**DESIGN OF ATOMIZERS AND BURNERS FOR
COAL-WATER SLURRY COMBUSTION**

QUARTERLY ACTIVITY REPORT

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Abstract

The Falling Droplet device was used to measure the extensional viscosity of a variety of aqueous polymer solutions. These solutions were atomized with the miniature “inverse” twin-fluid atomizer. Droplet size measurements were made with a Malvern laser diffraction particle sizing device. Droplet sizes measured did not rank strictly according to either low-shear rate shear viscosity or extensional viscosity.

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Progress Report for Period 10/1/99 - 12/31/99

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Executive Summary

The Falling Droplet device was used to measure the extensional viscosity of a variety of aqueous polymer solutions. These solutions were atomized with the miniature “inverse” twin-fluid atomizer. Droplet size measurements were made with a Malvern laser diffraction particle sizing device. Droplet sizes measured did not rank strictly according to either low-shear rate shear viscosity or extensional viscosity.

Introduction

This report reviews the activities and results of the period 1 October 1999 to 31 December 1999. Two principal activities were undertaken during this period. The first activity undertaken in this period is the measurement of the extensional viscosity of aqueous polymer solutions with the falling droplet instrument. The second activity is the measurement of droplet sizes in sprays of the polymer solutions measured in the first activity. The results of this work will be presented at the 12th annual meeting of the Institute for Liquid Atomization and Spray Systems, Americas section in Indianapolis, Indiana, on 18-21 May 1999. The extended abstract submitted to the conference organizers is appended to, and forms the bulk of, this report.

Previous work has shown that droplet size (both mean size and size distribution) does not always correlate with shear viscosity in non-newtonian fluids. It is suspected that extensional viscosity plays a dominant role in air-assisted atomization. Extensional viscosity is similar to the better-known shear viscosity, but relates the extension strain rate to the tensile stress rather than shear strain rate and shear stress. In the falling droplet technique, a droplet forms at the bottom of a vertical capillary tube. As the droplet falls, a thin filament is formed between the droplet and the capillary tube. Laser light sheets and photodiodes are used to measure the diameter and length of the filament, as well as the velocity and acceleration of the droplet by light attenuation. The resulting instantaneous ratio of tensile stress to extension rate gives a measure of the extensional viscosity of the fluid, in the form of a growth function versus time.

In the previous quarter, phase/Doppler measurements of drop sizes within a polymer solution spray showed approximately a 10% increase over a spray of water at the same conditions. In this quarter, the sensitivity of drop size to molecular structure will be examined as well.

Results and Discussion

Measurements of extensional viscosity were made with three aqueous polymer solutions: polyacrylimide (“E10”), xanthan gum (“XG”) and poly-ethylene oxide (“PEO”). As before, the ratio of the maximum observed extensional viscosity to the shear viscosity ranges from approximately 200 to nearly 10,000.

Droplet size measurements were made with a Malvern Model 2600 laser diffraction particle sizer. As shown in the attached paper, the droplet sizes do not correlate directly with either the extensional or the shear viscosities.

Conclusion

Utilizing the Falling Droplet instrument and the Triple-Concentric atomizer, it has been shown that extensional viscosity does appear to play an important role in drop formation during atomization of dilute polymer solutions. The exact mechanisms remain to be determined. Extensional viscosity is not the only parameter of importance, however, since the drop size ranking does not match one-to-one with extensional viscosity ranking.

Measurement of Extensional Viscosity and Its Effect on Atomization

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Introduction

The “Falling Droplet” instrument for measuring extensional viscosity of viscoelastic aqueous polymer solutions was used to investigate the effect of extensional viscosity on air-assisted atomization. Three different polymer solutions were examined.

Background

Many empirical, predictive equations exist for relating the physical properties of a liquid and the operating conditions and geometry of an atomizer to some measure of the atomization, usually the Sauter Mean diameter, or D_{32} . For example, Equation 1 [Nukiyama & Tanasawa, '39] is one of the first such equations for a simple air-assisted spray.

$$D_{32} = \frac{0.585}{U_R} \left(\frac{s}{r_L} \right)^{0.5} + 53 \left(\frac{m_L^2}{sr_L} \right)^{0.225} \left(\frac{Q_L}{Q_A} \right)^{1.5} \quad (\text{Eq. 1})$$

The liquid viscosity, m_L , affects the drop size observed in this case. However, the inclusion of the viscosity in this case clearly represents a single-valued Newtonian viscosity. Even in this case, it should be noted that the viscosity plays a lesser role than the surface tension, relative velocity, and relative flow rates of the air and liquid.

When the spray breakup processes are studied in detail, however, it is evident that, in the breakup region, the velocity gradients within the liquid necessary to generate shear stresses which could lead to liquid breakup are not present. Once beyond the initial breakup region, large droplets and “blobs” of fluid are stretched by aerodynamic forces. Atomizers which initially generate ligaments, such as rotary and some effervescent atomizers, also produce droplets by stretching, necking, and breaking of the ligaments.

The nature of the breakup of ligaments into droplets is predominantly a stretching motion rather than a shearing motion of the liquid within the ligament. For this reason, extensional viscosity is a critical

liquid property affecting the atomization of liquids such as high-molecular weight polymers. These liquids are important in a number of applications, including coatings and agricultural pesticide sprays.

All liquids exhibit an extensional viscosity. Newtonian liquids have an extensional viscosity which is approximately three times the shear viscosity. Non-newtonian liquids, such as polymer solutions in water, display a more complex behavior. In these fluids, shear viscosity is a function of shear rate, total shear, and the time history of shear. Extensional viscosity is also a function of extensional strain rate, total strain, and time history of strain. The ratio of the extensional to shear viscosity (the Trouton ratio) can reach values in the tens of thousands for dilute solutions of high molecular weight polymers.

For this work, the extensional viscosity “growth function” was measured for three different aqueous polymer solutions. The three materials are polyacrylimide, xanthan gum, and poly-ethylene oxide. They will be referred to as E10 (the manufacturer’s designation), XG, and PEO, respectively, henceforth. E10 is a long, flexible-chain molecule with locally charged regions along its length. For this reason, the solution of E10 is prepared in a solvent of 0.1M sodium chloride, so that the ions in the solvent will screen the charges on the molecules, preventing agglomeration of the particles. XG is a rigid, highly branched molecule. PEO is a semi-flexible molecule without molecular agglomeration problems of the E10. The molecular weights of the materials are: E10- approximately 20 million; XG- approx. 2 million, and PEO- approx. 1 million. Each material was used to create an aqueous solution containing 0.1% by weight of the polymer.

Theory

As the droplet falls, the filament experiences a tensile principal stress in excess of the radial principal stress. This excess stress, the first normal

stress difference, divided by the strain rate experienced by the filament, yields an extensional property of the polymer solution with units of viscosity — the apparent extensional viscosity.

The strain rate experienced within the filament is given by:

$$\dot{\epsilon} = \dot{L}/L = V/L \quad (\text{Eq. 2})$$

Here, $\dot{\epsilon}$ is the strain rate, L is the filament length, and V is the droplet velocity.

The total axial stress within the filament as it falls is governed by gravity, drop acceleration, surface tension, aerodynamic drag, and extensional viscosity:

$$T_{zz} = \frac{m(g-a)}{\pi r^2} - \frac{2s}{r} - \frac{F_D}{\pi r^2} \quad (\text{Eq. 3})$$

Here, T_{zz} is the axial tensile stress, m is the mass of the droplet, a is the acceleration of the droplet, s is the surface tension of the liquid, r is the radius of the filament, and F_D is the aerodynamic drag force on the droplet (estimated by a simple Stokes approximation to flow over a sphere).

The presence of surface tension creates an internal pressure within the filament. The radial stress caused by surface tension is given by:

$$T_{rr} = -\frac{s}{r}. \quad (\text{Eq. 4})$$

The difference between the axial and radial stresses, the first normal stress difference, is used in the definition of extensional viscosity. The extensional viscosity, h_E , is defined as the ratio of the first normal stress difference to the strain rate:

$$h_E = \frac{T_{zz} - T_{rr}}{\dot{\epsilon}}. \quad (\text{Eq. 5})$$

In order to measure the extensional viscosity with the falling droplet technique, one must determine the mass of the droplet, the velocity, and acceleration of the droplet as it falls, the drag on the droplet, and the length and diameter of the filament as functions of time during the extension of the filament. The mass is known from the density of the solution, the flow rate, and the frequency of droplet formation:

$$m = \frac{\rho Q}{n/\Delta t} \quad (\text{Eq. 6})$$

(where ρ is the solution density, Q is the flow rate, and n is the number of droplets in time, Δt). The filament length is determined from the position of the droplet. The position, velocity, and acceleration of the droplet and the diameter of the filament must be recorded during the process.

Apparatus

Falling Droplet Extensional Rheometer

The filament to be measured is formed by a pendant droplet at the end of a vertical capillary tube, as shown in Figure 1. The liquid is supplied by a stepping motor-driven syringe pump at very low flow rate to the capillary tube, such that one droplet forms and falls approximately every ten seconds.

The falling droplet extensional rheometer utilizes the uniaxial stretching, as described above, of a liquid filament between a droplet and the capillary tube from which it formed.

The falling droplet extensional rheometer employs laser light sheets to perform its measurements.

Figure 1: Falling Droplet Apparatus Schematic

Figure 1 shows the layout of the instrument. Two separate 15 mW Helium-Neon lasers are used. The first laser beam passes through a set of optical-quality cylindrical lenses to produce a wide, horizontal laser sheet. The stretching filament intersects this laser sheet, casting a shadow through the 4 mm-diameter aperture. The image of the filament's shadow is magnified and focused onto a photodiode. The reduction in total incident light flux onto the photodiode depends on the width of the shadow and, hence, the filament. The second laser beam passes through a glass rod to produce a very wide, vertical laser sheet. The filament is parallel to this laser sheet and the vertical array of photodiodes. As the droplet falls, it casts a shadow on each diode in sequence, allowing the elapsed time between the droplet's passing of any two adjacent diodes allows the calculation of the droplet's trajectory.

The photodiode used for the filament diameter measurement is calibrated using a high-magnification CCD camera. Water jets from very fine glass nozzles were imaged with the CCD camera and measured with the photodiode. The resulting calibration relationship is shown in Figure 2. Since the filament cannot be larger than the diameter of the capillary, the calibration curve's upper limit is equal to the outside diameter of the capillary tube.

All of the photodiodes are connected to a computer data acquisition system. The output of the photodiodes is recorded and stored for analysis at rates of up to 1000 Hz. Subsequent analysis produces simultaneous measurements of filament diameter and droplet position, velocity, and acceleration as functions of time during the fall of a single droplet.

Malvern 2600 Laser Diffraction Particle Sizer

The spray measurements were made with a Malvern Instruments Model 2600 laser diffraction instrument. The measurement volume is formed by the intersection of the spray cone with a 9 mm-diameter laser beam. The droplets within the measurement volume diffract the light at angles related to their size, in accordance with the Fraunhofer theory.

The diffracted light is collected by a Fourier lens which focuses all light which enters the lens at a given angle onto the detector plane at a fixed radial distance regardless of the location where the light enters the lens. A series of concentric semi-circular photodiodes measure the radial light intensity distribution that corresponds to the angular distribution produced by the diffraction of laser light by the droplets. The instrument computes the light distribution generated by an initial, assumed, volume distribution. The reported volume distribution is refined by an iterative process which seeks the minimum difference between the observed and calculated radial light distributions.

Since many thousands of droplets contribute to each measurement of the light distribution and the light distribution is measured thousands of times, the laser diffraction instrument provides an accurate and rapid assessment of the overall volume distribution of the droplets in a spray. The large measurement volume provides a global ensemble average of the whole spray.

Results

Polymer Solution Properties

The shear viscosity profiles were measured over two decades of shear rate in order to compare the relative effect of shear viscosity and extensional viscosity on the atomization of the polymer solutions. The shear viscosities are shown in Figure 3. The surface tension must be known for the computations

involved in the extensional viscosity measurements. Figure 4 shows the surface tension of the three materials tested and pure water.

Extensional Viscosity Growth Functions

The extensional viscosity of each solution was measured with the falling droplet instrument. The direct measurements consist of the droplet position versus time and filament diameter versus time (see Figures 5 and 6). The droplet position is measured relative the capillary tube exit, so the droplet position is equal to the filament length.

The distinct “shoulder” seen in the length-vs.-time plot with a logarithmic time scale appears to correspond to the change from necking (and combined strain and shear flow) to filament stretching (and uniaxial extensional flow). The droplet position with time is fit with a fourth-order polynomial (see Figure 5). This polynomial is differentiated twice to obtain the droplet velocity and acceleration as functions of time. The droplet velocity is required for determining the strain rate. The acceleration is used in the force balance to determine the “net” weight of the accelerating droplet.

Following the procedure described in Equations 2 through 6, above, the instantaneous extensional viscosity is calculated as a function of time. This curve is called the extensional viscosity growth function. Figure 7 shows the extensional viscosity growth functions for all three polymer solutions.

Droplet Size Measurements

All three solutions were atomized in a simple parallel-flow twin-fluid atomizer. The solutions were all atomized with a liquid flow rate of 50 mL/min. The indicated air flow rate of 50 L/min at 90 kPa gage pressure resulted in a relative velocity between the air and liquid of approximately 300 m/s. The air-to-liquid mass flow ratio was 1.65. The volume-classified diameters are given in Figure 8.

The volume distributions of water and E10 are shown in Figure 9.

Conclusion

It is readily apparent that the extensional viscosity of aqueous polymer solutions has a large influence on the atomization. According to “classical” size prediction equations, of the type presented earlier, the xanthan gum (XG) solution should have produced the worst (largest drop sizes) atomization due to its high shear viscosity. However, the extensional viscosity of the rigid xanthan gum molecules is much lower than that for either the polyacrylimide E10 or the poly-ethylene oxide (PEO), whose flexible molecules can become intertwined, strengthening the liquid considerably when under tension. Although the extensional viscosity of E10 does not reach as high a value as PEO, it undergoes much greater total strain before the filament breaks. Further study is needed to determine the relative effects of molecular weight and concentration on the extensional viscosity and

its effect on atomization of aqueous polymer solutions.

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