline. The highest mass flow rate in Fig. 4 and the lowest temperature in Fig. 5 give a wide distribution of wavelengths. This indicates the incompleteness of the disintegration process. Larger ligaments disintegrate into smaller ligaments before forming droplets.

Sheet length

We measured the length of the spray at one degree intervals, from –89° to 89° on either side of the spray centerline. Those measurements gave us the shape of the unbroken sheet. **Figures 9** and **10** present the effect of the mass flow rate and of excess temperature on the average length of the sheet, respectively. Increasing the mass flow rate increases the length of the sheet when the temperature is constant. It can be observed from Fig. 8 that there is no change in the sheet break-up mechanism in cases 4-6.

The reason why the sheet lengths are different is the increasing mass flow rate through a sector of the same size.

In Fig. 10, the sheet length differs greatly between ΔT_e = 17°-18°C and ΔT_e = 13°C. The disintegration mechanism clearly changes when the temperature increases 4°C (Fig. 8; cases 1, 4, and 7). At ΔT_e = 13°C, the break-up mechanism is a mixture of perforation and wave formation and the sheet is relatively long, approximately 0.6 m. When the spraying temperature increases, the flash breakup disintegrates the sheet in the nearby area of the nozzle. The measured length of the sheet in the flashing case was 0.36 m. The average length of the sheet at the spray centerline (l_S) is presented in Table I. We used the sheet length in the drop size calculations to determine the width of the sector (L_{sec}) at the break-up point, see Eq. 1. The standard deviation of the sheet length at the spray centerline varied between 56-130 mm. In cases 7-9, when the long uniform sheet appeared, the standard deviation was highest.

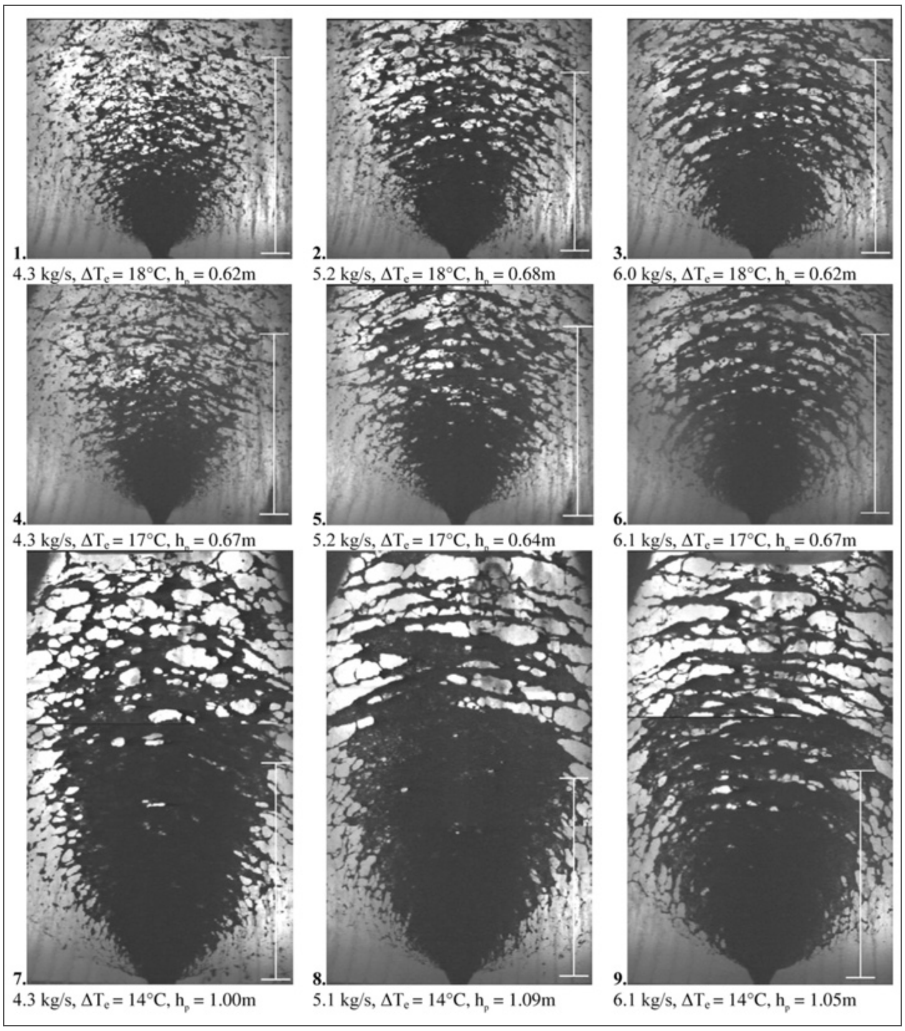
Dominating wavelength and drop size predictions

Autocorrelation was used over the intensity data and the dominating wavelength was found regardless of the disintegration mechanism of the sheet. Only the first peak (counted from the center of the autocorrelation) was used in the calculations. Other peaks existed, but they were not always multiples of the first peak (Figs. 4 and 5). This means that the shortest dominating wavelength of each case was used in the drop size calculations. A dominating wavelength was found in all the test cases, but not in all the images. In case 6 (printed in italics in Table I) we found the wavelength, but approximately 60% of the intensity profiles gave no result at all or the result had to be rejected. This indicates that a dominating wavelength does not always exist or cannot be found reliably by statistical methods. The dominating wave length was used in calculating the diameter of the ligament (d_L) to predict the drop size.

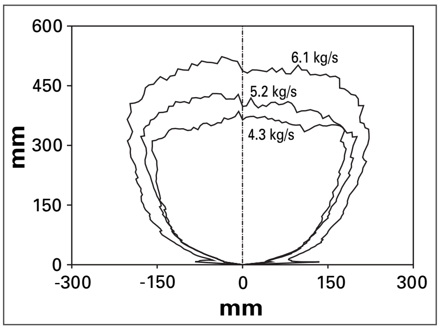
Calculated drop size is compared with the measured mass median diameter in **Fig. 11**. In cases 1-6, the drop size calculated from the wavelength theory (Eq. 3.) is larger than the corresponding measured mass median drop size. In cases 7-9, when the disintegration mechanism was not flash dominated, the calculated drop size was smaller than the measured drop size (Fig. 11). The ratio between the ligament diameter and drop size is

$$a = \frac{d_{S,measured}}{d_L} \tag{4}$$

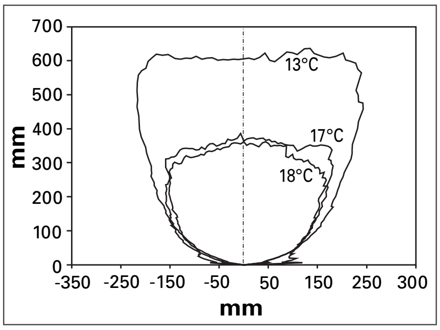
Equal drop sizes between measured and predicted were achieved with a = 2.33 in the cases where the sheet was long and flashing did not dominate the disintegration process. In the case of flashing, the correlation was not so clear, but the best



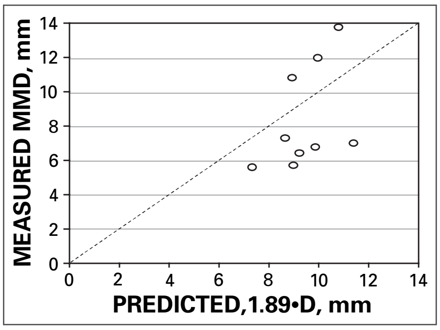
8. Example pictures of the nine spraying cases. Mass flow rates of 4.3, 5.2, and 6.1 kg/s. Excess temperatures of 18°C, 17°C, and 14°C. h_p = height of the picture. The white line represents a length of 0.5 m.



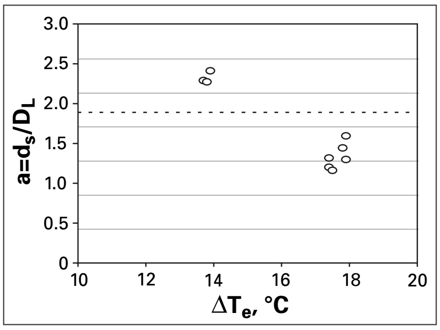
9. Effect of the mass flow rate on the average length of the sheet, ΔT_e = 17°C (cases 4, 5, and 6).



10. Effect of the spraying temperature (ΔT_e) on the average length of the sheet, $\dot{m}$ = 4.3 kg/s (cases 1, 4, and 7).



11. Predicted drop size and measured mass median diameter.



12. Coefficient a as a function of the spraying temperature.

result would be achieved with a = 1.31 (see **Fig. 12**).

SUMMARY AND CONCLUSIONS

Black liquor was sprayed through a splashplate nozzle into the spraying chamber at three mass flow rates and three spraying temperatures. The drop size distribution was measured at 4 m from the splashplate and different spray disintegration mechanisms were observed depending on the spraying parameters.

There is no method to measure the droplet size under in-furnace conditions available; that is why a method to predict the droplet size from spray characteristics near the splashplate was investigated. We searched the dominating frequency of ligaments forming from the spray centerline at the sheet break-up point to calculate the size of ligaments. The break-up point was determined by an image analysis-based algorithm, which was developed in this study. The algorithm also determines the shape of the uniform black liquor sheet. Rayleigh's model [9] of disintegration of a cylindrical ligament into drops was tested against the measured mass median diameter.

From this study, we learned the following:

- Image analysis can be used to determine the length of the black liquor sheet from a large quantity of data. The developed algorithm gives reasonable results, and can be used to scan the whole spray to determine the length of the sheet at desired intervals. When the sheet is short, the break-up mechanism is flash dominated.
- The wavelength distribution near the sheet break-up point can be used to determine the completeness of the disintegration process. If the distribution is wide, it indicates an incomplete disintegration.
- FFT and autocorrelation functions can be used to find the dominating wavelength of the sheet, but only one wavelength is probably not the case. It has been observed that several wavelengths exist simultaneously, resulting in various sizes of droplets. It was possible to receive a dominating wavelength even from the flashing cases, which means that disintegration mechanisms can exist simultaneously.
- In the flashing case, the drop size similar to the results from the drop size measurements would be achieved by multiplying the diameter of the ligament by 1.3, and 2.3 in the non-flashing case.

Our future work in this field focuses on analysis of the furnace endoscope data to study the applicability of the drop size predictions to furnace conditions. **TJ**

NOMENCLATURE

a = Coefficient, $d_{S,measured}/D_L$
d_L = Diameter of a ligament, m
d_S = Drop size, m
$d_{S,measured}$ = Measured drop size, mass median diameter, m
h_p = Vertical size of the picture, m
h = Sheet thickness, m
l_S = Average length of the sheet at the spray centerline, m
L_{sec} = Width of the sector at a sheet breakup point, m
$\dot{m}$ = Mass flow rate, kg/s
u_s = Velocity of the spray at the spray centerline, m/s
V_{sec} = Volume flow rate in a sector of one degree, m³/s
ΔT_e = Excess temperature = Spraying temperature – boiling temperature
λ = Wavelength, m
σ = Standard deviation of the wave length, m

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INSIGHTS FROM THE AUTHORS

For this study, we wanted to see if statistical methods are applicable with image analysis to predict the black liquor droplet size from the spray characteristics. If the prediction is possible (reliably enough), it would be an advantage to spraying models and adjustment of recovery boilers.

Black liquor drop formation has been studied for about 15 years at Helsinki University of Technology. Lots of experiments were carried out during that period. Connections between the spray properties and the final drop size have been sought. This study was our first step in trying to find that connection through a sheet disintegration mechanism and image processing technique when large amounts of data were processed.

In experimental work (full-scale and black liquor) it is always most difficult to carry out the experiments so that there are no open questions left during the data processing stage. We were surprised that it was possible to find the dominating wave length from the large amount of data. It is also interesting that wave length distributions give such a good correlation with the spraying parameters and disintegration mechanisms.

Drop size measurements under in-furnace conditions have not been available so far, but it is possible to measure the other required spray characteristics in an operating recovery boiler. To be able to predict the final drop size from spray properties makes it possible to better control the location of the combustion processes and char-bed formation in kraft recovery boilers.

Similar spray data is available from the in-furnace measurements carried out with a furnace endoscope. Our future work in this field focuses on the applicability of the drop size predictions to furnace conditions.

Miikkulainen, Kankkunen, Järvinen, and Fogelholm are with Helsinki University of Technology, Laboratory of Energy Engineering and Environmental Protection, P.O. Box 4400, FIN-02015 HUT, Finland. Email Miikkulainen at pasi.miikkulainen@hut.fi.

Miikkulainen **Kankkunen** **Järvinen** **Fogelholm**