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For

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ABSTRACT

An innovative method for spraying black liquor into a recovery boiler is disclosed which features independent control of median droplet size for virtually any percent solids liquor at temperatures below its flash point. Based upon effervescent atomization, air is mixed with the black liquor feed at some point upstream of the nozzle orifice such that a bubbly two-phase flow is produced. Upon exiting the nozzle, the fluid mixture "explodes" into ligaments and drops. The resulting median drop diameter is dependent upon the standard operating conditions of nozzle size, liquor solids and temperature (i.e., viscosity), and liquor flow rate, and upon air/liquor ratio. Using corn syrup as a model fluid, drop sizes for liquid viscosities ranging from 100 to 10,000 mPa·s were determined for flow rates of 30 and 45 liters per minute using a commercial Vee-jet nozzle. It is shown that effervescent spraying enables variation of the median drop size for specified (and fixed) normal operating conditions simply by changing the gas/liquid ratio. Application of this technology to recovery boiler operation is discussed.

Introduction

Independent control of droplet size when black liquor is sprayed into a kraft recovery boiler is highly desirable from the standpoint of decreasing shutdown frequency or increasing liquor solids throughput without getting excessive droplet entrainment and carryover to the superheater and boiler bank. Unfortunately, once liquor percent solids, temperature, and feed rate through a given nozzle are specified, the resulting median drop diameter and size distribution are fixed; they are a result of the balance of fluid mechanical forces which break up the liquor spray sheet and ensuing ligaments into droplets. Previous research on spraying black liquor through conventional splashplate and V-jet nozzles has quantitatively shown the dependencies of median drop size and size distribution on the operating parameters listed above [1]. With the present black liquor spraying technology, it is not possible to change the median drop size or size distribution without changing one or more of the operating parameters indicated.

Unfortunately, changing any of these operating parameters can not be done without impacting other operating parameters or other unit operations in the kraft recovery cycle. A change in liquor solids can only be accomplished by a change in the evaporator plant, and a feed rate change impacts both the evaporators and the caustic plant; a change in liquor temperature requires a change in the liquor heater operation. In short, to proactively change the liquor firing conditions, numerous other process operations must be altered to enable the change; it can't be done independently.

If a mill should want to go to high solids firing, it must raise feed liquor temperature to counteract the increased liquor viscosity. When it does this, there is a practical limit above which the spraying operation encounters a flashing condition. Effectively, the liquor reaches its boiling point in the feed line before the nozzle orifice, establishing a two-phase gas-liquid flow. The observed result is the formation of a finer spray issuing from the nozzle, leading to a condition of excessive droplet entrainment and carryover to the superheater. This then forces the operator to reduce the liquor feed temperature, often resulting in a "roping" condition, necessitating a subsequent reduction in percent solids.

Previous research (with normal solids liquors) at the Institute of Paper Science and Technology has shown that this flashing condition produces a significantly smaller droplet mass median diameter (MMD) with an apparent different drop formation mechanism as compared to conventional spraying in which drops are formed by liquid sheet disintegration [2]. Where and how flashing occurs in the feed line and spray nozzle is, for the most part, uncontrollable, resulting in spray properties that are unsteady and unpredictable.

This paper introduces an innovative method for delivering any percent solids black liquor to a recovery boiler without having to change the normal operating conditions for black liquor spraying. It uses a technique called effervescent spraying, which can be defined as a twin-fluid process in which an atomizing gas is injected into the liquid feed at some point upstream of the nozzle to form a bubbly two-phase flow [3].

When this mixture exits the discharge orifice, the rapidly expanding bubbles shatter the surrounding liquid into droplets whose mean size is largely determined by the ratio of injected air to flowing black liquor.

The important observation to make at this point is that this gas/liquor ratio can be set independently of all other recovery boiler operating variables.

Experimental

The experiments in this study were performed using nitrogen (as the atomization gas) and corn syrup as a model fluid, with its viscosity adjusted by adding or evaporating water. The details of the spraying facility have been described previously [1,2], although modifications were made for this study to handle a wide range of liquid viscosities and gas flows for specific black liquor nozzles (Figure 1). Starting at the 1500-liter storage tank, the liquid flows to a 4-stage Moyno pump, then through an electromagnetic flowmeter, ending with the spray nozzle, which is oriented in a horizontal direction parallel to the viewing window. Nitrogen is injected into the liquid approximately 0.1 meter prior to the nozzle orifice, with the gas flow being measured by a Hastings mass flowmeter. Pressure was measured (Ametek Model #851 transducers) before gas injection and just before (27 mm) the nozzle exit orifice. Experiments were run at room temperature, with the exact liquid temperature measured just before gas injection, thereby accounting for any small viscosity changes. Liquid viscosity was measured using a Brookfield viscometer (model RVT). Due to the presence of entrained gas in the liquid after spraying, experiments were run in semi-batch mode, allowing time for phase separation between runs, which reused the sprayed liquid.

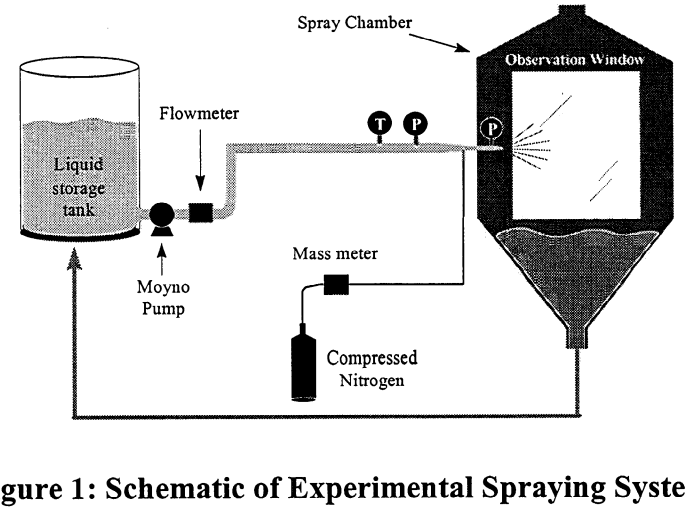

Figure 1: Schematic of Experimental Spraying System.

Corn syrup was used as a model fluid for black liquor, since preliminary testing showed its rheology to be very similar to black liquor, but with much better physical property uniformity and stability. Four levels of viscosity were evaluated (100, 750, 2800, 10,000 mPa·s) spanning the range of values for normal firing solids liquor (70-

75%) and high solids (80-85%). Experiments were performed at liquid flowrates of 30 and 45 liters per minute (LPM) and gas flowrates from 0 to 1300 standard liters per minute (SLM).

Initial experiments showed that the location of gas injection was critical to the quality of the spray produced. A schematic of the gas injection technique is shown in Figure 2. Gas was injected through many small holes (0.75 mm diameter) in the pipe wall, then distributed into the liquid using a 10-cm length of static mixer (Koch Engineering; model SMX; 25 mm diameter) before exiting the spray nozzle. Although effervescent sprays were effectively produced previously using a nozzle with a plain circular orifice [4], the experiments reported here were performed using a spray nozzle more commonly used in recovery boilers, namely, a Vee-jet nozzle (Spraying Systems #65200; 8.7 mm equivalent diameter).

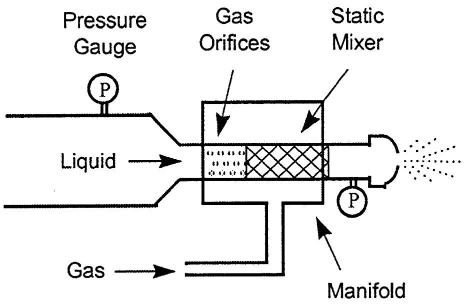

Figure 2: Schematic of Gas Injection/Mixing Method and Spray Nozzle.

Spray Analysis

Qualitative and quantitative spray results were obtained by analyzing video images produced by a high shutter speed camera (Xybion model ISG-250). The camera position for drop size analysis was approximately 1.3 meters horizontally from the end of the spray nozzle. A spray separation baffle was installed near the end of the nozzle to limit the quantity of spray within the camera's depth of field and to maintain a clean viewing window. Converting the video images into meaningful drop size data required making assumptions with respect to:

- converting a 2-D image into 3-D drop size information
- limitations in size determination
- defining edge boundaries, noise, and depth of field concerns
- assumptions for strands and other nonspherical shapes

A standardized image analysis filtering and arithmetic operations routine was developed (using Optimas image analysis software) to eliminate noise and define edge boundaries. Because most drops were not perfectly spherical, it was necessary to translate the 2-D drop images into equivalent drop diameters. For an individual drop, the measured area and perimeter can be assumed to be proportional to the actual drop volume and surface area. The applied method uses the area and perimeter of each drop image and converts it into a cylinder with hemispherical ends (because most nonspherical drops appear as such). Then, after calculating the cylinder volume, the diameter of a sphere of equal volume was calculated, this being referred to from this point on as the equivalent drop diameter. The lower limit of drop diameter detection was approximately 0.3 mm; below this size, it was impossible to accurately define drop shape or size. For each set, between 1000 and 10,000 drops were measured (15-25 image frames). The maximum level of uncertainty to be expected in the drop size MMD measurements was ± 20%.

Spray angles were determined from video images of the near-nozzle spray structure. Using a top view of the spray sheet leaving the nozzle orifice, a "fanning out" angle, as defined by the sheet boundaries, is easily quantified. From a side view of the spray sheet, a "sheet thickness" angle can be measured, relative to the flat sheet (zero sheet thickness angle) for liquid-only spraying. Due to the inherently unsteady nature of effervescent spraying, it was necessary to produce an average image of 32 individual images (using image analysis), thereby allowing a more accurate determination of the sheet thickness angle from the nozzle orifice.

Drop Formation

Conventional spraying (liquid-only) with a Vee-jet nozzle was analyzed in order to make meaningful comparisons with effervescent spraying. Similar to splashplate nozzles, the Vee-jet nozzle operates under the principle of spreading the liquid into a thin sheet, which is disrupted by wave thinning and perforation mechanisms, forming strands and eventually drops [5]. At low viscosity, the liquid sheet breaks up quickly after exiting the nozzle, forming nearly spherical drops by the time they reach the video imaging area. As viscosity increases, however, the liquid sheet disintegration process is significantly slowed, such that strands persist further downstream from the nozzle. Eventually a viscosity is reached where a continuous stream is formed from the nozzle (traditionally classified as "roping").

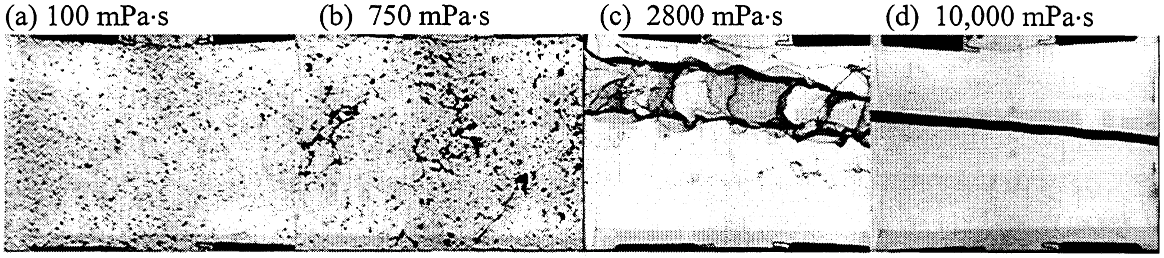
(a) 100 mPa·s (b) 750 mPa·s (c) 2800 mPa·s (d) 10,000 mPa·s

Figure 3: Images of Liquid-only Spray Showing Viscosity Effect (liquid flowrate = 45 LPM; images 1.3 meters from nozzle)

Effervescent spraying does not rely on forming a liquid sheet for disintegration; rather, atomization is accomplished by the explosion of gas exiting the nozzle. Images of the near-nozzle spray structure illustrate this effect, as shown in Figure 4. Initial addition of gas (extent indicated by the gas/liquid mass flow ratio: GLR) causes the liquid sheet to disappear, being replaced by liquid strands exiting immediately from the nozzle orifice.

As shown for the lowest GLR levels, the curve-shaped strands are oriented normal to the direction of flow with fairly uniform spacing. As discussed by Crapper *et al.* [6], the appearance of these "waves" of liquid strands indicates a frequency imposed by some external force, in this case, the unsteady release of bubbles from the nozzle orifice. Pressure fluctuations, as measured just before the nozzle orifice, verifiy the unsteady forces being imposed on the liquid by the rapidly expanding bubbles.

For each tested viscosity level, increasing the GLR causes thinner strands with a more disordered orientation (as shown in Figure 4). The thinner strands are most likely the result of: 1) increasing number and size of bubble explosions, and 2) increasing liquid velocity, caused by the gas reducing the effective cross-sectional area for liquid flow at a constant liquid flow. The thinner strands result in smaller drop sizes. At the highest GLR level evaluated, drops appear to form immediately upon exiting the nozzle orifice.

The rate at which the strands disintegrate into drops depends on the liquid viscosity. Increased viscosity causes strands to persist further downstream from the nozzle. At high viscosity levels (2800 and 10,000 mPa·s) and low GLRs, strands do not break up into spherical drops before reaching the end of the spray chamber (1.8 m downstream of nozzle). At the highest evaluated GLR level, however, the differences between sprays of different viscosity are minimized.

Drop Size

For conventional liquid-only spraying, drops do not form above a certain viscosity threshold (dependent on liquid flowrate and nozzle size). As the images in Figure 3 indicate, individual drop distinction becomes indeterminable at high viscosity levels, since all or portions of the spray are a continuous stream of liquid. One aspect of this research was to determine if an effervescent spraying process could effectively spray a liquid that is too viscous for conventional spraying methods.

As shown in Figure 5, the gas flowrate has a dramatic effect on the drop size MMD for effervescent spraying. At the highest viscosity levels (2800 and 10,000 mPa·s), adding a small amount of gas (GLR=0.0001-0.0002) disrupts the continuous 30 LPM liquid stream to form large drops of 10-20 mm diameter. Further increases in the gas flowrate reduce the drop size MMD to the desirable range for a recovery boiler (2-4 mm). Lower viscosity levels require a lower GLR to attain a similar drop size.

Although not shown, increasing the liquid flowrate from 30 to 45 LPM results in the exact same trends as shown in Figure 5, but reduces the drop size MMD by approximately 20-30%. This decrease can be attributed to the higher flow velocity and pressure drop experienced in the nozzle orifice, subsequently causing more intense bubble explosions and faster moving liquid strands. Both effects reduce the spray drop size.

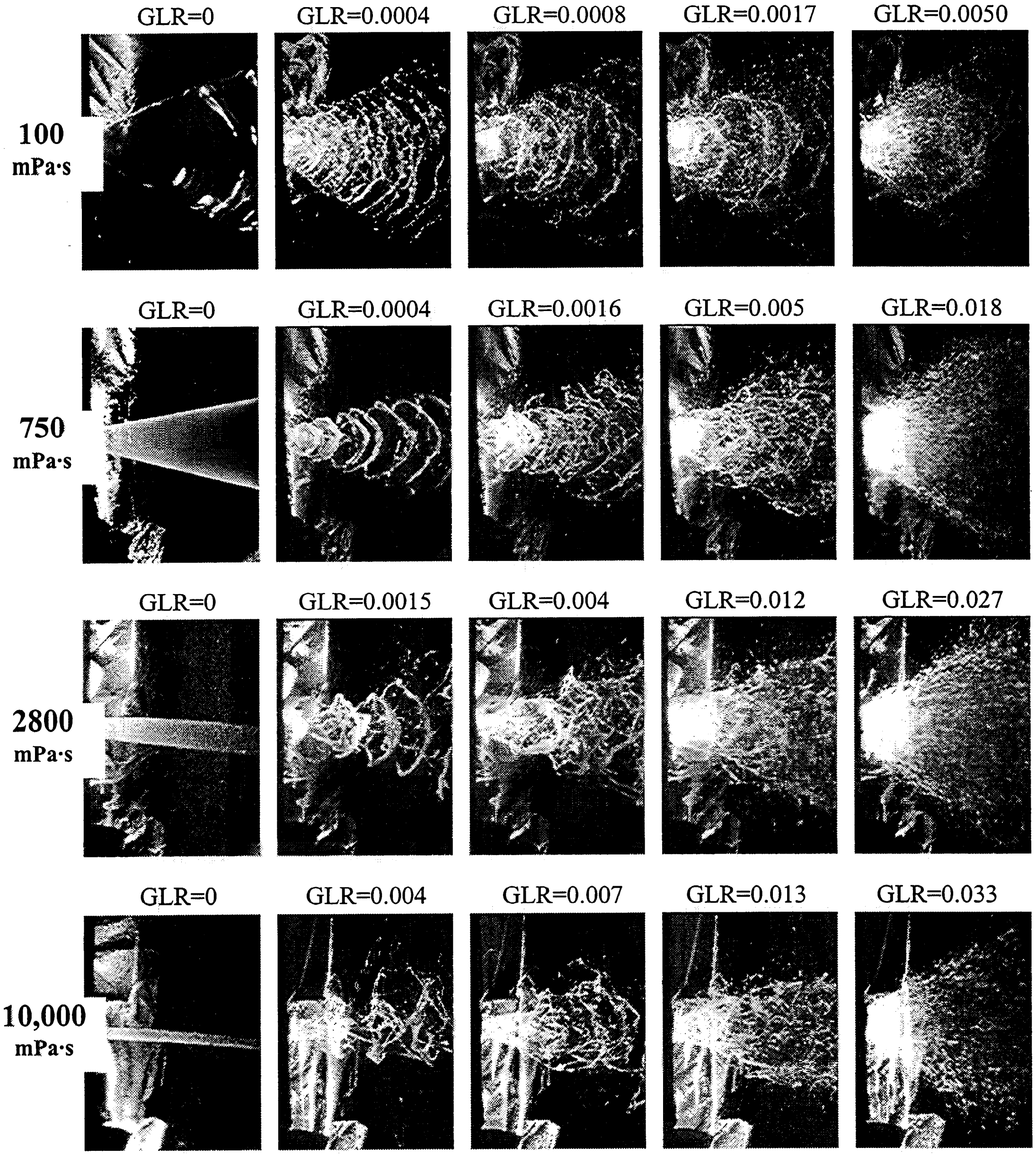

Figure 4: Images of the Near Nozzle Spray Structure for Liquid-only (GLR=0) and Effervescent Sprays at Increasing Gas Flowrate and Liquid Viscosity (liquid flowrate=30 LPM).

Close inspection of the lower viscosity data in Figure 5 (100 and 750 mPa·s, shown more clearly in Figure 6) reveals, as GLR increases from zero, an initial increase in MMD compared to liquid-only spraying. This increase in drop size at low GLRs was unexpected, since previous research concerning black liquor flashing reported a smaller drop size than spraying without flashing [2]. More recently, Helpio et al. did report a small increase in MMD at the start of flashing using a splashplate nozzle [7]. The larger drops can be attributed to the different drop formation mechanism for effervescent spraying compared to sheet disintegration (as discussed previously). At low GLR levels, the strands that form immediately upon exiting the nozzle orifice are thicker than the strands formed through disintegration of the liquid-only sheet, thus resulting in a larger drop size MMD compared to liquid-only spraying.

The implications for this effect when applied to black liquor delivery to a recovery boiler are profound. Without changing black liquor feed rate, percent solids, or temperature or nozzle size or type, the median drop diameter can be changed by almost a factor of two simply by mixing air with the liquor feed ahead of the nozzle using a GLR of about 0.0005. If smaller MMDs should be desired, raising the GLR will accomplish this without having to alter the liquor feed conditions. Changes elsewhere in the pulping and recovery cycle would not be needed to achieve these results.

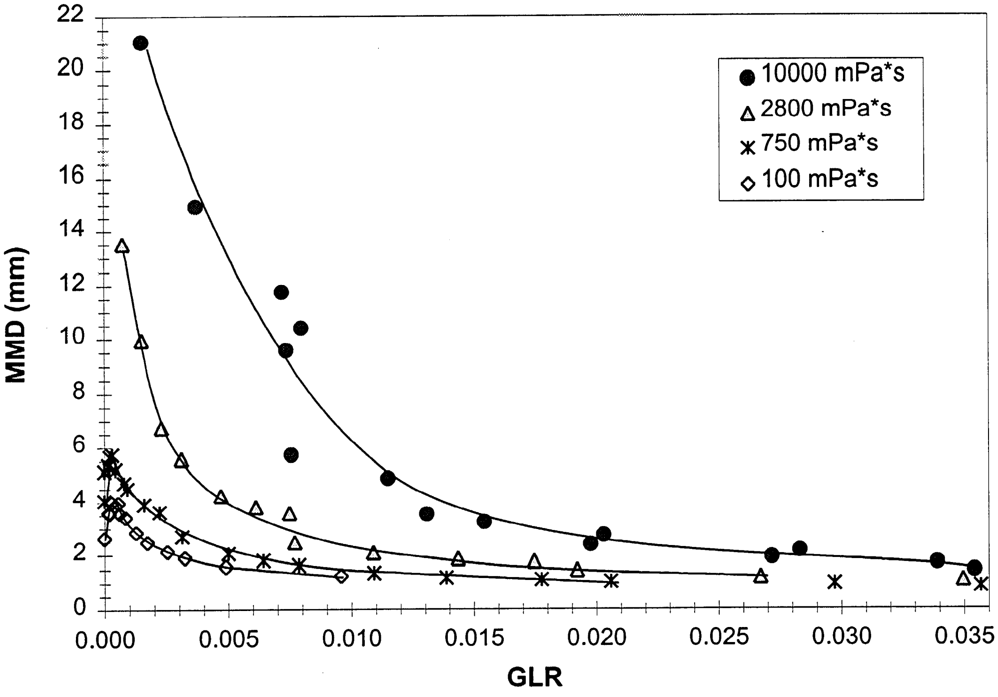

Figure 5: Drop Size MMD as a Function of GLR and Viscosity for Effervescent Spraying (using Vee-jet nozzle at 30 LPM; uncertainty estimates are ± 20% for MMD and ± 1% for GLR; trendlines are for graphical purposes only).

It would appear from Figure 5 that the drop size MMD levels out at about 1 mm as the GLR is increased, even at the highest viscosity. This is by no means believed to be true because other studies show drop sizes below 0.1 mm are possible at very high GLRs [8]. Unfortunately, however, current video camera and image analysis limitations do not allow drops smaller than 0.3 mm diameter to be accurately detected. Although the mass fraction of these tiny drops is very small at the lower GLRs, higher GLRs should cause a higher mass fraction of these tiny undetectable drops. Thus, results reported here are probably slightly higher than they actually are for reported MMDs in the 1-1.5-mm range. With respect to recovery boiler operation, knowing the relative quantity of these small drops is critical because they collectively contribute to the carryover problem mentioned earlier.

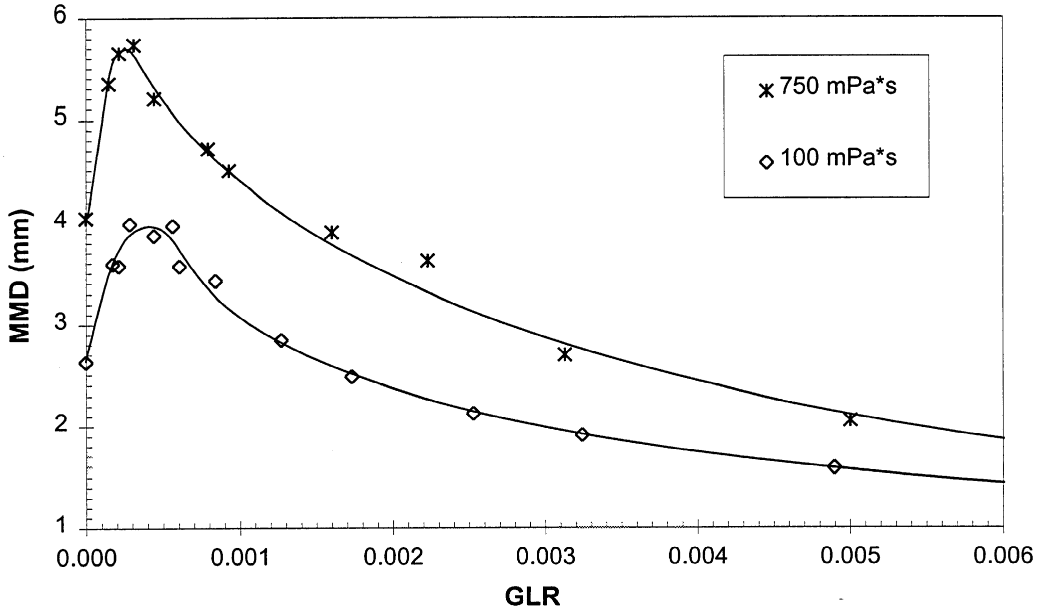

Figure 6: Drop Size MMD as a Function of GLR for Viscosities 100 and 750 mPa·s (close-up view of data in Figure 5).

Drop Size Distribution

Previous research for black liquor spraying has shown that the drop size distribution was best modeled mathematically by the square-root normal distribution, and that the normalized standard deviations (standard deviation divided by the square root of the MMD) were relatively constant at 0.20 ± 0.03 [1]. After analyzing effervescent spray drop size results, it was determined that the square-root normal function still provided the best overall fit to the data, although sprays containing abnormally large strands and globules (observed at high viscosity and low GLR conditions) were better represented by a normal distribution. However, in order to make meaningful comparisons among drop size distribution data, standard deviation values were calculated based on the square-root normal function for all drop size data sets.

Experimental results of this study showed an average value for the normalized standard deviation of about 0.25 ± 0.02 for liquid-only spraying at 100 mPa·s, and about 0.29 ± 0.02 at 750 mPa·s. One possible reason for these values being larger than those of the earlier black liquor study is that a more advanced image analysis software (Optimas) was used in the present study.

Although not taken into account in the drop size analysis, conventional liquid-only sprays contain a relatively thicker portion of liquid in the outer region of the sheet, sometimes referred to as the rim. It is speculated that the rim may have a significant effect on the drop size distribution. At low viscosity (100 mPa·s), the rim forms relatively larger drops and strands compared to the central portion of the spray; however, at 750 mPa·s and higher, the rim is essentially a continuous stream of liquid (Fig. 7(a)). In either case, if quantified into an equivalent drop size, the rim would skew the drop size distribution and likely produce a much larger normalized standard deviation value for the total spray (compared to the spray's center-view only). Effervescent sprays, however, do not show a rim effect (Fig. 7(b)); thus, the drop size distributions, as measured from the central portion of these sprays, are more representative of the entire spray.

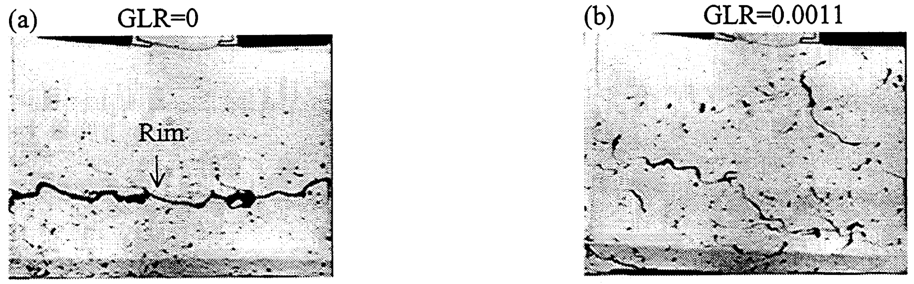

Figure 7: Outer Region of Spray Showing (a) Rim Effect for Liquid-only Spray, and (b) No Rim Effect for Effervescent Spray (both sprays are at 750 mPa·s viscosity and 45 LPM).

Graphs of normalized standard deviations, as they varied with gas flowrate and viscosity, are shown in Figure 8. At 100 mPa·s, values are comparable to liquid-only spraying (≈ 0.25), decreasing at high gas flow rates due to video measurement limitations (i.e., when the MMD approaches 1 mm, the drop size distribution appears more narrow due to the lower detection limit of 0.3 mm diameter).

At 750 mPa·s, adding a small amount of gas causes a sharp decline in the normalized standard deviation. The liquid-only value of 0.27 at GLR = 0 falls to 0.14 at GLR = 0.0001 to 0.0002. Increasing the gas flow causes values to become closer to liquid-only spraying, then decline again due to measurement limitations. At low GLRs, the relatively uniform strands disintegrate into drops of more uniform diameter. Increasing the gas flowrate creates a greater variety of strand size and thickness, thus producing a wider drop size distribution more typical of liquid-only spraying. A similar trend was observed at 2800 and 10,000 mPa·s; however, comparison to liquid-only spraying was not possible under these conditions, since a continuous liquid "rope" exited the nozzle at these high viscosities. In general, it can be concluded that effervescent spraying produces a similar drop size distribution as liquid-only spraying, but in certain cases at low GLR values, it actually gives a narrower distribution (particularly when the spray rim effect is taken into account).

Applying this finding to recovery boiler operation has interesting implications. There has been a long-standing question of whether or not a narrow drop size distribution is beneficial for conventional recovery boiler operation. Until now, this was an academic question because we had no way to independently change the distribution. Effervescent spraying now provides a limited "window" for addressing this question. A small adjustment in the GLR can independently narrow or widen the drop size distribution.

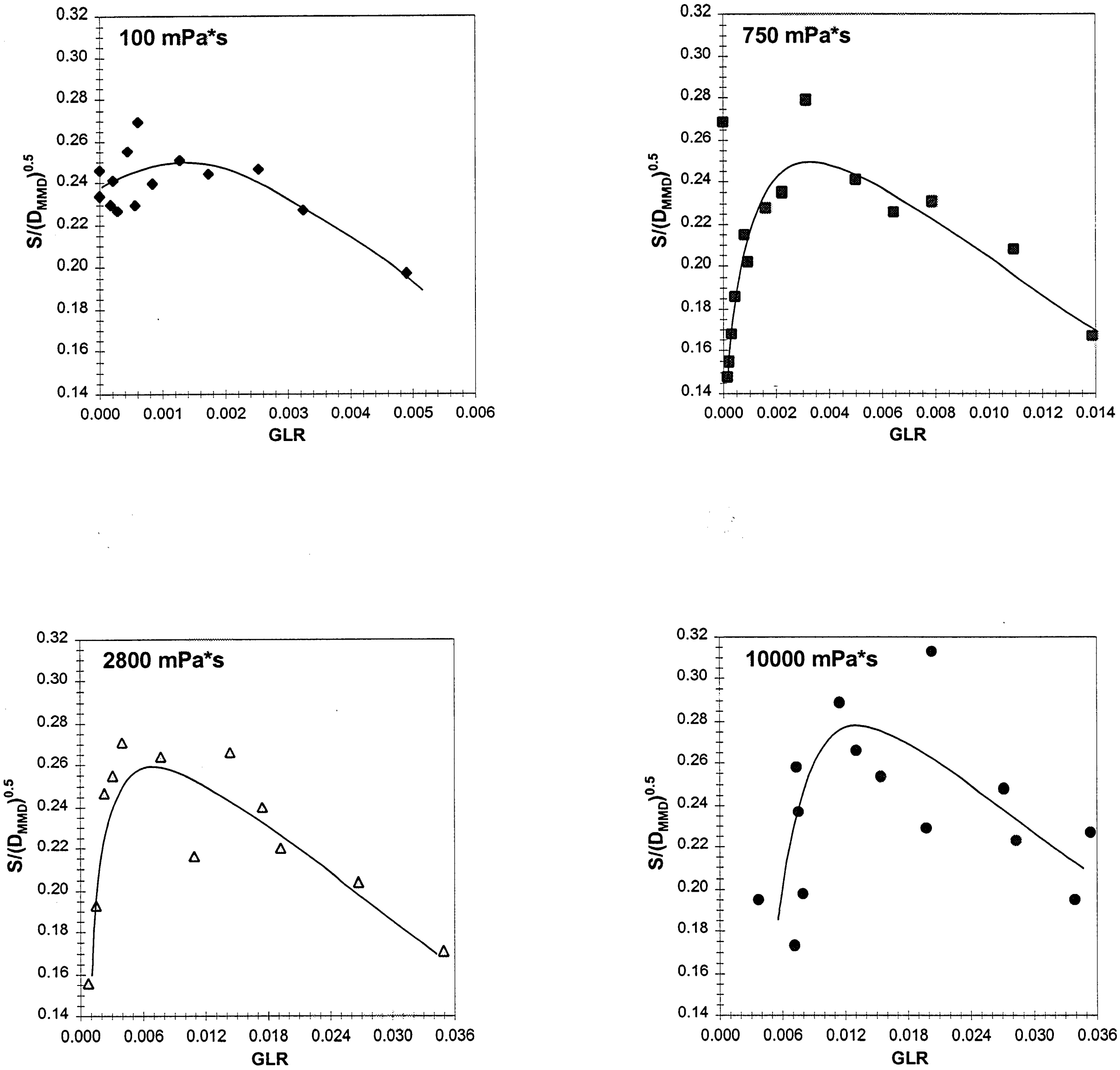

Figure 8: Drop Size Normalized Standard Deviation Varies with GLR for Effervescent Sprays at Different Viscosities (Vee-jet nozzle at 30 LPM; trendlines are for graphical purposes only).

Spray Angle

Previously, it was mentioned that liquid-only spraying with the Vee-jet nozzle distributes the liquid in a sheet that fans out in relatively two-dimensional form, whereas effervescent sprays exit the nozzle in a more conical form. Increasing the GLR causes an increase in the spray fanning-out angle (shown qualitatively in Figure 4; quantitatively in Figure 9). Liquid viscosity acts as a resistance to changes in momentum; thus, for a given GLR, increasing viscosity causes a decrease in spray angle (as shown in Figure 9). The error in spray angle determination is estimated at ± 5°.

The spray "sheet-thickness" angle (as measured from a view perpendicular to the fanning-out view) is zero for liquid-only spraying (a flat sheet); whereas effervescent spraying shows a relatively constant angle of about 60° (at a GLR > 0.003) that is relatively unaffected by liquid flow rate, viscosity, and GLR.

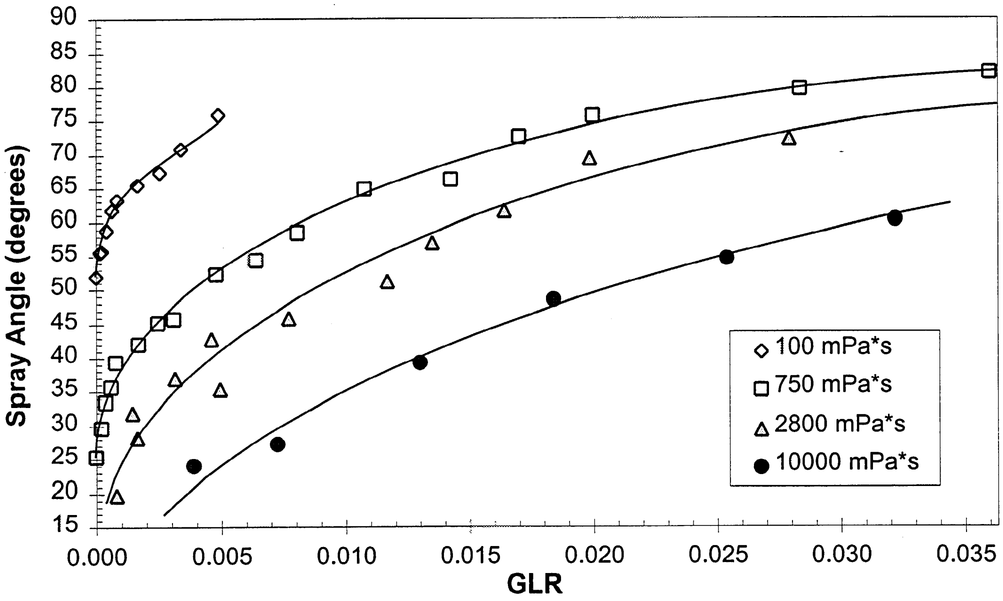

Figure 9: Spray Angle (sheet fanning-out view) Increases with GLR and Decreases with Viscosity for Effervescent Spraying at 30 LPM with Vee-jet Nozzle (trendlines are for graphical purposes only).

Nozzle Pressure

Nozzle pressure was measured at a point 27 mm upstream from the orifice; thus, the actual discharge pressure should be considered only slightly less than the measured pressure readings due to unaccounted frictional effects between the pressure port and nozzle orifice. Liquid-only spraying pressures were very stable with time, as opposed to unsteady pressure fluctuations observed in effervescent spraying (caused by bubble "explosions" at the nozzle orifice).

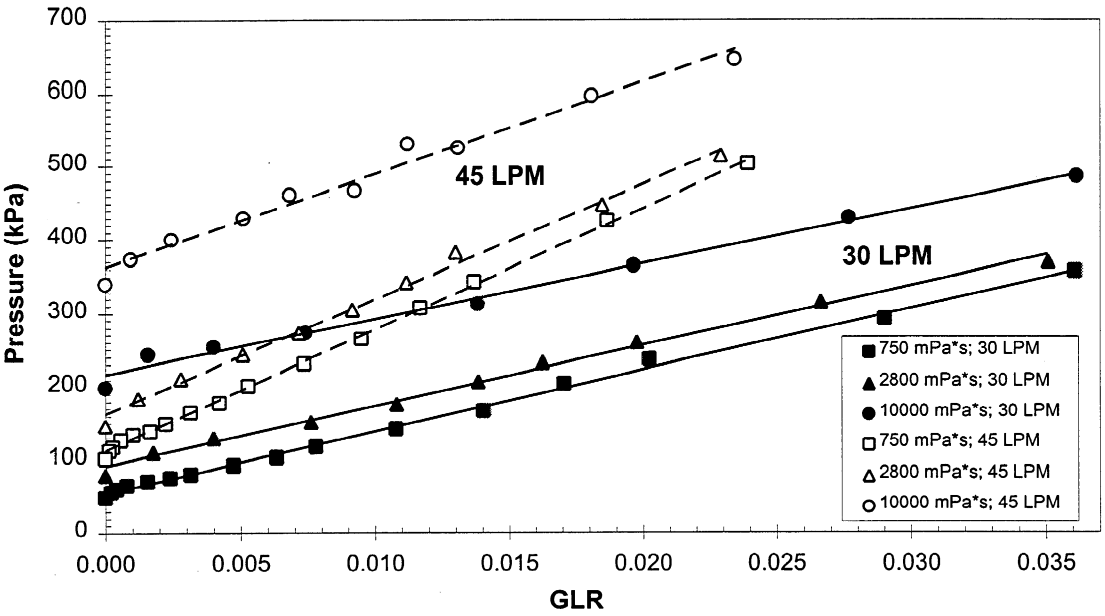

Figure 10: Nozzle Pressure Increases with GLR and Viscosity for Vee-Jet Nozzle.

Average nozzle pressure results are shown in Figure 10. As expected, nozzle pressure increases with gas flow rate, viscosity, and liquid flow rate. Increasing the gas flow rate at constant liquid rate causes an increase in nozzle flow void volume, subsequently resulting in less volume for liquid flow, and thus causing higher frictional pressure losses in the nozzle. The slope of the pressure vs. GLR linear curve was found to be relatively constant for the different viscosity levels tested, depending on the liquid flow rate. An increase in the GLR by 0.001 units caused the nozzle pressure to increase by about 7.7 kPa for 30 LPM liquid flow, and about 15.5 kPa per 0.001 GLR unit increase at 45 LPM. Hence, effervescent spraying of black liquor in a recovery boiler will require slightly higher nozzle pressures than conventional spraying.

Conclusions

- Effervescent spraying enables an effective means of independently controlling the spray drop size by adjusting the injected gas flow rate (as indicated by the gas/liquid mass ratio). The level of gas required to produce a particular drop size MMD depends on the liquid viscosity and flow rate.
- Effervescent spraying at low viscosity (100 and 750 mPa·s) and at low GLR gives an unexpected increase in drop size compared to liquid-only spraying. At a low GLR (≈ 0.0004), the drop size MMD increased by approximately 50 percent compared to liquid-only spraying. The mechanism for producing larger drops appears to be initiated by the gas induced creation of waves of liquid strands that subsequently form larger drops (as compared to the sheet perforation mechanism for liquid-only sprays).

- Effervescent spraying produces a drop size distribution best represented by a square-root-normal function, similar to previous black liquor spraying studies. At low viscosity, the normalized standard deviations showed no significant difference between effervescent spraying and liquid-only spraying. At the higher viscosities, a low GLR resulted in low values for the normalized standard deviation, which increased to normal (i.e., liquid-only) values as GLR was increased. It should be noted, however, that liquid-only sprays were observed to have an outer "rim" of liquid not found in the effervescent sprays. Although not included in drop size determinations, the rim would be expected to increase the normalized standard deviation values. Thus, the actual drop size distribution for liquid-only spraying may be wider compared to effervescent spraying in an overall spray comparison at similar viscosity (≤ 750 mPa·s).

- The "fanning-out" angle of liquid distribution from the nozzle increased as GLR and/or liquid flow rate increased, and decreased as liquid viscosity increased. Compared to liquid-only spraying, the liquid exiting the nozzle was not in a flat sheet form, but rather had an elliptical cone shape.

- Results from this study have shown that liquids that are normally too viscous to spray using a commercial sheet-producing type nozzle can be sprayed effervescently to produce drops in the desirable range for recovery boiler operation (MMD = 2-4 mm). This should lend itself to being an independently controlled, economical method for firing high solids black liquor to a recovery boiler.

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