

# **Characterization of Spray-Drying Nozzles at Industrially Relevant Conditions**

**Continuation Proposal  
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## Abstract

This proposal is a continuation of our current IFPRI research on the atomization process by swirl and twin-fluid nozzles. The current research is on homogenous viscous fluids, and the present proposal extends it to slurries and fluids with suspensions. We will measure the droplet size distributions from sprays of suspensions with different particle types and sizes, and solutions with elongational viscosity, surface tension, and molecular weight. Both pressure-swirl and twin-fluid nozzles will be studied. We will develop physical models for the atomization of these fluids. This will be achieved by performing breakup studies on droplets and ligaments of fluids with suspension. Breakup process of a droplet or a ligament will also allow for a quantitative determination of the effect of the cross-flow temperature on the breakup process.

## Summary of the Current IFPRI Project

A detailed description of the project was submitted in our final report to IFPRI. Here, only a brief discussion of our previous project is provided.

We performed experiments on the sprays generated by swirl and twin fluid nozzles and measured droplet size distributions and obtained near-nozzle images of the atomization process for these nozzles. Glycerin/water and CMC/water solutions were used as test fluids. Mass flow rate, spray angle and liquid sheet breakup lengths were measured for each case.

### 1. Liquid Sheet Breakup Based Model for Swirl Nozzles

The droplet size distribution of high viscosity and polymeric fluids can have a bimodal distribution, with a minor and a major peaks (modes). A mixture of two lognormal distributions fit the volume distribution of all testing conditions. The minor peak (mode) of the size distribution represents the long tail of the volume distribution and it corresponds to droplets of less than 10  $\mu\text{m}$ . These small droplets are mainly generated by the breakup of thin and stretched ligaments generated by the breakup of the swirling liquid sheet. The transition from minor to major peak is smoother for polymeric fluids. The thin ligaments are stretched into a wide range of sizes in the polymeric cases, while they rapidly breakup in glycerin solutions.

A distribution function of the following form was developed that fits the data well:  $f_3(d) = k_v f_{3,1}(d; \hat{\mu}_1, \hat{\sigma}_1) + (1 - k_v) f_{3,2}(d; \hat{\mu}_2, \hat{\sigma}_2)$ . Here,  $k_v$  represents the volume ratio between the major mode for large droplets ( $f_{3,1}$ ) and the minor mode for small droplets ( $f_{3,2}$ ). Based on our model, the droplet size distribution can be predicted using the following steps:

- 1) Determine the fluid properties including density  $\rho$ , kinematic viscosity  $\nu$ , surface tension  $\sigma$  and the operating conditions, including the injection pressure  $P$  and the swirl nozzle orifice diameter  $d_o$ . Measure the mass flow rate  $\dot{m}$  and the spray angle  $\theta$ .
- 2) Calculate the sheet velocity  $u$  using  $u = k_d \sqrt{2P/\rho}$ , where  $k_d$  is the discharge coefficient of the nozzle.
- 3) Solve for the sheet thickness,  $t_{or}$ , from  $\dot{m} = (\rho u \cos \theta) \pi t_{or} (d_o - t_{or})$ .
- 4) Solve for the dominant wavenumber  $k_{max}$  using the dispersion equation

$$\omega = -2\nu k^2 + (4\nu^2 k^4 + \rho_r u^2 k^2 - (\sigma k^3 / \rho_l))^{0.5},$$

where  $\omega$  is the growth rate and  $k$  is the wavenumber.

- 5) Calculate the dominant wavelength  $\lambda_{KH}$  using  $\lambda_{KH} = 2\pi/k_{max}$ .
- 6) Calculate the parameters in the distribution equation using

$$\hat{\mu}_1 = 1.96\lambda_{KH}^{0.589} \text{ and } \hat{\sigma}_1 = 0.065t_{or}^{0.517}.$$

- 7) The volume distribution is given by  $f_3(d) = f(d; \hat{\mu}_1, \hat{\sigma}_1)$ , where  $f$  is a lognormal distribution:

$$f(d) = \frac{1}{d\sigma\sqrt{2\pi}} \exp\left(-\frac{1}{2}\left(\frac{\ln(d/\hat{\mu})}{\hat{\sigma}}\right)^2\right).$$

SMD and other averages can be determined from the above distribution function. In addition, the following correlation is developed for the Sauter Mean Diameter (SMD) of sprays formed by a swirl nozzle and for high viscosity fluids.

$$\frac{d_{32}}{d_o} = 42.52 Re_p^{-0.151} We_p^{-0.519}$$

High resolution closeup imaging of the swirl nozzles revealed that the liquid sheet perforation process is the dominant mechanism for the atomization process. In order to quantify this process, experiments on the atomization of flat sheets of liquid is currently being performed. A flat sheet of liquid, as in fan sprays, eliminates the sheet curvature, and allow for better imaging. Figure 1 shows some typical images of a 60% glycerin-water solution at different injection pressure. At 600 psi injection pressure, the sheet is smooth. As the pressure increases to 700 psi, the liquid inside the nozzle becomes turbulent. Increasing the pressure to 800 psi and then to 900 psi, increases the turbulent intensity, and makes the sheet more perturbed right at the exit of the nozzle. These perturbations expedite the breakup of the sheet. Therefore, laminar sheet breakup models overpredict sheet breakup length. The size of these turbulent structures are at the scale of Taylor microscale. We are currently developing a turbulent based model for this process.

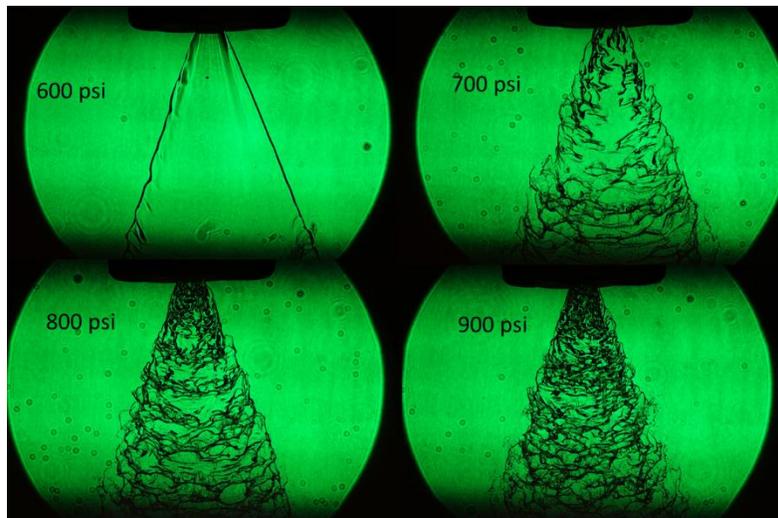


Figure 1. Atomization of 60% glycerin-water solution in a flat fan nozzle at different injection pressures.

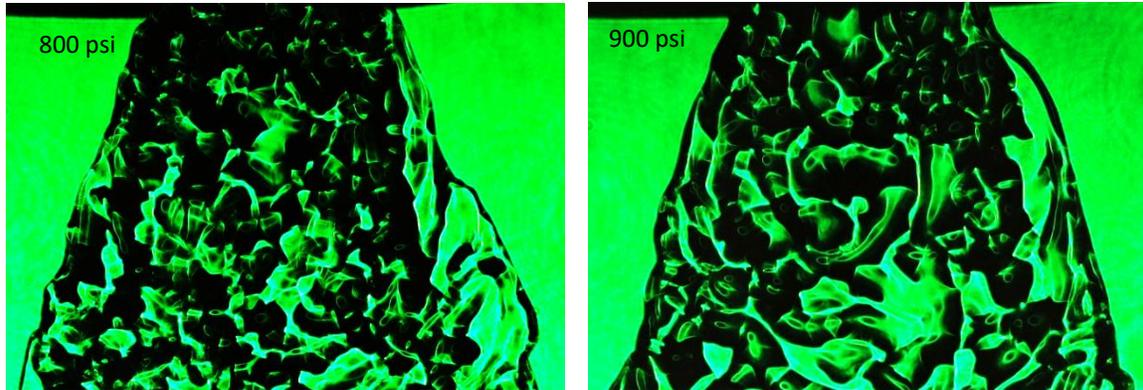


Figure 2. Turbulent structures right at the exit of the nozzle.

## 2. Droplets Breakup Based Model for Twin Fluid Nozzles

For twin fluid nozzles, a new phenomenological model was developed that predicts the size distribution from the primary and secondary atomization. The details of the experience and the model are present in the final report and two journal articles<sup>1,2</sup>. The model is outlined as follows: Starting from an initial characteristic size and relative velocity, the formation of the ligaments (i.e., the rims) is predicted via the droplet deformation rate. The characteristic size can be obtained from the KH wavelength, the liquid jet diameter, or the drop diameter. Three modes of ligament breakup are modeled. Several mechanisms contribute to the breakup of each. Taking the mean and standard deviation of the predictions for each mechanism, the distribution resulting from each mode is predicted. Combining the distributions of the modes, the multimodal distribution of the spray is predicted. In cases where modes overlap considerably, a weighted mean and standard deviation can be used to predict the overall distribution of the constituent modes.

This model is applied to the atomization of twin fluid nozzles, by using the oscillation characteristic of a liquid jet exiting the nozzle (figure 3). The Kelvin Helmholtz wave is used as the size of the droplet that goes through bag breakup. Then, the droplet sizes from this breakup are used to determine the droplet size distribution in the spray as depicted in figure 4. The results of the predictions are compared with the experimental data in figure 5, which shows a good prediction.

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<sup>1</sup> Jackiw, I., and Ashgriz, N., "On aerodynamic droplet breakup," *Journal of Fluids Mechanics*, Volume 913, 25 April 2021, A33.

<sup>2</sup> Jackiw, I., and Ashgriz, N., "Prediction of the droplet size distribution in aerodynamic droplet breakup," *Journal of Fluids Mechanics*, (2022), vol. 940.

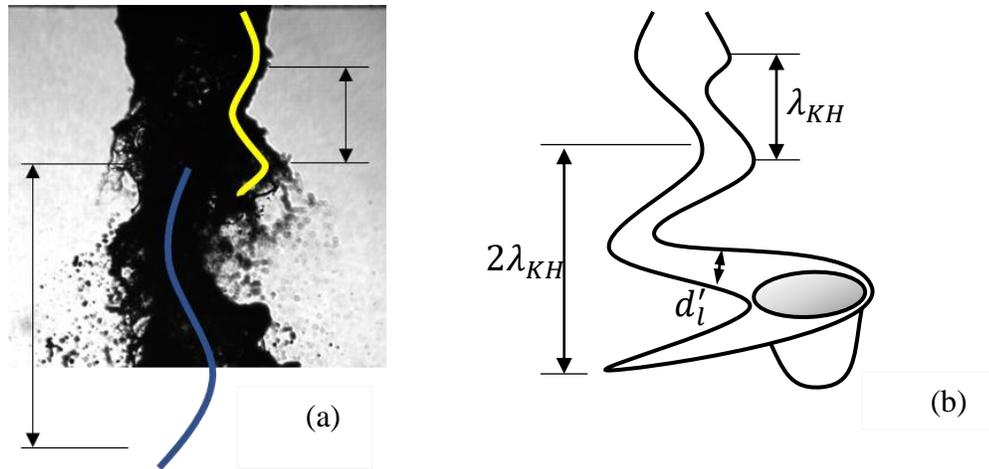


Figure 3. (a) In a twin fluid atomization process, the waves on the liquid grow to an extent that the high velocity air breaks them into small droplets. (b) This is modeled through a bag breakup process.

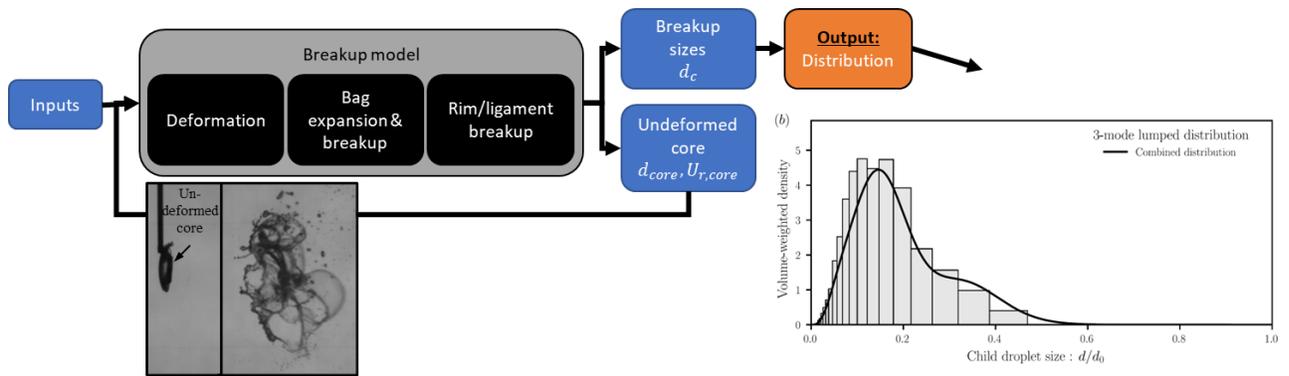


Figure 4. Using droplet breakup model to predict the droplet size distribution in a twin fluid spray.

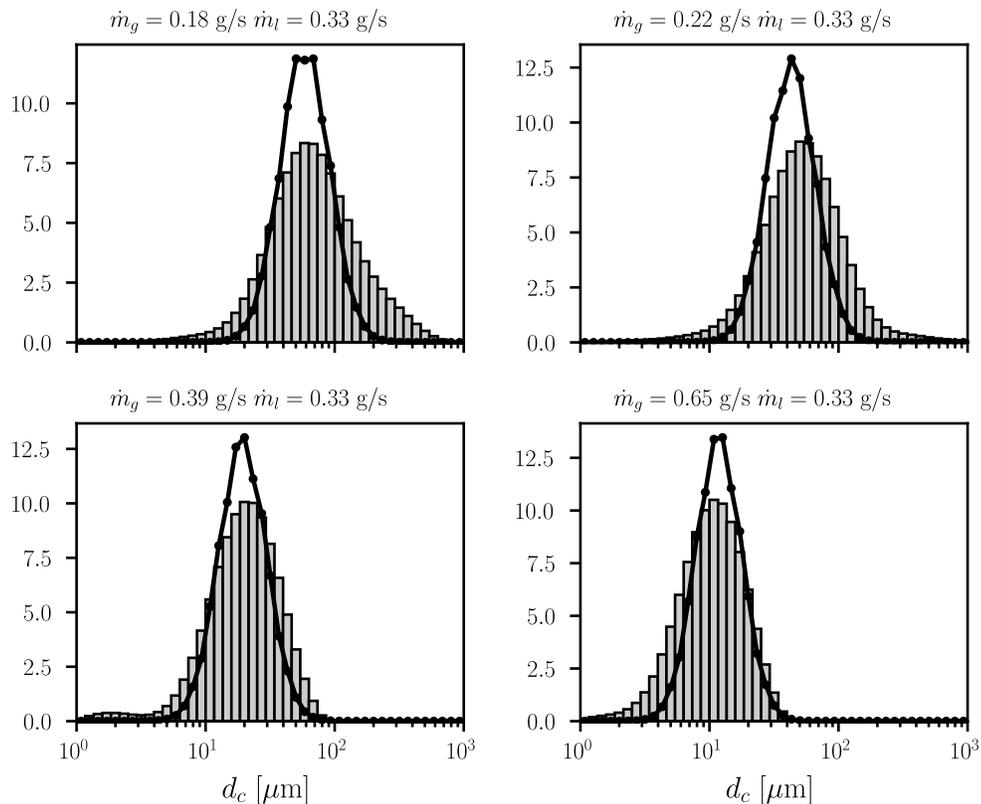


Figure 5. Comparison of our atomization model with the experimental measurements for droplet size distribution for four different flow conditions of a twin fluid nozzle.

## Literature review on the atomization of slurries

The prior studies on the atomization of liquids with suspensions have identified the followings: The influence of the particles suspension on the atomization intimately relates to the particle size and concentration. No change in the spray droplet sizes was found when atomizing slurries with 4 to 16  $\mu\text{m}$  particle sizes and concentrations of 0 to 30 weight percent.<sup>3</sup> However, solid particles of larger than 20  $\mu\text{m}$  separate from water droplets with 70% weight percentage of solid suspension.<sup>4,5</sup> Several studies have shown bimodal distributions for slurry sprays.<sup>6</sup> On the other hand, unimodal size distributions were reported for slurries with mean particles sizes of less than 50  $\mu\text{m}$ , and bimodal distribution for larger than 50  $\mu\text{m}$ . One peak corresponded to the solid particle diameter in the suspension, and the other peak represented pure liquid spray size. At low injection pressures, the peak of the liquid droplet diameter was controlled by the solid particle size. Suspension with relatively fine solid particles result in a smaller diameter peak than the suspension with relatively large solid particles. At high injection pressures, this diameter peak is

<sup>3</sup> Isenschmid, T., 1992 Diss. ETH Nr. 9609.

<sup>4</sup> Glaser, H. W., 1989 VDI Düsseldorf.

<sup>5</sup> Mulhem, B. Fritsching, U. Schulte, G. and Bauckhage, "Characterisation of Twin-Fluid Atomisation for Suspensions," 2001 ILASS-2001 Zurich.

controlled by the gas velocity. Atomization of lime particle slurries using a pressure-swirl nozzle has shown that SMD increases with increasing solid percentages of lime slurry.<sup>6</sup>

Generally, the shear thinning behaviour of the suspension viscosity, which increases with increasing solid concentration and decreasing solid particle size, makes the influence of the viscosity on the suspension break-up within a twin-fluid atomizer less important with increasing velocity of the atomizing gas<sup>7</sup>.

Based on the current state of knowledge on the slurry atomization, the following issues are raised:

- 1- How does a solid particle separate from the solution? It is not clear whether the liquid is sheared off the solid particle or the particle separates during the breakup of the mixture.
- 2- Is there a correlation between the particle separation from the solution with respect to its size, the suspension concentration, and other solution rheological properties?
- 3- Does the SMD of the spray increase with an increase in solid particle size? There are conflicting results on this, as some show no increase, whereas some show an increase in SMD with particle size.
- 4- At what conditions does the unimodal size distribution turn into a bimodal size distribution? Clearly this is a pressure and nozzle dependent condition. However, we aim to determine the physical conditions that may result in this. We will develop a correlation for a critical suspension parameter, at which point the size distribution becomes bimodal.
- 5- What are the differences between the suspension fluid atomization in pressure-swirl and twin-fluid atomization? In pressure-swirl atomization, a solution is converted into a thin sheet, which is then atomized into droplets, whereas in a twin-fluid atomization, a high gas velocity tears the solutions into droplets and ligaments. The influence of particles in each of these processes can be different. For instance, particles in a sheet may expedite preformation and breakup, whereas particles may not break off the core liquid in the shearing process of twin-fluid atomization.

The present proposal is designed to address these questions.

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<sup>6</sup> Yan, Y., Zhang, L., Pan, W., and Pu, G., Experimental Investigation of Atomizing Performance of Low Pressure Swirl Nozzle, *Advances in Mechanical Engineering*, Volume 2014, Article ID 782064, 10 pages.

<sup>7</sup> Mulhem, B., Schulte, G., Fritsching, U., "Solid-liquid separation in suspension atomization," *Chemical Engineering Science* 61 (2006) 2582 – 2589.

## Objectives of the current Proposal

1. Spray droplet size measurement of liquids with solid suspension.
2. Development of correlations for droplet size distribution for slurries using swirl and twin fluid nozzles.
3. Secondary breakup of ligaments of fluids with suspensions.
4. Development of a perforation-based model for slurry atomization by swirl nozzles.
5. Development of a droplet breakup-based model for slurry atomization by twin fluid nozzles.

## Proposed Research

### 1. Spray Characterization of Liquids with Solid Suspension

There have been several requests from IFPRI members to perform tests with slurry fluids, as well as considering the effects of other rheological properties on the atomization. We propose to investigate the effect of the following specific properties on the atomization: **slurries with different particle types and sizes** to determine how the atomization is affected; **fluids having significant elongational viscosity**, which influences the breakup of the ligaments during the secondary atomization; **surface tension**, which was not specifically studied in the previous work; and **molecular weight**, which was also not studied in the previous work. We will use both pressure-swirl and twin-fluid nozzles in this study.

The objective of this part of the research is to measure droplet size distributions generated by different slurries. There are numerous studies on the coal-slurry particles<sup>8,9</sup>. However, there are very limited systematic studies on other types of suspension. The non-Newtonian rheological properties of the suspension play an important role in their disintegration process. We will measure the droplet size distribution for a range of suspension fluids. We plan to use glass and zeolite particles in our study (other suspension fluids of interest to IFPRI members can be considered if suggested). The solubility effect can also be studied using zeolite with a surrogate fluid. The maximum concentration of the particles to be tested depends on the sizes of the particles. For large particle sizes, higher pressures may be needed to push high particle concentration through the nozzle orifice. Therefore, the particle concentration will be increased to as high as our pumping facility will allow.

We have been using Mavlern Spraytec Dropsizer to characterize our sprays. We also use high resolution imaging instruments to obtain close-up images of the atomization process. The same instruments will be used in the proposed study.

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<sup>8</sup> Tsai, S.C., Ghazimorad, K., and Viers, B., "Airblast atomization of micronized coal slurries using a twin-fluid jet atomizer," FUEL, 1991, Vol 70, 483.

<sup>9</sup> Andreussi, P., Tognotti, L., Graziadio, M., and De Michele, G., "Atomization of Coal-Water Fuels by a Pneumatic Nozzle: Characteristics of the Spray," Aerosol Science and Technology, 13, pp 35-46 (1990).

## 2. Correlations for Droplet Size Distribution

Malvern Dropsizer generates volume percentages of droplet size distribution. Figure 6 shows typical histograms generated by Malvern for water and glycerin-water solutions at three different operating pressures. For almost all the test cases, the histograms have one peak with a long tail on the left corresponding to the smaller droplets of less than  $10\ \mu\text{m}$ . The results show that as the injection pressure increases, the volume percentage of large droplets decreases and that of smaller droplets increases (e.g., the peaks move to the left).

The volume percentage is converted to the volume distribution by dividing the height of each bin by the corresponding bin width. Figure 7 shows the volume distributions corresponding to the volume percentage of figure 6. The volume distribution amplifies the changes in the effect of droplet size on volume. Therefore, Fig. 7 shows bimodal distributions for the unimodal distribution of Fig. 6. The small tail in Fig. 6, results in the first peak in volume distribution curves. In order to find a size distributions corresponding to a volume distribution, we first guess a size distribution, and then calculate the volume distribution based on that and compare it with the experimental results. The parameters of the size distribution are then changed to match the experimental data. This procedure is simple for unimodal distributions, but more complex for bimodal distributions.

We will performed similar work for slurry sprays and will develop correlations for both the SMD and also for the droplet size distributions. The PDF of the size distribution provides much more information than just the SMD.

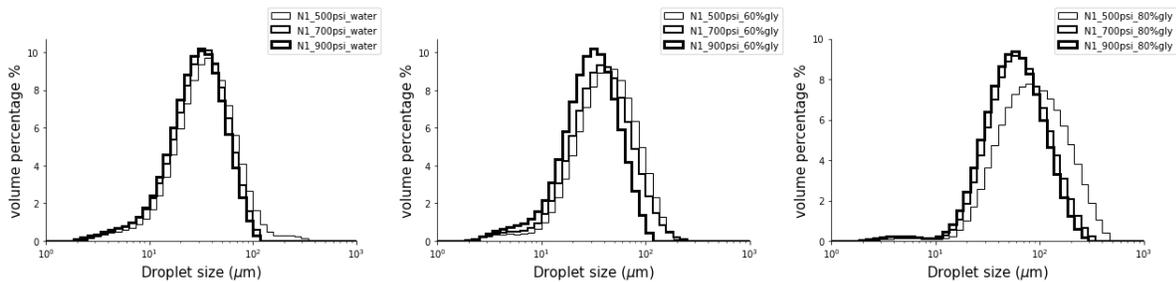


Figure 6: Volume percentage of each testing cases.

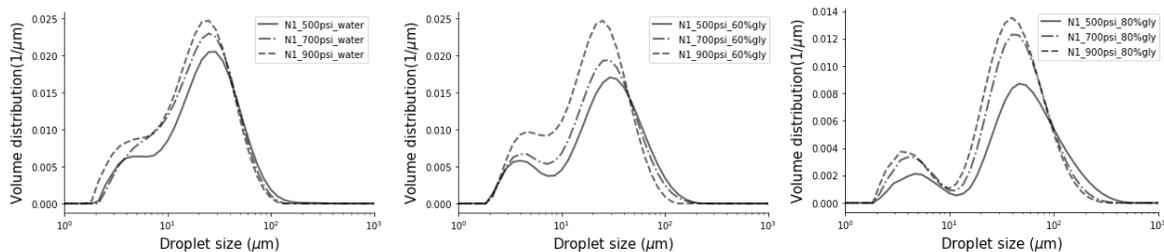


Figure 7: Volume distributions for all testing cases.

### 3. Secondary Breakup of Ligaments of fluids with suspensions

The atomization process for high viscosity fluids is a two step process: primary atomization generating relatively large droplets and ligaments, and secondary atomization further breaking the droplets and ligaments into smaller ones.

Our current near nozzle images of high viscosity liquids show the existence of long ligaments that may persist far downstream of the primary atomization region. This indicates a shortcoming in the traditional definitions of the atomization regions. Typically, the primary atomization results in the formation of droplets, while the secondary atomization is the breakup of those droplets. However, for viscous liquids, rather than only droplets being formed in the primary atomization region, ligaments are also produced that may undergo secondary breakup. Figure 8 shows images downstream of the nozzle where large ligaments still exist.

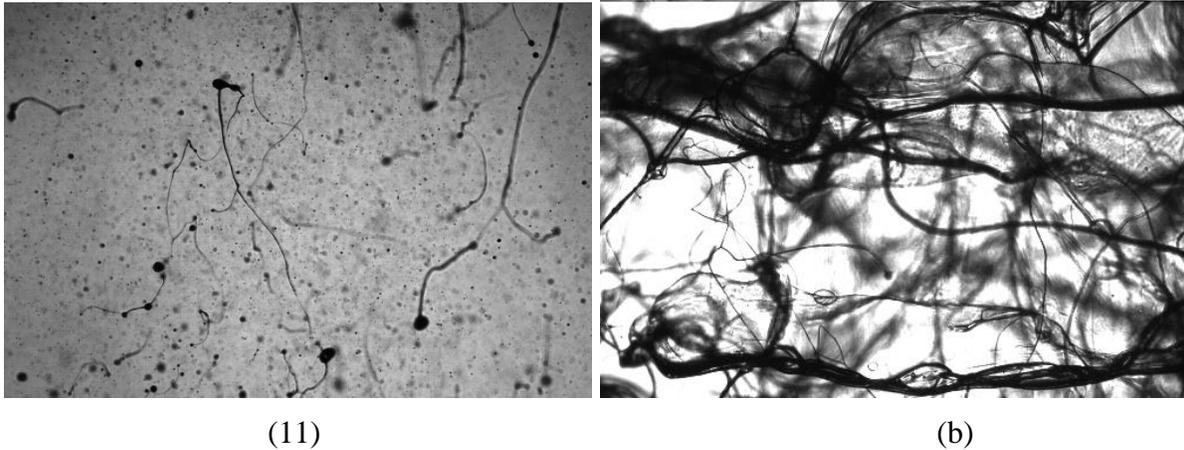


Figure 8: Image of (a) a 0.5% CMC spray at 20-25mm, and (b) 80% Glycerin-water spray at 10mm downstream of the nozzle, showing the persistence of ligaments far downstream of the primary atomization region.

Although the secondary breakup of droplets has been studied extensively in the literature, the secondary breakup of ligaments has not. The question of the secondary breakup of the ligaments poses a few interesting problems, the most relevant of which is “do the ligaments break into small droplets or collapse into large droplets, and does this process occur over the timescales of spray drying?” If the timescale of the ligament secondary breakup is shorter than the drying process, then the resulting powder will have mainly spherical particles. However, if the ligaments do not break within the drying time, then the powder may have long, fibrous particles that decrease the quality of the powder. A better understanding of the secondary atomization of the ligaments will provide a basis for designing sprays to ensure that the ligaments do break within the drying time.

As we have found in our previous work, analyzing these ligaments based on commercial nozzle sprays is highly challenging. Due to the crowding of the images, ligament characterization and temporal tracking become difficult. Furthermore, it is difficult to create controlled experiments of the ligament dynamics, as they are generated somewhat chaotically. By focusing on the fundamental aspect of the secondary breakup of single ligaments, we aim to provide models and understanding of the secondary breakup of ligaments that can be used as a basis for analyzing the complex ligament networks in commercial sprays more effectively.

We propose to study the fundamental behaviours of the secondary breakup of droplets and ligaments. The study of the breakup of droplets will be a direct continuation of our previous IFPRI term,<sup>11</sup> which sought to understand and model the processes that lead to ligament formation and breakup. Some of the images that resulted from this study are shown in Fig. 9.

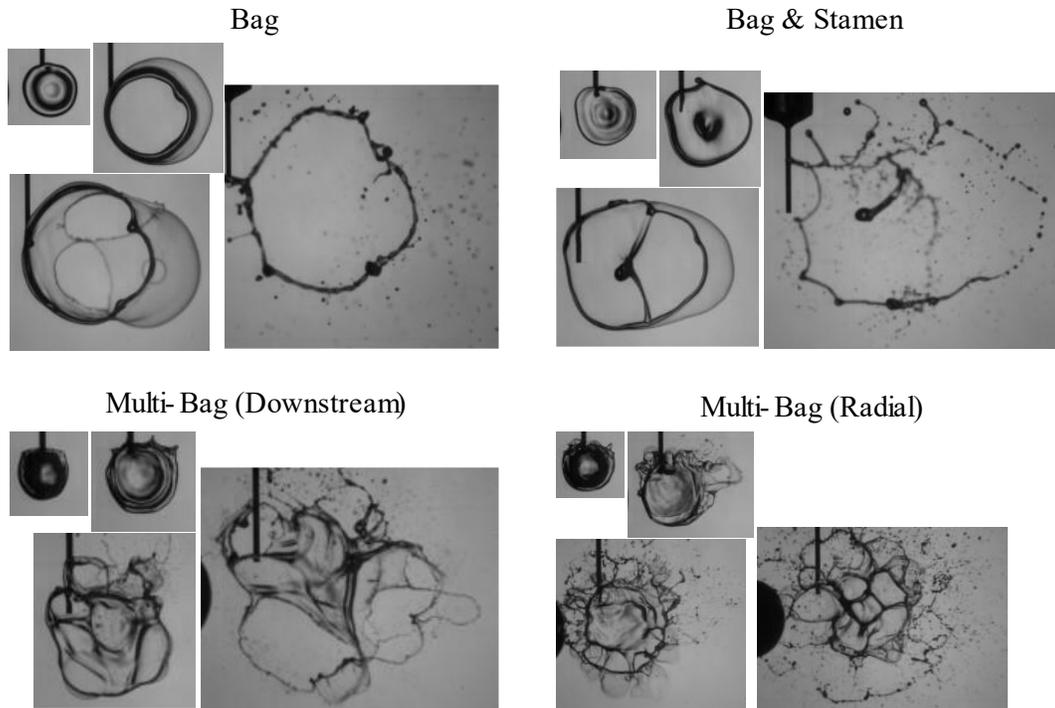


Figure 9: Images of droplet breakup for various morphologies showing the formation of ligaments.

To study the breakup of ligaments, we will generate small diameter ligaments of viscous, polymeric, and slurry fluids that we will expose to a variety of conditions such as a gaseous cross flow to study their breakup. We will use both an elongated liquid bridge and also inject a liquid jet into a gaseous cross flow. These methods are illustrated in Figure 10. The dynamics of the ligaments will be captured using high-speed video so that the temporal behaviour of the ligaments can be resolved. Key factors to be determined in these experiments are the effects of the extensional viscosity on the breakup and the timescales of the ligament breakup in addition to the sizes generated. These factors are expected to be related by the critical conditions required for the ligament breakup or collapse, thus, we will aim to determine what the critical conditions are and what parameters affect or determine them.

In these experiments, we may also study the effects of elevated ambient temperatures. Since the primary atomization occurs rapidly for two-fluid nozzles, there is no significant heat transfer to the fluid during the primary atomization. However, since the ligaments in viscous sprays persist for a long time after they are formed, their dynamics may be affected by the elevated temperatures, either in the changing of the liquid properties due to heating or by the evaporation of the solvents in the liquid.

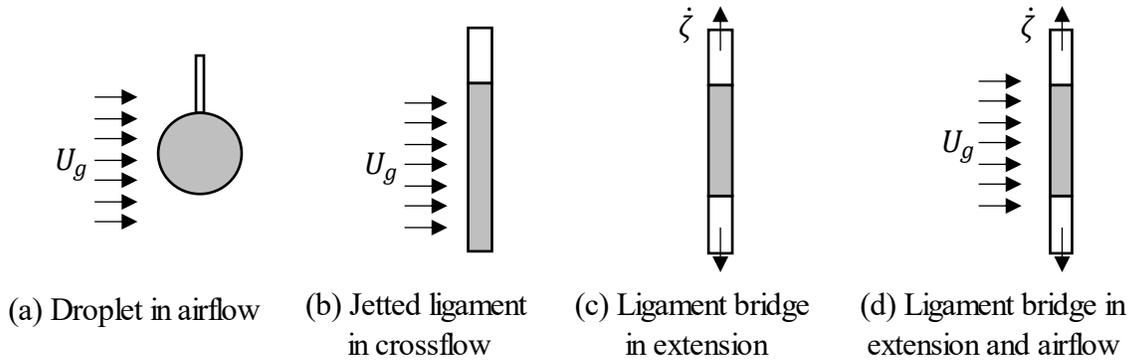


Figure 10: Illustrations of experimental setups for study of droplet and ligament breakup

#### 4. Perforation Based Model for Pressure-Swirl Nozzles

Based on studying the near nozzle images, we have developed a new atomization model for the pressure-swirl nozzles. The new model is based on a combination of a Kelvin-Helmholtz wave (KH) and a sheet perforation. In order to extend this model to fluids with suspensions, we need to determine the breakup length of the swirling sheet of such fluids, characterize the diameter of the ligaments formed, and determine the number and frequency of the perforations on the sheet. These are planned for the proposed period.

A perforation-based atomization model as depicted in Fig. 12 is proposed. The liquid sheet breaks up into droplets in the following steps:

1. As the high-speed swirling conical liquid sheet exits the orifice, surface waves are generated due to the Kelvin-Helmholtz (KH) instability.
2. As the liquid sheet moves downstream, the amplitude of wave crests become larger, and eventually break the sheet into segments.
3. Each segment is perforated based turbulent structures.
4. The perforation process results in a set of rectangular cells, that have span-wise and stream-wise sides.
5. The cells eventually breakup forming span-wise and stream-wise ligament with different diameters, which eventually breakup to result in droplet with different diameters.

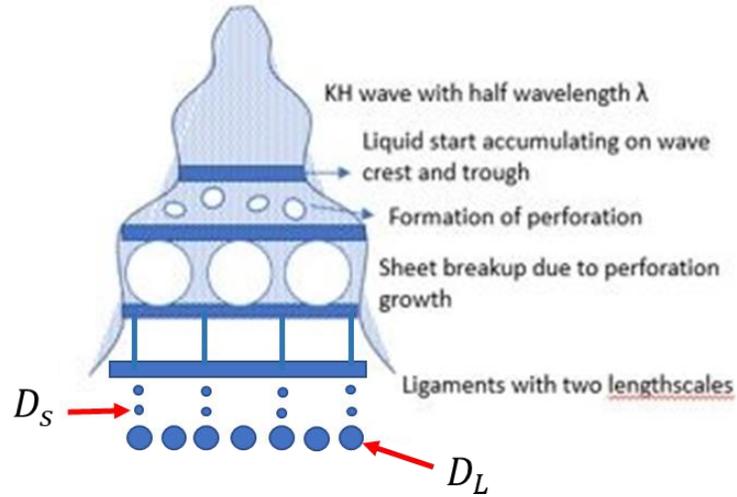


Figure 12. A schematic of the perforation-based atomization model

This breakup model provides two different length scales, which take up different weight percentages: One based on the thickness of the thin sheet between the crests and troughs of the waves ( $D_s$ ) with a total mass of  $m_s$ , and one based on the diameters of the crests and troughs ( $D_L$ ) with a total mass of  $m_L$  and these can be given as

$$D_L = \sqrt{\frac{4m_c}{\pi^2 D_b}}, D_s = \sqrt{\frac{4(m - m_c)}{\pi n \lambda}}$$

where  $D_b$  is the breakup length,  $n$  is the number of perforations on the thin sheet and  $\lambda$  is the distance between the wave crests.

We will model the spray size distribution using two size distribution (PDF) each for one of the spray length scales. In order to complete this model, we need to determine correlations for the characteristic sizes of both the spanwise and the streamwise ligaments, in terms of the fluid properties and the operating conditions. In order to develop a correlation that can be generalized to a wider range of fluids and operating conditions, the following tasks are planned

- Determine the frequency of perforations on the thin sheets with fluids with suspensions.
- Determine how much mass is accumulated at the wave crest
- Develop a theory for how long the sheet can stretch in Newtonian case
- Determine the breakup length of the fluids sheets with suspensions

These data will be combined with the ligament breakup model (Rayleigh-Plateau instability) for non-Newtonian fluids to generate a model for droplet size distribution. More detailed information on the testing cases and their results including droplet size distribution and near nozzle images are provided in the final report.

## 6. Physics Based Model for Twin-Fluid Nozzles

Two key advantages to the atomization models that we developed for twin-fluid nozzles in our previous project term are the ability to relate the rate of the deformation to the breakup and to predict the formation of ligaments in the spray. This allows for the opportunity to directly include rheological effects such as extensional viscosity in our models and to support their development by using what we learn in our experiments on the breakup dynamics of the ligaments and the spray characterization. Our previously developed model for the atomization of twin-fluid nozzles is comprised of the following processes: KH waves form on the jet that exits a nozzle and cause small droplets to break off its surface; the liquid core in the jet (the liquid left behind after KH droplets break off) goes through flapping instability, forming larger droplets with length scale of the flapping wavelength, and the secondary atomization process breaks the large droplets to smaller droplets. The breakup model is related to the rate of deformation of the waves. The advantages of modelling the spray in this way is the isolation of the slow wave growth from the fast wave breakup and the prediction of ligaments in the breakup, which previous models do not consider. While the development of the KH and flapping instabilities will be somewhat affected by the rheological fluids, the greater effect is expected to be on their breakup, especially when the extensional viscosity is high. We aim to extend our existing models to include these effects.

Our currently developed model for the atomization of the twin-fluid nozzles comprises of the following processes:

- High-speed gas flow over the liquid breaks off small droplets from the surface of the liquid jet issuing from the nozzle based on Kelvin-Helmholtz (KH) instability. The droplet sizes are related to the wavelength of the fastest growing KH waves.
- The liquid core in the jet (the liquid left behind after KH droplet break offs) goes through flapping instability, forming larger droplets with length scale of the flapping wavelength.
- The secondary atomization process breaks the large droplets to smaller droplets. The following secondary atomization model is developed: The high-speed gas flow deforms the front of the droplet into a disk shape. The volume-fraction in the frontal disk determines the breakup morphology:
  - A model is developed that can predict the following morphologies: single bag and multibag breakup, as well as bag and stamen breakups. The breakup is related to the rate of deformation of the droplet.
  - Each disk shape droplet forms a bag with a relatively thick rim. The bag grows and eventually breaks forming small droplets, and the rim breaks forming larger droplets.
  - Rims are analogous to the ligaments seen in high-viscosity spray breakup.
- The final spray droplet size distribution is predicted based on the primary and the secondary atomization models.

The advantages of modelling the spray in this way is the isolation of the slow wave growth from the fast wave breakup and the prediction of ligaments in the breakup, which previous models do not consider. While the development of the KH and flapping instabilities will be somewhat

affected by the rheological fluids, the greater effect is expected to be on their breakup, especially when the extensional viscosity is high. Using what we learn about the breakup dynamics of the ligaments, we aim to extend our existing models to include these effects. Further details on the developed twin-fluid nozzle is given in the final report.