THE CHARACTERISTICS OF BLACK LIQUOR SPRAYS

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ABSTRACT

Knowledge of the characteristics of black liquor sprays is essential when the performance and location of the combustion process in a recovery boiler are considered. Drop size distribution and velocity are the most important properties of a spray. They define combustion time and essentially the location where the reactions take place in the furnace. The drop size is defined by black liquor sheet properties such as velocity, thickness, length and the breakup mechanism.

The spray properties mentioned above have been studied in a test chamber and in a furnace. It was observed that small changes in operating parameters could cause dramatic changes in the spray properties. Models for correlating velocity of a spray and drop size distribution were developed and their range of application was studied.

INTRODUCTION

Splashplate nozzles produce a thin liquid sheet which rapidly breaks up, resulting in the formation of rather large drops with a wide size distribution. The drop size and size distribution are both of great importance for controlling the combustion process in the furnace. They define combustion time and thereby also the location in the furnace where the drying, devolatilization and char burnout take place.

Liquor dry solids content, temperature, feed rate and the nozzle geometry are the main operating parameters that determine spray properties. As an example, there are three different spraying situations presented in Figure 1, where only excess temperature, $\Delta T_b$ (defined as the temperature difference between spraying temperature and atmospheric boiling point of the black liquor) has been changed. The spray disintegration mechanism changes from wave disintegration to perforation and finally flashing takes place.
This study presents the results of experiments used for finding correlations between operating parameters and spray properties. The focus is on the spray disintegration mechanism, velocity and drop size distribution. The majority of the experiments were carried out at a modern Finnish recovery boiler within about its normal operational range. Spray characteristics were measured with a furnace endoscope in the furnace environment as well as in a test chamber. The spraying temperature was well above the boiling point of the liquor which in some cases resulted in heavy flashing. Drop size measurements were carried out only in the test chamber.

Optimal spraying parameters can be chosen only if the drop sizes are known with sufficient accuracy. As it is not possible to measure drop size distribution on-line in a furnace, there is a need for a set of probability density functions, which can be adapted for estimating the drop size distribution for varying spraying parameters /2/. Spray formation was observed to be similar in the spraying chamber and furnace and therefore the results from spraying chamber are considered applicable to furnace conditions /3/.

**SPRAYING EXPERIMENTS**

The spraying experiments took place at a modern Finnish pulp mill where softwood liquor was sprayed at a temperature of 129 to 135°C, which was 13 to 19°C (excess temperature, $\Delta T_b$) above the atmospheric boiling point. The liquor dry solids content was relatively high as it varied between 75 and 79%. Three different mass flow rates of 4.3 kg/s, 5.2 kg/s and 6.1 kg/s were used to observe the effect of the load. A mass flow of 5.2 kg/s at 16°C above the atmospheric boiling point approximates normal operating conditions. Two different types of commercial splashplate nozzles were used in the tests - see Figure 2: Nozzle A, where the nozzle exit area is partly reduced by the splashplate and nozzle B, where the splashplate is attached to a nozzle tube with a jacket whose inner diameter is larger than the nozzle tube exit. Black liquor spray properties and their effect on drop formation were studied both in the horizontal spraying test chamber that was built in the boiler room right next to the operating recovery boiler and in the furnace as to ensure the applicability of the results.
A schematic experimental configuration is shown in Figure 3. The main dimensions of the spraying chamber itself were 5.5 m x 3 m x 2 m. The facility for the liquor gun and endoscope insertion extended the total length to 10 m. The endoscope was located at 0.3 to 0.5 m above the splashplate nozzle, which was positioned horizontally to ensure a close similarity to spraying in a furnace. The liquor spray was illuminated from beneath through a plexiglass window. In the furnace, the necessary backlight came from the char bed. For making overall pictures of the spray break up mechanism a black and white high shutter-speed camera was located in the roof structure of the spraying chamber. Temperature and pressure were measured also, directly from the nozzle pipe. /3/

The furnace endoscope was used for comparison of the spray properties, such as the velocity at the spray centerline and the opening angle, from two different spraying environments. This air-cooled endoscope tube is approximately 3 m long with a high shutter-speed CCD –camera at one end of the tube and a prism to get a right-angle view at the other. The furnace endoscope was put into the furnace using the same liquor gun entrance hole (i.e. immediately above the liquor gun) so that the distance from the endoscope lens to the liquor sheet was about 30 cm. The velocity was measured by using the triple-exposure mode of the camera and an image-analysis system. In addition, the shape and the length of the black liquor sheet were measured and the sheet break-up mechanism was determined.

The drop size and shape was determined by a combination of a video camera and image analysis. A video camera was used to record pictures from the spray at a distance of 4 meters from the nozzle. The drop size and shape were assumed to be final at that distance. Spray separation baffles restricted the spray width so that a narrow undisturbed part of the spray could reach the drop size measurement chamber. The spray was lit by a stroboscope from the opposite side of the chamber, so the droplets could be detected by a standard video camera without any motion blur - see Figure 3. In the study, 1500 picture frames were studied for each case /4/. Most of the drops observed were not spherical /5/. Only the particles that were completely inside the focus area were accepted for analysis. The volume of each accepted particle was calculated. Non-spherical particles were assumed to form spheres of equal volume. Particles were categorized into size classes with a width of 1 mm, and a least sum of squares method was adapted to compare measured and calculated volume fractions in the size classes.
RESULTS AND DISCUSSION

By comparing the spray properties measured in the furnace with the ones measured in the test chamber it was possible to see how the furnace environment affected a liquor sheet and its break-up mechanisms. It is not possible to measure drop size and size distribution directly in the furnace and therefore those were measured, almost at the same time, in the test chamber. Spray formation was observed to be similar in the spraying chamber and furnace and therefore the results from the spraying chamber are applicable to the furnace conditions.3/3/

Disintegration and velocity

It was observed that spraying temperature is one of the most important parameters that affect the spray characteristics. Typically, in a modern recovery boiler the liquor is sprayed at temperatures well above the atmospheric boiling point. At the mill where the experiments were made the normal excess temperature was 16 °C. At this temperature, no actual liquid sheet is formed since flashing takes place inside the nozzle tube and breaks up the continuous liquid phase. Figure 4 presents the spray disintegration mechanisms at three excess temperatures as a function of mass flow rate. At a constant excess temperature the increasing mass flow rate results in a longer uniform liquid sheet before the sheet disintegrates by perforation or flashing. Examples of remarkably changing disintegration mechanisms can be observed for both nozzle types. Even a minor change in spraying temperature, especially in the latter case, can cause a dramatic change in sheet break up.
mechanism. At a 2°C higher excess temperature, flashing breaks up the liquid sheet rapidly on the splashplate and no uniform liquid sheet is formed. For nozzle B, the uniform and long liquor sheet was unexpected. The splashplate is attached to a nozzle tube with a jacket, the inner diameter of which is larger than that of the nozzle tube exit. It may be assumed that with an excess temperature as high as 14 °C, flashing takes place in the liquor flow immediately after the nozzle tube exit before the flow hits the splashplate and no sheet will form.\(^6\)

The velocity at centerline of the spray varied in the range of 9.7 to 14.5 m/s for nozzle A, and 10.2 to 15.3 m/s for nozzle B. For both nozzles an increasing excess temperature increases the velocity of the spray, but the effect of mass flow rate is not clear. Increasing the mass flow rate diminishes the effect of excess temperature. The spray velocity is highly dependent on the spraying temperature and pressure: at a higher temperature, flashing produces water vapor with a large specific volume, which accelerates the flow. At lower pressure i.e. at lower mass flow rate, flashing takes place more easily. The dimensionless velocity (ratio of measured velocity at the centerline of the black liquor sheet and the velocity of the non-flashing case at the smallest cross-sectional area of the nozzle with the same mass flow rate) can be used to describe this phenomenon:

\[
u^* = \frac{u_s}{u_p} = \frac{u_s}{m \rho_{BL} A}
\]

where \(u_s\) is the velocity measured at the centerline of the black liquor sheet and \(u_p\) is the velocity of the non-flashing case at the smallest cross-sectional area, \(A\), of the nozzle at the same mass flow rate, \(m\).\(^7\)

Increasing the mass flow rate decreases the effect of flashing inside the nozzle tube. The smaller the mass flow rate or the higher the excess temperature, the higher the dimensionless velocity - see Figure 5. Furnace environment appears to have a negligible effect on the spray velocity in the cases studied here. For different cases the dimensionless velocity can be connected to a corresponding sheet disintegration mechanism. When dimensionless velocity is low, flashing doesn’t dominate the sheet disintegration mechanism and an unexpectedly long, uniform liquid sheet is formed.\(^6\)

**Drop size and shape**

The disintegration mechanisms can be related to the resulting drop size. The median drop size was affected mostly by excess temperature. An increase of excess temperature by only 2 °C (i.e. from 14 to 16 °C) decreased median drop size by approximately 50% for the case of nozzle B. Note also the very large change in spray appearance. Figure 6 gives pie charts below each of the spray pictures: the white area in the chart presents the fraction of spherical particles detected. The ratio of spherical and non-spherical particles and the method to measure sphericity are reported in \(^5\). In Figure 6, all
Figure 4. *The effect of excess temperature and mass flow rate on sheet disintegration, (height of a single picture is approximately 0.7 m)* /6/.

Figure 5. *Dimensionless velocity as a function of the excess temperature. The open and closed symbols represent the test chamber and furnace respectively* /6/.

Figure 6. *Drop size as a function of excess temperature. The white area in the pie charts presents the fraction of spherical drops* /6/.
particles, droplets and non-spherical particles are assumed to eventually form spherical particles. The resulting mass median diameter is larger than the mass median diameter of single spherical droplets, or than the equivalent smaller mean diameter of non-spherical particles for the corresponding case. This is a natural consequence of the transformation of the volume of non-spherical particles into equivalent spherical particles. Either a decreasing temperature or an increasing mass flow rate increased the mass median diameter. /2/ In the case of nozzle B the fraction of spherical particles increases noticeably when sheet disintegration changes to the flash break-up dominated mode. In the case of nozzle A, this change in sheet disintegration mechanism is not as sharp but the mass median diameter of drops decreases as excess temperature increases.

The most important observations of this study were the large median drop sizes and the large fraction of non-spherical particles in the spray. These are essential properties of high solids content black liquor sprays and must be taken into account when, for example, making boiler calculations with CFD - software. The volume fraction of non-spherical particles varied from 60 to 85%. Under normal spraying practice conditions as used by boiler operators the fraction of non-spherical particles was around 70%.

Median drop size was affected most by excess temperature as presented above in Figure 6. Increasing the mass flow rated increased median drop size slightly for the type A nozzle, and neither was the median drop size for nozzle B affected by mass flow rate, see Figures 7 and 9. The fraction of non-spherical particles was high, between 60 and 90 vol-%.

![Figure 7. The effect of mass flow rate on median drop size, nozzles A and B. /2/](image)

**Drop size distribution**

Although the spray consisted of spherical and non-spherical particles, it is assumed here that non-spherical particles would eventually form spherical droplets. Rosin-Rammler, normal distribution, square-root normal distribution and log-normal distribution were fitted to experimental data. All four distribution functions fit the experimental data quite well, especially for the mean part of the distribution. The least square method produced best results most often for the square-root normal distribution function - see Appendix 1. The log-normal distribution function produced the next best fit. The normal distribution and the
Rosin-Rammler distribution overestimated the fraction of small particles. An example for normal spraying condition is presented in Figure 8.

**Figure 8.** Experimental particle size distributions and four size distribution curves for nozzles A and B. /2/

**Figure 9.** The effect of mass flow rate on the calculated drop size distributions for nozzles A and B. /2/
The drop size distribution curves can be used, for example, when studying the quality of the spray from the viewpoint of avoiding boiler operation problems. The fraction of small particles, which can cause fouling of heat transfer surfaces and the fraction of large particles, which can cool down the char bed are both of great concern. The median diameter varied from 5 to 11.6 mm, when the square-root normal distribution was used. The volume median diameters detected here were remarkably higher than those found earlier for lower solids content black liquor./8/

CORRELATION MODELS FOR SPRAY VELOCITY AND DROP SIZE

A correlation model for the spray velocity (at the centerline of the spray) was developed. The dimensionless velocity equation presented below consists of three elements, being critical velocity, \( u_c^* \), excess temperature, \( \Delta T_b \) and mass flux, \( \dot{m}^* \). Those elements were found to be reasonable characters for spray velocity and could be fitted to experimental data by two constants, \( a \) and \( b \). These constants differ for different liquor types and nozzle types, and have to be experimentally determined.

\[
\frac{u^*}{u_c^*} = 1 + \left( \frac{\Delta T_b}{\Delta T_{bc}} \right) \frac{a}{\dot{m}^*_{pb}}
\]  

(2)

Figure 10. Experimental data of dimensionless velocity compared to the correlation model

The critical velocity is the velocity for the non-flashing case and the temperature difference (\( \Delta T_b - \Delta T_{bc} \)) between excess temperature and critical excess temperature has to equal to or larger than zero. Critical excess temperature is typically 7 - 8 °C./9/

One observation from the spraying experiments was that the drop size correlates very well with spray velocity. The correlation model for mass median diameter (MMD) consists of two elements, being excess temperature and dimensionless velocity. The constants, \( c \), \( d \) and \( e \), again differ for different liquors and nozzle types. MMD can be expressed as

\[
MMD = c \Delta T_b^d \, u_{\Delta T}^e
\]  

(3)
Figure 11. Experimental data of mass median diameter compared to the correlation model

These correlation models can be reliably used in the operational regions near the temperature and dry solids ranges of the experiments. The experiments were carried out within the widened operational range of the mill. The lowest experimental spraying temperature was achieved by cooling the long black liquor pipe from the ring header. A minor decrease of excess temperature (only by 2 °C, i.e. from 16 to 14 °C) for the case of nozzle B increased median drop size by approximately 50%. There was also a huge change in the spray disintegration. The fraction of non-spherical droplets increased also rapidly. Taking also the viscosity of liquor into account may widen the range of application of the correlation models.

FUTURE WORK

The application range of the developed empirical correlation models for velocity and drop size is limited. In order to broaden the application range, physical or semi-physical models are required. The experimental work is irreplaceable for model validation. Liquor specific characteristics and their effect on atomization performance must be verified. The effect of nozzle geometry must not be forgotten.

The relation between spray disintegration and resulting drop size is obvious and it should be studied further. When viscosity of black liquor is high and flash break-up mode of atomization process does not occur, the portion of non-spherical particles is high. Different trajectories and the differing combustion behavior of such particles must be taken into account.

CONCLUSIONS

In their normal range of operation modern kraft recovery boilers implies that flash-breakup is the dominating spray break up mechanism. This operational range is, however, quite narrow. When in the experiments the firing temperature was lowered to 2°C below the operational temperature range of a recovery boiler, an unexpectedly long, uniform liquor
A very important observation of this study was the large median drop size and the high fraction of non-spherical particles in the spray. These are essential properties of high solids content black liquor sprays and should be taken into account when, for example, designing or modeling kraft recovery boilers. The volume fraction of non-spherical particles varied between 60 and 85%. Under normal spraying practice conditions as used by boiler operators the fraction of non-spherical particles was around 70%.

The studied distribution functions fit the experimental data quite well. The square-root normal distribution function gave the best result when assumed that non-spherical particles would eventually form spherical droplets.

The developed correlation models of spray velocity and mass median diameter of drops can be reliably used near the ranges of temperature and dry solids covered by the experiments presented here. Taking the viscosity of liquor into account the application range of the correlation models can be extended, but reliable physical models for this are still needed. Different trajectories and the differing combustion behavior of non-spherical particles must be taken into account.

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Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>The smallest cross-sectional area of the nozzle, m$^2$</td>
</tr>
<tr>
<td>$D$</td>
<td>Drop diameter, mm</td>
</tr>
<tr>
<td>$d_p$</td>
<td>Diameter of a nozzle pipe, mm</td>
</tr>
<tr>
<td>$DT$</td>
<td>Excess temperature, °C</td>
</tr>
<tr>
<td>$dTb$</td>
<td>Excess temperature, °C</td>
</tr>
<tr>
<td>MMD</td>
<td>Mass median diameter, mm</td>
</tr>
<tr>
<td>$m$</td>
<td>Mass flow rate, kg/s</td>
</tr>
<tr>
<td>$m^*$</td>
<td>Mass flux, kg/m$^2$ s</td>
</tr>
<tr>
<td>$p$</td>
<td>Pressure, bar</td>
</tr>
<tr>
<td>$q$</td>
<td>Differential distribution function</td>
</tr>
<tr>
<td>$s$</td>
<td>Standard deviation</td>
</tr>
<tr>
<td>$s^*$</td>
<td>Normalized standard deviation</td>
</tr>
<tr>
<td>$u^*$</td>
<td>Dimensionless velocity</td>
</tr>
<tr>
<td>$u_p$</td>
<td>Velocity of the non-flashing pipe flow at the smallest cross-sectional area, m/s</td>
</tr>
<tr>
<td>$u_s$</td>
<td>Measured velocity at the centerline of the black liquor spray, m/s</td>
</tr>
<tr>
<td>$\Delta T_b$</td>
<td>Excess temperature, °C</td>
</tr>
<tr>
<td>$\rho_{BL}$</td>
<td>Density of black liquor, kg/m$^3$</td>
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REFERENCES

APPENDIX 1

Square-root normal distribution /2/

The volume distribution of square-root normal distribution function can be expressed as

\[ q_{\text{sqrt}}(D) = \frac{1}{2s\sqrt{2\pi}} D^{-0.5} e^{-\left(\frac{\sqrt{D-V_{0.5}}}{s}\right)^2} \]  

(4)

where \( s \) is the standard deviation of \( \sqrt{D} \). Simmons reviewed in /10/ a very large quantity of experimental data using jet engine nozzles. He concluded that the parameter \( s \) is related to the volume median diameter of the spray by the following equation for normalized standard deviation

\[ s^* = \frac{s}{\sqrt{D_{V0.5}}} = 0.24 \]  

(5)

This means that the shape and the normalized width of the square-root normal distribution are equal for all drop sizes. This observation was later adopted to black liquor spraying. Adams et al. in /11/ and Empie et al. in /12/ obtained similar results based on experiments with a splashplate nozzle. Adams et al. found that the normalized standard deviation \( s^* \) increases up to 0.32 with increasing velocity and Empie et al. in /9/ even measured a value of 0.38. In spite of this variation the value of \( s^* \) is commonly assumed to be constant at 0.24. Recently Loebker and Empie /13/ measured slightly higher values of 0.25-0.29 for \( s^* \).