Institute of Paper Science and Technology
Atlanta, Georgia

IPST TECHNICAL PAPER SERIES

NUMBER 468

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FEBRUARY 1993

Effect of Temperature on Black Liquor Droplet Formation from Commercial Spray Nozzles

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Submitted to
AIChE Symposium Series

Originally Presented at
1992 AIChE Annual Meeting
Forest Products Division
November 6, 1992
Miami Beach, Florida

ABSTRACT

The spraying characteristics of black liquor nozzles are important to the operation of the kraft recovery boiler. Ultimately, to optimize the combustion process, control of the black liquor drop size distribution needs to be established. The initial droplet diameter has a direct impact on the condition of the char bed and the amount of carryover. Droplet size distributions from commercial nozzles have been measured for several mill liquors sprayed at typical operating conditions. The effect of temperature on drop size has been quantified, showing a weak dependence on temperatures up to the boiling point. At a temperature a few degrees above the boiling point, there is a step change in the mean droplet size which is related to flashing. The mechanism for this demands that several degrees of superheat are required for flashing to occur, resulting in smaller-than-expected droplet diameters. The temperature is sharply defined, implying that it represents an unstable point for droplet formation. Since black liquor firing temperatures in today's mills have been steadily increasing as nozzle solids levels climb, the observed phenomenon is important to be aware of so that carryover difficulties might be minimized.

INTRODUCTION

The most important unit operation employed in the recovery cycle of the kraft pulping process is combustion of black liquor in the recovery boiler. This step is initiated by spraying the concentrated liquor through one of several types of commercial nozzles, the most common being the splashplate, the V-jet, and the swirl cone. The liquor issues from the nozzle as a thin sheet, which subsequently breaks up into droplets. These droplets then go through the sequential processes of drying, pyrolysis and gasification, combustion, reduction, and smelt coalescence (1). The rates at which these physical and chemical processes occur are highly dependent upon the size and size distribution of the droplets formed from the spray. The smaller the droplet, the greater the surface area per unit mass of liquor and hence, the greater the rates of heat and mass transfer. While this is desirable for increasing capacity, it is offset by higher entrainment and carryover, which are characteristic of small particles in an upward flowing turbulent gas stream. Inevitably, this results in accelerated fouling of the relatively cool boiler tubes and more rapid pluggage of the heat transfer section of the boiler.

A common way to debottleneck kraft recovery boilers is to fire higher solids liquors (75-80%), thereby reducing the water vapor content of the furnace gas and allowing for a greater flow of combustion products. Obviously, control of black liquor droplet size and size distribution is crucial to successful firing of higher solids liquors so that the proper balance can be maintained between in-flight combustion and char bed burning. A major operational difficulty arises with firing high solids liquors, namely being able to contend with the accompanying increased viscosity. The easiest way to reduce the viscosity of concentrated liquor to the level of standard nozzle solids liquor is to raise liquor firing temperature. Thus it becomes important to understand the dependence on temperature of droplet size and size distribution.

The work reported here presents results of an experimental study in which mill black liquors were sprayed through commercially available black liquor nozzles at several different temperatures. We noticed a dramatic change in the behavior of the spray discharge from the nozzle above certain temperatures; namely, much smaller drops resulted. This phenomenon appeared to be due to operation above the liquor boiling point, resulting in flash evaporation of water from the liquor within the nozzle.

The initial drop diameter is a critical parameter in the operation of a recovery boiler and has a direct effect on the amount of carryover and the condition of the char bed. Tests were performed to determine the operating conditions where this change in spraying behavior occurs. Performance characteristics have been determined, including flow rate/pressure drop correlations, droplet size and size distribution.

The performance of spray nozzles for kraft black liquor has been the subject of several recent investigations (2-7). Although research is incomplete, some information is available about the performance of the three basic commercial types of black liquor nozzles mentioned above. Data have been reported on the flow/pressure drop characteristics and droplet size distributions, along with treatments of the mechanism of droplet formation from sprays, and correlation of the resultant mass median diameters in terms of liquor properties and operating parameters, as well as combinations of appropriate dimensionless groups.

Bennington and Kerekes (2) were the first to report on the size and size distribution of black liquor droplets from spray nozzles. They used small grooved-core nozzles similar in design to the swirl cone nozzles used in some mills for firing black liquor, but about one-twentieth the size of a standard commercial nozzle. Not too surprisingly, the droplet sizes from their nozzles were about one-tenth those from commercial black liquor nozzles. Data were reported for a 56% solids black liquor at 100 to 135C. They observed mean droplet size to decrease continuously and weakly as temperature was increased through its boiling point (112C). There were not enough data to make any conclusions on the effect of flashing on liquor droplet size, although there is some evidence in the literature (3) that claims a few degrees change in liquor temperature can change a coarse spray into a fine mist.

In earlier work related to the present study (4-6), commercially available black liquor nozzles have been evaluated. Droplet size distribution data were correlated well with a square root-normal distribution function which involves only two parameters, the mass-median diameter and the normalized standard deviation (standard deviation/[mass median diameter]$^{0.5}$). Results from three nozzle types showed the normalized standard deviation to be relatively constant at 0.2, effectively rendering the distribution function a one-parameter model. Of the design and operating parameters examined, the three which most strongly influenced mass-median drop diameter were nozzle diameter, velocity, and viscosity. Limited temperature data showed a weak inverse dependence similar to the work of Bennington and Kerekes. This work expands significantly on the effects of temperature, including above the liquor boiling point.

EXPERIMENTAL

Black liquors were obtained from several kraft mills and sprayed continuously through commercial black liquor nozzles using a heated, recirculating pump-around loop system. The central component of the system was a spray chamber which served the dual purpose of providing a visible spray pattern while containing and storing the liquor inventory. The stainless steel chamber was 2.1m long, 1.8m deep, and 3.0m high with a V-shaped heated bottom that could store up to 1100 liters of liquor at 94C. The front and rear walls contained 1.5m x 1.2m tempered glass windows to allow back-lighting and video-taping the liquor spray sheet and droplets. The side of the chamber had an opening for the spray nozzle which was oriented to deliver the liquor sheet in a horizontal direction parallel to the windows. Baffles were used to restrict the spray pattern to a narrow path parallel to the windows. Another set of baffles was attached to the front window around the field-of-view of the video camera to keep that area clean.

A two-stage Moyno pump circulated the liquor from the spray chamber through a spiral heat exchanger, which could raise the liquor temperature by 15C at 190 l/min, then through a Brookfield viscometer and back to the chamber. When the liquor was at the desired conditions for testing, the bypass loop was closed and the flow directed through the nozzle. The system had the capacity to deliver to the nozzle up to 150 l/min (as measured by a Foxboro magnetic flowmeter)at 5.5x10E5 N/m2 (80 psi). A continuous water addition line was included in the pump-around loop to replace any water vapor which escaped from the hot liquor spray, thereby maintaining a constant liquor solids content. Because of odor control considerations, the spray chamber was operated under a slight negative draft. An ID fan drew air in through eight adjustable dampers, after which this flow plus any volatiles coming from the liquor spray were pulled through a carbon absorber (provided by Westvaco Corp.) before being exhausted to the atmosphere.

To achieve liquor temperatures in excess of 130C, the pump-around loop was revised to allow for direct injection of steam into the liquor just ahead of the spiral heat exchanger (c.f. Fig.1). After the heat exchanger, the liquor had to flow through about 8m of piping before it reached the nozzle. This was expected to provide adequate mixing length for the liquor to be homogeneous prior to being sprayed. On-line knowledge of the liquor solids content was achieved through a correlation with liquor temperature and viscosity, which were monitored continuously. Direct injection of steam in general decreased the liquor solids content by 3 to 5 percentage points.

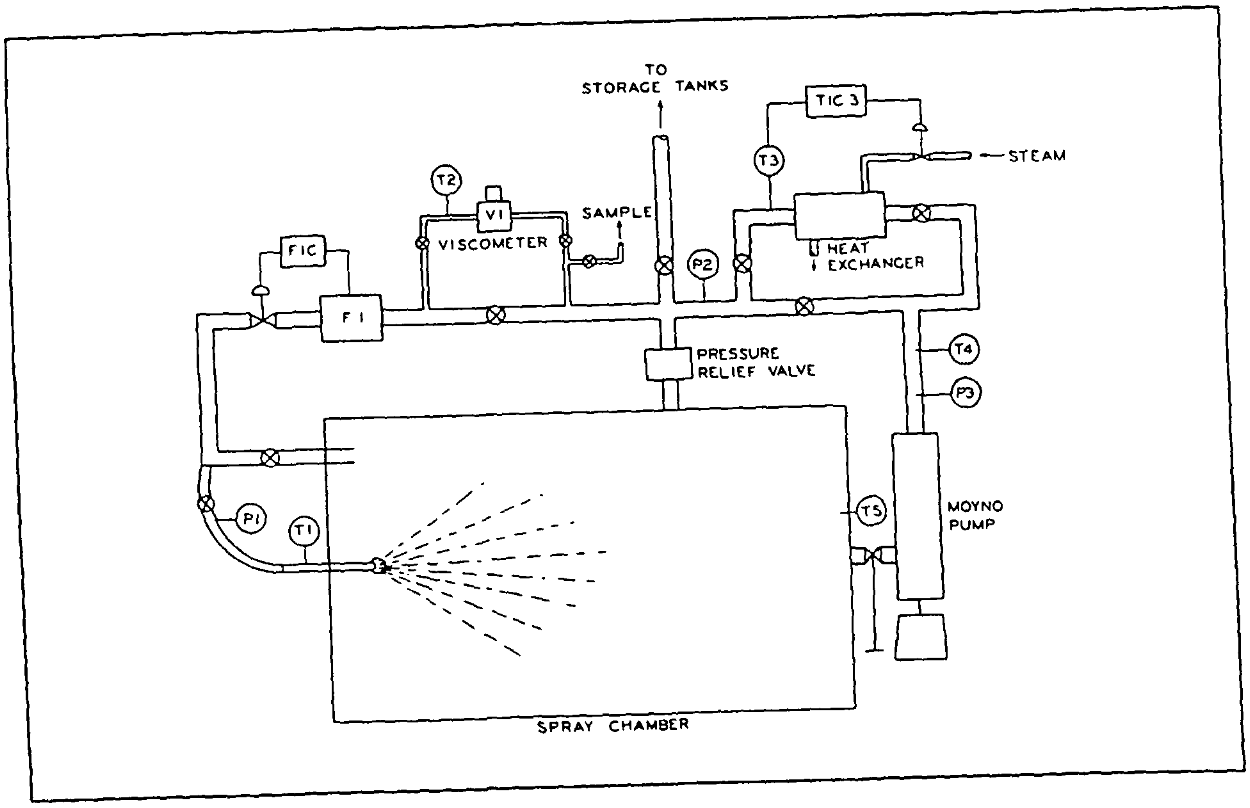

Figure 1. Schematic of Experimental Spraying Apparatus.

The nozzles used in this work are representative of the types being used in recovery boilers. The three nozzles used were a Babcock & Wilcox 12/45 splashplate, a Spraying Systems V-jet 65200, and a Combustion Engineering swirl cone. All three were approximately the same size and are at the small end of the size range that could be considered industrial scale (9.5mm orifice, 45 liters/min at 1.72x10E5 N/m^2). The tests were performed by adjusting the nozzle pressure to a given flow level at a relatively low initial temperature (about 95C). The liquor temperature was gradually increased by using the heat exchanger and direct steam injection, until the transition in spray behavior was observed. The steam was then shut off and the temperature allowed to fall back below the transition point. Values during a typical test run are shown in Fig. 2.

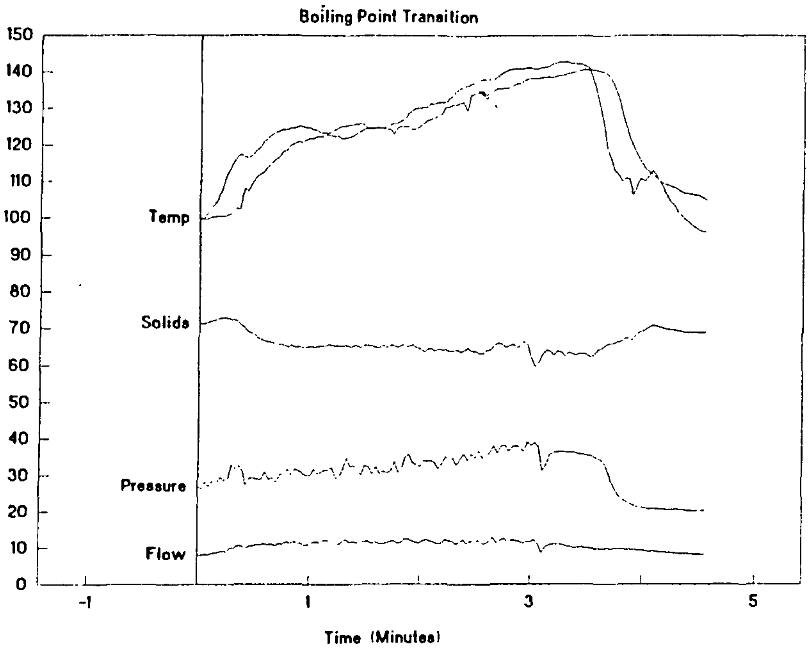

Figure 2. Test with B+W 12/45 Splash-plate Nozzle.

A high-speed video camera, manufactured by Xybion Electronics Systems (Model ISG-250), was used to record drops in a section of the spray pattern at a position approximately 1.2 m from the end of the spray nozzle. A light source was located behind the rear transparent window to give a high contrast video image. The camera uses an image intensifier and electronic gating to obtain images at very high shutter speeds of about 50 micro-seconds. The video images were recorded on a Super-VHS recorder and later transferred to a Tracor Northern image analyzer (Model TN-8502). After subtracting an averaged background image to eliminate images of spots on the windows, the gray level image was converted to a binary image and then analyzed to determine number and sizes of drops.

An assessment of the accuracy of the image analysis techniques was made using ball bearings and cylinders of known diameter. Measurement error was found to be less than 6% for diameters greater than 2mm; the minimum quantitatively measurable (+/-50%) diameter was about 0.3mm.

Spraying at the higher solids levels tended to give problematic images; strands and ligaments, along with discrete droplets, appeared in the field-of-view in the same manner as reported by Bousfield (7). In order to obtain drop size distributions from these images, we assumed that the strands would break into droplets with the same size distribution as the drops already in the images. To justify this, images taken along the centerline of the spray at different distances from the nozzle were analyzed with and without the strands included. The difference in mean diameters was less than 10% when the strands/ligaments portion of the image was less than 30%.

RESULTS

Transition Temperature

As black liquor temperature was raised continuously, there was a temperature (designated the transition temperature) above which the spray droplet size decreased dramatically. At the same time, another change was observed in the shape and direction of the spray pattern. For both the V-jet and splashplate nozzles, the normally flat spray sheet opened up into an oval-shaped cone; also, with the splashplate nozzle the central plane of the spray moved away from the plane of the nozzle plate. The angle of this deviation was not measured, but it was estimated to be 20 - 40 degrees. These effects were observed to be reversible as temperature was lowered continuously to a point below the transition value.

Both of these changes can be attributed to the vaporization of water. The physical effects of flashing can be analyzed by considering the flow of black liquor through the nozzle. Although the specific designs of the nozzles are quite different, they are similar in that the black liquor flows through a restriction before the spray issues from the nozzle. As the liquor flows through the restriction, its velocity increases and its pressure decreases. The pressure at the inlet of the nozzle is higher than the vapor pressure of the liquor and hence single phase flow prevails at this point. As the liquor flows through the nozzle orifice and its pressure decreases, a point is reached where the vapor pressure exceeds the line pressure and vaporization occurs.

Drop size is known to decrease as velocity increases (2,6). A lower density, two-phase flow emerging from the nozzle above the transition temperature and having a higher velocity at the same total mass flow as a low temperature, single phase flow, would therefore be expected to have a smaller diameter. The change(s) in shape and/or direction of the spray sheet above the transition temperature with a splashplate nozzle can result from the flashing steam contributing a component of momentum normal to the plate and therefore to the original plane of the spray.

The transition temperature was estimated to be several degrees above the atmospheric boiling point of the liquor and is dependent upon the dissolved solids content of the liquor. In this work, the transition temperatures were observed to be about 5C above the atmospheric boiling point at 60% solids and about 9C at 70% solids.

Since the transition temperature is the liquor boiling point at some elevated pressure, it should be predictable in the same way that the atmospheric boiling point of black liquor is (8):

$$T_{bp} = 100 + K_{bp} \left(\frac{S}{100 - S}\right) \quad (1)$$

Hence,

$$T_{tr} = 100 + K_{tr} \left(\frac{S}{100 - S}\right) \quad (2)$$

Subtracting eq.(1) from eq.(2),

$$T_{tr} - T_{bp} = (K_{tr} - K_{bp}) \left(\frac{S}{100 - S}\right) \quad (3)$$

Using the two rough data points above, the value of $K_{tr} - K_{bp}$ is 4C. Typical values of K_{bp} range from 6 to 10C (8).

Using the Clausius-Clapeyron Equation, it is possible to estimate the vapor pressure of the liquor at any given temperature. Values were calculated for the observed transition temperatures, using an atmospheric boiling point from Eq.(1). As expected, the transition temperature vapor pressure is below the inlet nozzle pressure, which simply confirms that the flashing is initiated somewhere between the inlet and outlet of the nozzle. The vapor pressure values calculated ranged from 1.6 to 6.7 psig. These relatively low values indicate that vaporization should not start until the liquor is almost at the nozzle outlet.

Flow Coefficients

Since the black liquor flow rate to a boiler is controlled by maintaining a constant header pressure, a correlation describing the relationship between nozzle pressure and black liquor flow rate is very useful. In earlier work (6), a simplified equation derived from the Bernoulli equation for incompressible flow was used to predict the pressure drop across the nozzle:

$$\Delta P = \frac{V_n^2}{2} Cf \qquad (4)$$

Using experimental pressure drop and flow rate data, Cf -values and Reynolds numbers were calculated for a wide range of test conditions (but not at temperatures above the boiling point). These results were used to determine correlations for the splashplate and V-jet nozzles. For several different splashplates:

$$Cf = 1.24 + 458/Re^{0.9} \qquad (5)$$

For the Spraying Systems V-jet:

$$Cf = 1.09 + 540/Re^{1.35} \qquad (6)$$

In this work at operating conditions near the transition temperature, the flow coefficients were found to be higher than these correlations would predict (i.e., pressure drop was higher than expected).

In our previous work at lower temperatures (6), the pressure drop across the spray nozzle was assumed to equal the gauge pressure at the nozzle inlet because the outlet of the nozzle was at atmospheric pressure. For the high temperature tests where the temperature is above the atmospheric boiling point, a simple correction for ΔP can be made by using the difference between the inlet pressure and the vapor pressure of the black liquor. The flow coefficients calculated using this method agree closely with the correlations established earlier at low temperatures for all nozzles tested.

That this empirical approach seems to work comes from examining the physical flow situation. The minimum pressure in the nozzle occurs at the vena contracta near the nozzle outlet. The pressure at this point cannot drop below the vapor pressure of the liquor. If it did, vaporization of water from the liquor would occur to maintain vapor-liquid equilibrium. As more water evaporates, the liquor temperature would slowly drop (assuming adiabatic operation) and so would the pressure. The result is that the outlet pressure is maintained near the water vapor pressure for temperatures above the atmospheric boiling point.

Using this approach, the calculation of flow coefficients has been redone for all of the pressure vs. flow data which have been collected. The effect of using this pressure correction is shown in Fig. 3 for the B & W 12/45 splashplate nozzle. The scatter in the flow coefficient data is reduced significantly after the vapor pressure is employed. A regression analysis applied to all the splashplate nozzles tested (two B & W, one Gotaverken, and one Tampella) has resulted in Eq.(7):

$$Cf\text{-}adj = 1.35 + 371/Re^{0.9} \qquad (7)$$

Despite differences in the nozzle design which could affect flow patterns, all the results appear to lie on the same curve ($r^2 = 0.856$). The result is similar to, but slightly higher, than the earlier low temperature correlation.

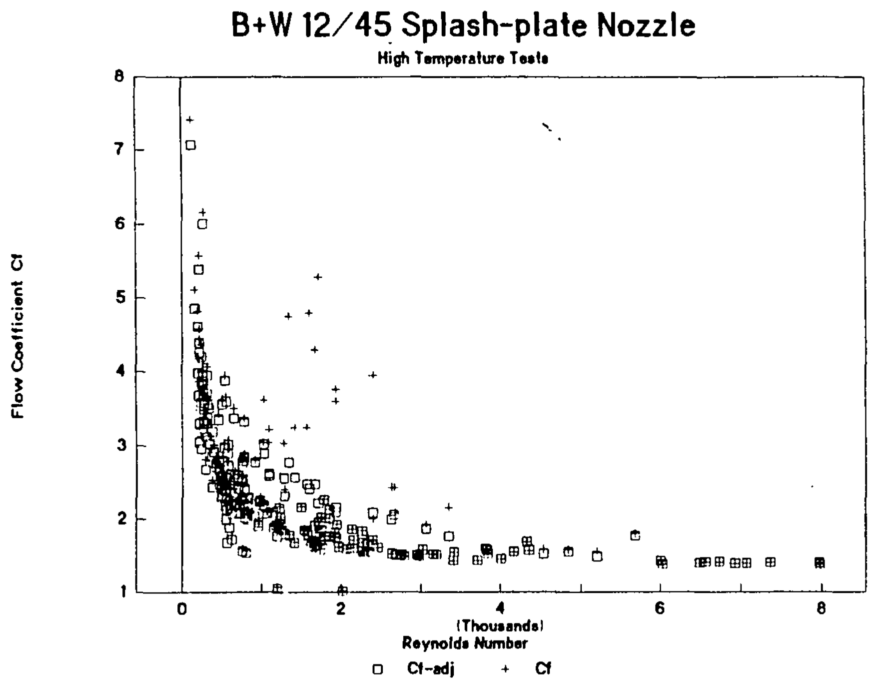

Figure 3. Correct Calculation of the Flow Coefficient Requires the Liquor Vapor pressure.

Three different sizes of V-jet nozzles produced by Spraying Systems Inc. gave results which all followed the same curve, independent of nozzle diameter (c.f. Fig.4). The resulting equation is comparable to that reported earlier for low temperatures; the exponent for the Reynolds Number decreased from 1.35 to 1.0 (r^2 = 0.362):

$$Cf\text{-}adj = 1.06 + 66.4/Re \quad (8)$$

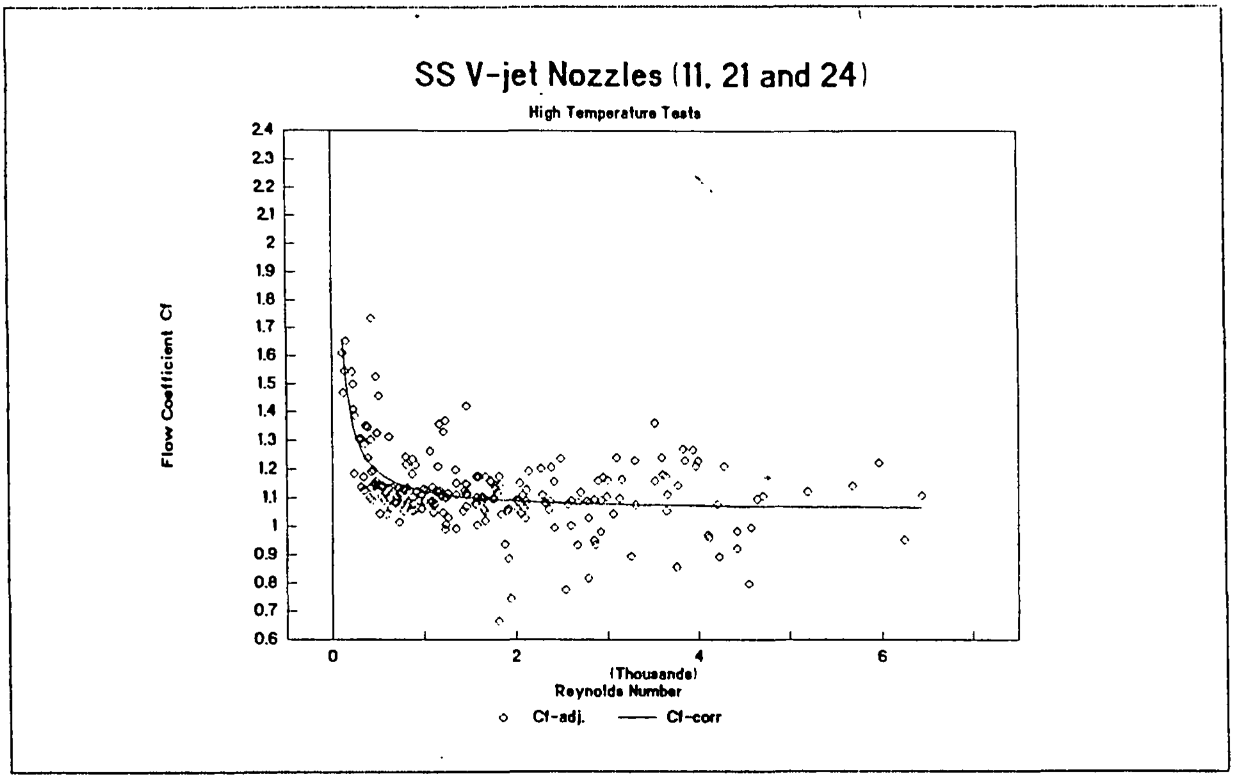

Figure 4. Flow Coefficients for three sizes of SS V-jet Nozzles.

As in the low temperature work, this technique applied to the swirl cone nozzle does not yield a good correlation with Reynolds Number. The geometry of the swirl cone is more complex, with the liquor passing through two separate restrictions before discharging. The previous analysis of flow coefficients at low temperatures gave an average value of 1.11. With high temperature data included, again Reynolds Number was not significant, and Cf-adj averaged 1.12 (c.f. Fig 5).

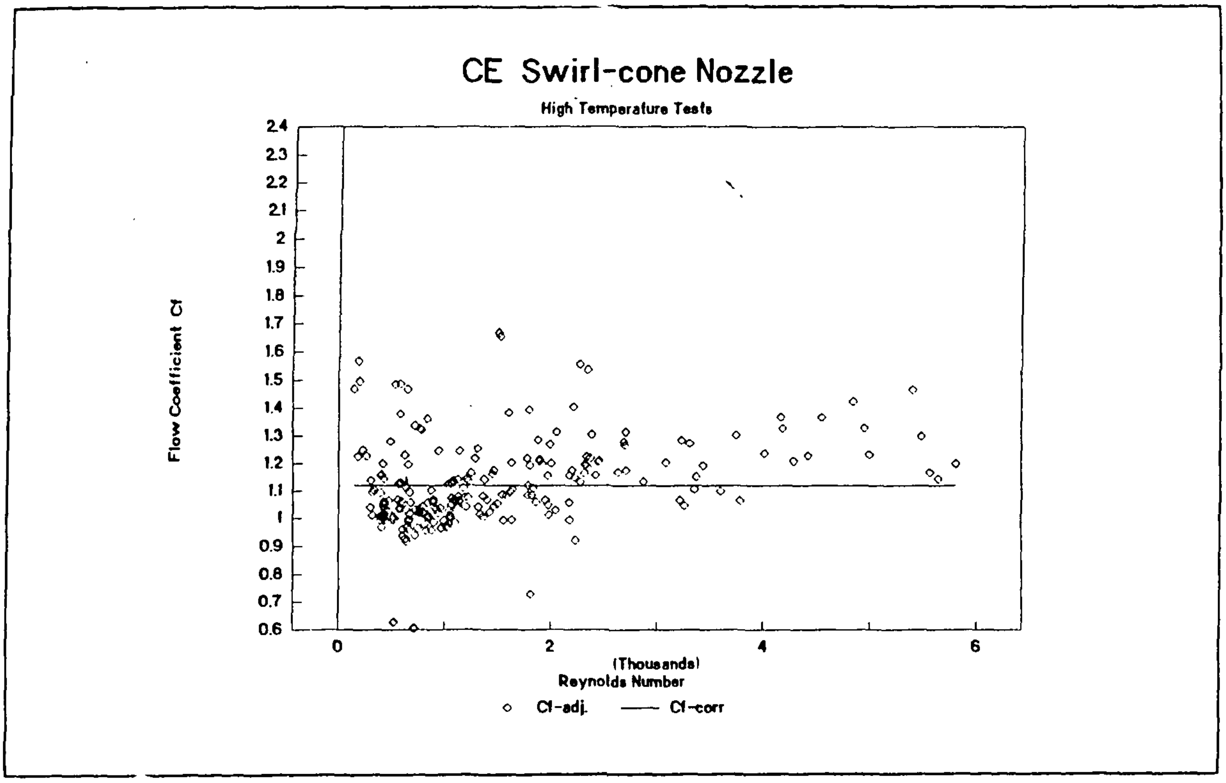

Figure 5. There is no clear trend of C_f with Re for the CE Swirl-cone Nozzle.

Droplet Diameter

The average median drop size above the transition temperature was about 80% of the size measured just below the transition temperature, but because of the difficulty in obtaining accurate data at high temperatures, the actual difference in diameter may be more pronounced.

In addition to the normal amount of error associated with drop size measurements, there is additional variability in the droplet data at high temperature because the video recordings were made during unsteady state conditions where the liquor was being heated rapidly. Also, as the mean drop size decreases, the error increases, since the minimum drop diameter which can be measured on the image analyzer is about 0.3mm. Finally, the movement of the spray pattern toward the camera, when operating above the transition temperature, changes the calibration slightly, and this has not been accounted for in these results.

When operating above the transition temperature, the smaller drop sizes and the displacement of the spray pattern from the splashplate both result in over-estimating the drop size. Therefore, the actual drop size at temperatures above the transition will be smaller than reported.

As the spray temperature was gradually increased, a continuous videotape of the spray pattern was recorded. The image analysis of these tapes was performed by first analyzing the segment of the tape above the transition temperature which appeared to have a minimum drop size. Then, temperatures below the transition (during both heat-up and cool-down) were analyzed to determine mass median drop size and standard deviation. Because the timing of the videotape with instantaneous spraying conditions was not accurately known, a best estimate was made to determine the liquor transition temperature. The measured median drop size is plotted against the estimated nozzle temperature in Fig. 6 below. Qualitatively, the graph shows that median drop size decreases as temperature increases.

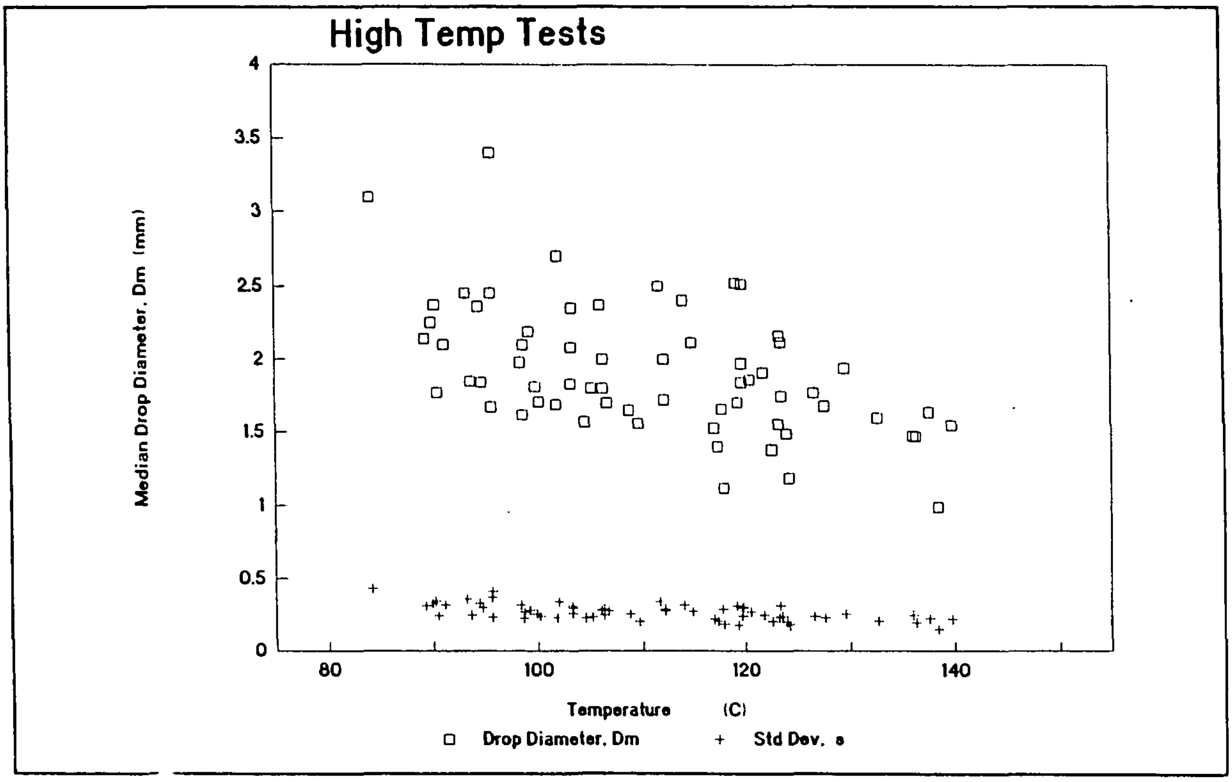

Figure 6. Median Drop Diameter vs Nozzle Temperature.

Since the transition temperature is a function of liquor solids level, it is useful to define an excess liquor temperature (Tx) as the difference between the liquor temperature (Tl) and the estimated transition temperature (T_{tr}). Hence, Tx is positive when the liquor temperature is above the transition and negative when it is below. Drop diameter is plotted against Tx in Fig. 7. If a linear dependence of Dm on Tx is assumed for the region of Tx < 0, a zero slope (based on a standard t-test) results; similarly, for Tx > 0, a statistically significant negative slope is determined.

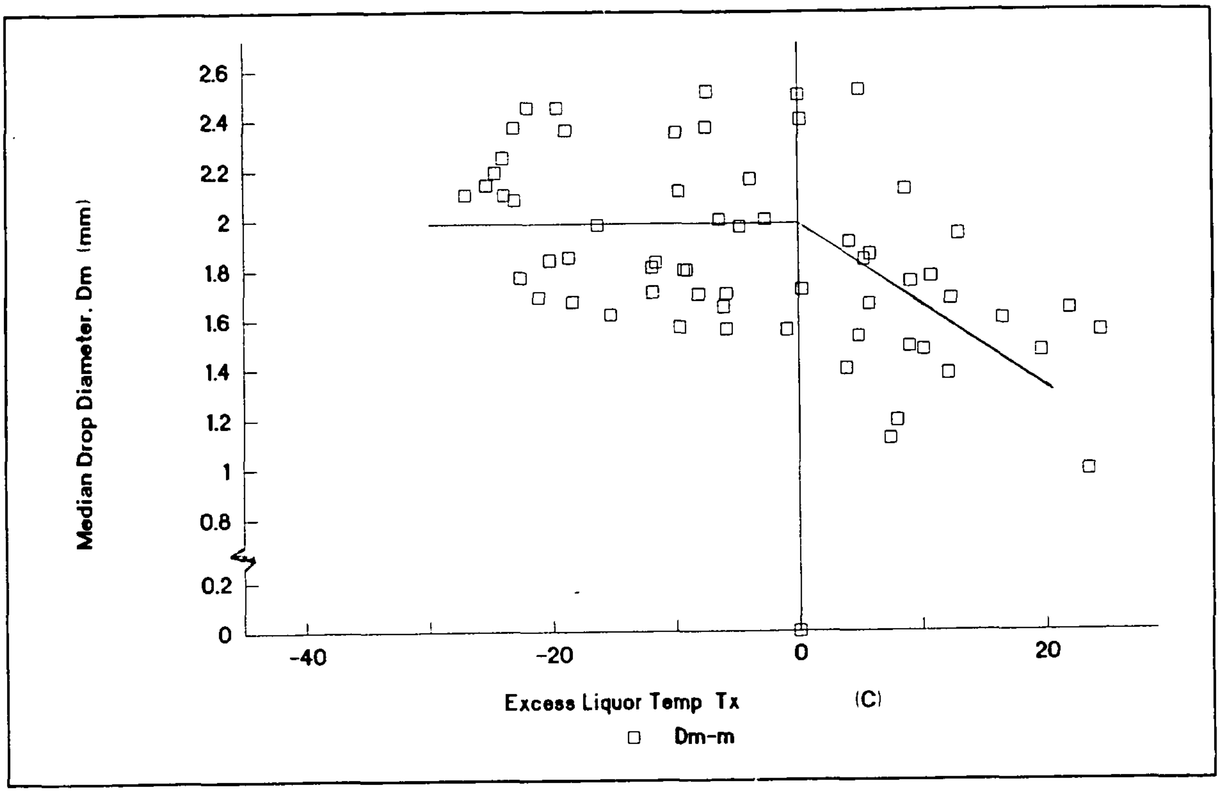

Figure 7. Median Drop Diameter Drops when T_x is greater than zero.

An average drop size was calculated for temperatures both above and below the T_{tr}. For Tx < -2.5, there were 35 data points with an average Dm = 1.99 mm; for Tx > 2.5, Dm averaged 1.63 mm. Hence, the diameter ratio above and below the transition temperature is 0.82. It should be noted that a more accurate value will be somewhat lower, since, as discussed earlier, the drop size measurements above the transition temperature are on the high side. Applying this result to recovery boiler operation, a 20% decrease in median drop diameter will mean that for the same liquor mass flow, the number of drops will be doubled and the surface area will be increased by about 30%.

In addition to measuring the median drop size, the standard deviation of a square root-normal distribution was also calculated for each test. Previous work at temperatures below T_{tr} has shown that the normalized standard deviation (s^*), defined as $s/Dm^{0.5}$, is essentially invariant at 0.20 (+/- 0.02). For temperatures above T_{tr} in this work, the average value for s^* was 0.19, signifying essentially no change in s^* and leaving the square root-normal distribution model as a one parameter model.

Previous work with black liquor and V-jet nozzles below the transition temperature (6) reported an empirical dependence of median diameter on velocity raised to the -0.34 power and temperature to the -0.09 power. If we assume this to hold above the transition temperature as well, we can calculate a value for the gas phase volume fraction in the nozzle. Based on a 20% decrease in drop diameter in going from 115C to 127C at a constant total liquor flow rate, the correlation predicts a gas phase volume fraction of 0.48. At the estimated pressure in the nozzle at this point, vaporization of less than 1% of the water in the liquor would give this volume fraction. This percentage seems low, and more data above the transition temperature are needed to establish the velocity/temperature dependence of drop diameter in this region.

CONCLUSIONS

1. The transition temperature approximates the temperature in the nozzle at which point liquor boiling starts at the local nozzle pressure. Transition temperature correlates with black liquor solids in the same functional way as does the atmospheric boiling point. At typical liquor firing solids levels, the difference between the transition temperature and the normal boiling point was about 8C.

2. Operating at temperatures above the transition results in three phase flow before the liquor exits the nozzle, thus increasing nozzle exit velocity. The resultant additional energy release causes an increased rate of sheet break-up and more than a 20% decrease in median drop size. This effect is accompanied by a change in the spray sheet cross-section from planar to oval, and, for a splashplate nozzle, a change in the direction of the spray sheet that leaves the plate.

3. For splashplate and V-jet nozzles operating below the boiling point of the black liquor, the relation between nozzle inlet pressure and flow rate can be predicted from previously derived correlations. At temperatures above the boiling point, the pressure drop for these correlations must be based on the nozzle inlet pressure and the liquor vapor pressure.

4. The average change in droplet size as a consequence of operation above the transition temperature was a decrease of about 20%. This would result in almost doubling the number of drops being fired into the boiler. The smaller drop size would increase droplet surface area and hence in-flight rate processes; however, liquor carryover rates would also increase. The normalized width of the droplet size distribution was the same, whether below or above the transition temperature.

LIST OF SYMBOLS

C_f - Flow coefficient at atmospheric pressure
$C_{f\text{-}adj}$ - Flow coefficient based on liquor vapor pressure
D_m - Mass median droplet diameter (mm)
K_{bp} - Boiling point rise of 50% solids black liquor at atmospheric pressure (C)
K_{tr} - Proportionality constant for eqn.(2) (C)
ΔP - Liquor pressure drop across nozzle (N/m^2)
Re - Reynolds Number
s - Standard deviation
s* - Normalized standard deviation
S - Black liquor solids concentration (%)
T_{bp} - Boiling point of black liquor at atmospheric pressure (C)
T_1 - Liquor temperature (C)
T_{tr} - Transition temperature (C)
T_x - Excess liquor temperature (C)

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ACKNOWLEDGMENTS

The authors thank the U.S. Department of Energy for financial support and specifically Stan Sobczynski of DOE for his guiding hand and commitment to the program. Doug Samuels provided skilled and innovative operation of the equipment. Contributions of the IPST support staff are also gratefully acknowledged.