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**The Physics of Non-Newtonian Liquid Slurry Atomization
Part II: Twin-Fluid Atomization of Non-Newtonian Liquids
Contract Number: DE-FG22-92PC92152**

**First Quarterly Technical Report
Period of Performance: 1/1/94- 3/31/94**

**Adel Mansour and Norman Chigier
Mechanical Engineering Department
Carnegie Mellon University**

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THE PHYSICS OF NON-NEWTONIAN LIQUID SLURRY ATOMIZATION

PART2: TWIN-FLUID ATOMIZATION OF NON-NEWTONIAN LIQUIDS

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ABSTRACT

The changes in the physical processes of atomization as a result of adding a high molecular weight polymer in low concentrations to liquid have been studied. Both Newtonian and non-Newtonian liquids were investigated with particular emphasis on the non-Newtonian rheological characteristics. It was found that viscoelastic liquids are much more difficult to atomize than viscoinelastic liquids. Viscoinelastic liquids showed a breakup behavior similar to that of water sprays. Viscoelastic materials showed remarkably different breakup patterns. The ligaments were seen to undergo a very large stretching motion before they breakup, resulting in long threads of liquid attached to droplets. The normal stresses developed in viscoelastic materials are much higher than their associated shear stresses. Consequently, the development of the large normal stresses appears to be the most important rheological mechanism that inhibits breakup.

INTRODUCTION

Airblast atomization of viscous non-Newtonian liquids is carried out using a co-axial twin-fluid atomizer [1]. As the atomizing air interacts with the liquid jet downstream from the nozzle exit, waves form on the surface of the liquid jet. As a result, the liquid jet sheds ligaments which rapidly collapse into droplets. The atomized drop sizes can be described in terms of three dimensionless groups, namely the liquid/air mass ratio (M_l/M_a), Weber ($We = \rho_g D U_g^2 / \sigma$) and Ohnesorge ($Z = \mu_l / (\rho_l \sigma D)^{1/2}$) numbers in simple forms whose exponents and coefficients are determined by least squares fit to the experimental data. A phase Doppler particle analyzer was used for the determination of particle size and velocity. A digital spray analyzer was used for die-swell measurements. The Non-Newtonian liquids selected for the experiment were aqueous solutions of Xanthan gum (supplied by Aldrich Chemical Co.) and Polyacrilamide E10 (supplied by Allied Colloid Co.). Tests were performed on 0.05, 0.10, 0.15, 0.20, 0.30% concentrations by mass for the Xanthan Gum polymer, and 0.0125, 0.025, 0.05, 0.1, 0.15, 0.30, and 0.50% concentrations by mass for the Polyacrilamide E10.

RHEOLOGICAL MEASUREMENTS

Shear Viscosity Measurements The rheology of the polymeric solutions was modeled by the Power Law Model.

$$\tau = k |\dot{\gamma}|^{n-1} \dot{\gamma} \quad (1)$$

where τ is the shear stress, $\dot{\gamma}$ is the shear rate, n is the power law index and k is the consistency index. Viscosity measurements at low shear rates ($\dot{\gamma} < 13.4 \text{ s}^{-1}$) were performed using the Brookfield viscometer. To obtain viscosity data at higher shear rates ($\dot{\gamma} > 13.4 \text{ s}^{-1}$), capillary tube viscometers were used. A large and small capillary tube were operated in the range of shear rates, $10 < 8V/D < 2000 \text{ s}^{-1}$ and $1800 < 8V/D < 20000 \text{ s}^{-1}$, respectively. In order to eliminate the errors associated with the velocity adjustment region in the entrance and exit of the capillaries a differential pressure measurement system was utilized. A highly accurate differential pressure transmitter was used to obtain the pressure drop across two points on each capillary tube. The large and small tube diameters were 4.39 mm and 1.80 mm, with a length to diameter ratio of 80 and 189, respectively.

The apparent viscosity data for the Xanthan and E10 solutions is shown in Figs 1 and 2, respectively. In the mid-range of shear rates ($10 < \dot{\gamma} < 3500 \text{ s}^{-1}$), the apparent viscosity conforms to a power-law function. In the highest range of shear rates, $\dot{\gamma} > 3500 \text{ s}^{-1}$, the apparent viscosity becomes nearly constant and the solutions tend towards Newtonian behavior, with viscosities relatively close to that of water. The degree of shear thinning exhibited by each solution was characterized by the apparent viscosity power law index in the mid-range of shear rates. For each concentration, the power law

parameters and apparent viscosity limiting values are summarized in Table 1. η_0 is the viscosity value that is reached in the limit as the shear rate tends towards zero. The value η_∞ is the apparent viscosity value which is reached as the fluid becomes Newtonian at high shear rates. It is observed from Table 1 that n is a strong function of the solution concentration with a decreasing value of n for increasing values of concentration.

Table 1: Power-Law Parameters and Limiting Viscosities for Xanthan and E10 solutions

Conc. (%)	k (cP-S ^{$n-1$})	n	η_0 (cP)	η_∞ (cP)
0.05XG	28.6	0.604	30	1.6
0.10XG	110.2	0.487	115	1.7
0.15XG	321.0	0.383	822	1.8
0.20XG	664.4	0.333	2416	2.2
0.30XG	1804.8	0.248	14761	2.8
0.0125E10	1.9	0.923	2.10	1.2
0.025E10	3.2	0.855	3.40	1.3
0.05E10	7.3	0.757	7.912	1.5
0.10E10	18.7	0.700	20.21	2.2
0.15E10	84.4	0.595	93.00	3.9
0.30E10	517.7	0.444	1243	6.3
0.50E10	1644.4	0.387	9342	12.1

Die-Swell Measurements The phenomenon of die-swell is attributed to the relaxation of the normal stresses. In order to calculate the normal stresses from die-swell, the following relationship proposed by Metzner and co-workers [2] is used.

$$N_1 = \tau_{11} - \tau_{22} = \frac{\rho D^2 (8V/D)^2}{64 n'} \left\{ (n'+1) \left(\frac{3n+1}{2n+1} \right) - \left(\frac{D}{d_j} \right)^2 \left[n'+1 + \frac{d \log D/d_j}{d \log 8V/D} \right] \right\} \quad (2)$$

Measurements of the die-swell ratio (D/d_j) along with the volumetric flow rate are used to evaluate the normal stress differences. A digital image analyzer with a close-up lens was positioned near the outlet of the capillary tube to measure the jet diameter. The jet diameter and volumetric flow rate were measured at different flow conditions. Figure 3a and 3b show a typical image of the die-swell phenomenon for 0.3% Xanthan and 0.5% E10 solutions respectively at a shear rate $8V/D = 5246 \text{ S}^{-1}$. In order to calculate the normal stress difference from die-swell measurements, a reasonably accurate estimate of the shear dependency as a function of $8V/D$ is needed. Over the range of $8V/D$ for which die-swell measurements were made ($300 \text{ sec}^{-1} < 8V/D < 5000 \text{ sec}^{-1}$), the shear dependency of the polymeric solutions is represented by the values of n shown in Table 1. The calculated values of the first normal stress difference N_1 as a function of shear rate for the Xanthan and E10 solutions are given in Fig.4. The single most important difference between the Xanthan solutions and the E10 solutions is the much higher levels of normal stresses developed in the E10 solutions when compared to the Xanthan gum solutions.

AIRBLAST ATOMIZATION

Dependence of SMD on Fluid Viscosity or the Ohnesorge number. Local point measurements of the Sauter mean diameter were made at $r = 0$ and $x/D_a = 20$, where x is the downstream distance from the injector, r is the radial distance, and D_a is the annular air jet diameter. The variation of the Sauter mean diameter of droplets with changes in the air/liquid ratio is shown in Fig. 5. Throughout this experiment the air flow rate was maintained constant at 12.89 g/s and the liquid flow rate was varied between 2.27 g/s and 27.21 g/s. This corresponds to an air velocity of 93.4 m/s and a liquid velocity varying from 0.310 to 3.72 m/s. Atomization data for the highest concentrations of Polyacrilamide E10 solutions (i.e., 0.05, 0.1, 0.15, 0.3, 0.5) are not included in Fig. 5. For these concentrations the liquid did not atomize. Long threads of liquid interspaced by droplets were formed and the rejection rate due to phase errors exceeded 30% for the highest concentration.

Figure 5 shows that, although the shear viscosity data for the 0.025% solution of E10 is orders of magnitude lower than that of the 0.3% solution of Xanthan at all values of the shear rate,

the atomization quality of the Xanthan solution is substantially better at all values of the liquid flow rate. The single most important difference between the Xanthan solution and the E10 solution is the much higher levels of normal stresses developed in the E10 solution. The large die-swell ratio (d_j/D) that has been observed for the E10 solutions in the capillary tube experiment suggests that the extensional viscosity levels for the E10 solutions are large and that the extensional viscosity is probably the most important rheological mechanism that inhibits breakup. Figure 5 also shows the significant reduction in droplet size that can be obtained by increasing the air/liquid mass ratio.

The effects of solution rheology on atomization are first examined in order to determine which viscosity should be used to define the Ohnesorge number. The spray mean drop size data shows that the SMD increases with apparent viscosity of the fluid. The liquid flow rate was varied between 2.27 g/s and 27.21 g/s corresponding to an apparent shear rate ($8V/D$) inside the injection tube varying between 786 S^{-1} and 9754 S^{-1} . Due to the very high shear rates encountered inside the atomizer, it was decided to correlate the data in terms of η_∞ . The reason for maintaining the air flow rate constant was to remove any extraneous effects of the additional shear introduced by the air flow on the fluid viscosity. The data in Fig. 5 was fitted to the following equation:

$$\text{SMD} \propto Z^{X_z} \quad (3)$$

Only water and the 0.05, 0.1, 0.15, 0.2 and 0.3% solution of Xanthan are considered for the correlation. The data consisted of 19 data sets representing the various variations of liquid flow rate, and each data set contained 6 points representing the various polymer concentrations. Each of the data sets was fitted to Eq. (3) separately to show the functional dependence of the SMD on fluid viscosity. The power-dependency X_z of the Sauter mean diameter on fluid viscosity, in the limit of infinite shear rates, is shown in Fig. 6. In Fig. 6 X_z is plotted as a function of the apparent shear rate ($8V/D$) inside the injection tube. It is seen in Fig. 6 that the exponent X_z is not constant and is a function of the shear rate. At the low end of shear rates ($8V/D < 3600 \text{ S}^{-1}$), the exponent X_z decreases with shear rate. Beyond $8V/D = 3600 \text{ S}^{-1}$, X_z remains sensibly constant. The variations of X_z with respect to the shear rate is relatively simple to explain in terms of the apparent viscosity of the fluid. At the low end of shear rates, the liquid inside the injection tube is sheared to a consistency which intermediate between η_0 and η_∞ . The use of η_∞ to correlate the SMD data with is not appropriate. At shear rates above 3600 S^{-1} , the liquid inside the atomizer is sheared to a consistency η_∞ for all concentrations and the use of η_∞ to correlate the SMD data with is indeed the most appropriate choice. The average value of X_z at shear rates greater than 3600 S^{-1} is $X_{z,av} = 0.417$. Attempts at correlating the data in terms of a mass averaged shear viscosity, which is seemingly a more appropriate choice, were unsuccessful. The reason for this is that correlation of the data in terms of a mass averaged shear viscosity requires knowledge of the velocity profile at the exit of the injection tube, which is not known a priori. Even if the velocity profile were known, correlating the data based on a mass averaged shear viscosity would make the problem untractable since the mass average shear viscosity is not single valued; it varies with shear rate. Figure 6 shows that SMD increases in proportion to the increase of the apparent viscosity. Note that the correlation is valid for viscoelastic liquids only (Xanthan solutions). The coefficient of dependence in the high shear rate region ($X_z = 0.417$) is not far removed from previous results with Newtonian liquids.

The air flow field in airblast atomization is unknown. The air flow imparts additional shear on the liquid column before breakup. Although, at low liquid flow rates, the liquid inside the injector might not have been sheared to a consistency η_∞ , the additional shearing imparted to the liquid due to its interaction with the air flow can result in additional thinning of the liquid before breakup. Thus, a second set of experiments were carried out to investigate if the additional shear, on the liquid column before breakup, introduced by the air flow has any effect on the power dependency (X_z). The liquid velocity was maintained constant at $M_l = 7.46 \text{ g/s}$, corresponding to $8V/D = 2622 \text{ S}^{-1}$ and the air mass flow rate was varied between 8.51 g/s and 23.64 g/s corresponding to an air velocity in the range of $61.66 \text{ m/s} < V_a < 171.278 \text{ m/s}$. This resulted in Weber number variation between 189 and 1461. This experiment was carried out using pure water

and the 5 Xanthan solutions. The data consisted of 17 data sets representing the various variations of air flow rate, and each data set contained 6 points representing the various polymer concentrations. Figure 7 shows that the SMD decreases with increase in the Weber number. Each of the data sets was fitted to Eq. (3) separately to show the functional dependence of the SMD on fluid viscosity at variable air flow or Weber number. Figure 8 shows that X_z is insensitive to the variations in air mass flow rate or air velocity. It was found that the power dependency X_z remains almost the same with $X_{z,av} = 0.420$.

Dependence of SMD on $1+M_1/M_2$ The power-dependence on $(1 + M_1/M_2)$ is examined by plotting the SMD data as a function of $(1 + M_1/M_2)$ at constant Weber number. Figure 9 shows the same data displayed in Fig. 5 as a function of $(1 + M_1/M_2)$ plotted using a logarithmic plot. It should be noted that the slope of the line shown in Fig. 9 is effectively unity. Therefore the power of $(1+M_1/M_2)$ in the SMD equation, should be unity.

Dependence of SMD on Weber Number Experiments were conducted to examine the influence of Weber number on the Sauter mean diameter. The effect of Weber number on droplet SMD was examined by varying the air exit velocity. No variations of surface tension were undertaken. The data presented in Fig. 7 was used to establish the functional dependence of the SMD on the Weber number. The power-dependence on Weber number is examined by plotting the SMD data as a function of Weber number at constant $(1 + M_1/M_2)$. In the previous sections we have established that the effects of the air flow on the viscous behavior of the polymeric solutions are negligible. Recall that the power dependency X_z of the SMD on η_∞ was nearly constant, irrespective of the Weber number, with $X_{z,ave} = 0.420$. In the previous section we have also established that the power-dependency of SMD on $(1 + M_1/M_2)$ is unity. Thus, in order to extract the Weber number dependency from the data presented in Fig.7, the SMD should be divided by $(1 + M_1/M_2)$. The data of Fig.7 was thus correlated by the following Equation:

$$\text{SMD}/(1 + M_1/M_2) \propto \text{We}^{-X_w} \quad (4)$$

Table 2 shows the best least squares solutions of the unknown variables X_w for water and the 5 Xanthan solutions. The correlation coefficients are found to be in the range 0.977 to 0.992. The average value of Weber number dependency X_w is $X_{w,av} = 0.503$.

Table 2. Weber Number Dependency

Solution	X_w	C.O.C
Water	0.507	0.992
0.05% XG	0.482	0.977
0.10% XG	0.518	0.981
0.15% XG	0.504	0.986
0.20% XG	0.505	0.980
0.30% XG	0.502	0.990

General Semi-Empirical Model for Airblast Atomization The wave mechanism has found the most wide acceptance among the mechanisms of atomization. Atomization is a process whereby a volume of liquid is converted into waves, ligaments and drops. When the dynamic pressure ($\rho_a U_a^2/2$) of the air stream in airblast atomization is large enough, the amplitude of the surface waves will grow if their wavelength (λ) exceeds a minimum value ([3]; [4]). When the amplitude becomes sufficiently large, the waves shed ligaments which rapidly collapse, forming drops. When the aerodynamic pressure force predominates, waves propagate as acceleration waves and are governed by $(Z/\text{We})^{2/3}$. In contrast, when the surface tension predominates, waves propagate as capillary waves and are governed by (Z^2/We) [3, 5]. Therefore, both We and Z , with exponents of the Z number equal to or twice that of the We -dependency, are included in our modeling of drop sizes for airblast atomization.

Based on the aforementioned wave mechanism, the following two and three-parameter models are proposed along with the basic drop size equation for air blast atomizer, Eq. (5), [6]. Equations (5), (6a), (6b), (6c), and (6d) are used to correlate the data in terms of the Weber

number, the Ohnesorge number, and the non-dimensional group $(1 + M_l/M_a)$. The linear dependence on $(1 + M_l/M_a)$ is based on momentum balance and energy considerations [5]. The power dependency of unity was verified in a previous section.

$$SMD/D = (1 + \dot{M}_l/\dot{M}_a) [X_2 We^{-0.5} + X_3 Z^{1.0}] \quad (5)$$

$$SMD/D = (1 + \dot{M}_l/\dot{M}_a) [X_2 We^{-X_1} + X_3 Z^{X_1}] \quad (6a)$$

$$SMD/D = (1 + \dot{M}_l/\dot{M}_a) [X_2 We^{-X_1} + X_3 Z^{2X_1}] \quad (6b)$$

$$SMD/D = (1 + \dot{M}_l/\dot{M}_a) [X_2 (Z/We)^{X_1}] \quad (6c)$$

$$SMD/D = (1 + \dot{M}_l/\dot{M}_a) [X_2 (Z^2/We)^{X_1}] \quad (6d)$$

The exponents and coefficients are determined by least squares fit of these equations to more than 220 experimental points. Table 3 shows the best least squares solutions of the three unknown variables X_1 , X_2 , and X_3 of Eqs. (5), (6a), (6b), (6c), and (6d) along with the coefficients of correlation. The coefficients of correlation were found to be in the range 0.933 to 0.972. The best correlations are found for the equations involving the exponent of the Z -dependency equal to the exponent of the We -dependency. This verifies the basic intuition that at the conditions of the study the waves propagate as acceleration waves. The correlation coefficient of Eq. (6c) is particularly good especially that the equation involves only two parameters (X_1 and X_2). Figure 10 demonstrates the agreement between the proposed models with the best least squares solutions listed in Table 3 and the experimental data. The agreement is considered excellent in view of the diverse liquid properties and experimental conditions.

Table 3 Semi-Empirical Model for Airblast Atomization of Non-Newtonian Viscoelastic Fluids

	X_1	X_2	X_3	c.o.c
Eq. (3)	—	0.169	0.581	0.964
Eq. (6a)	0.616	0.264	0.125	0.972
Eq. (6b)	0.390	0.0707	0.275	0.969
Eq. (6c)	0.496	3.414	—	0.969
Eq. (6d)	0.328	2.930	—	0.933

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Fig. 1 Shear Viscosity Data Xanthan Gum Solutions

Fig. 2 Shear Viscosity Data Polyacrilamide E10 Solutions

Fig.3 Die-Swell a) 0.3% Xanthan Gum, b) 0.5% E10 at $8V/D = 5246 \text{ S}^{-1}$.

Fig. 4 First Normal Stress Difference

Fig.5 Variation of SMD as a function of Air/Liquid Ratio

Fig. 6 Power Dependence of SMD on Ohnesorge (Z) Number at Various Shear Rates

Fig. 7 Variations of SMD with Weber Number

Fig. 8 Power Dependence of SMD on Ohnesorge (Z) Number at Various Weber Numbers

Fig. 9 Variation of SMD with $1 + M_l/M_a$

Fig.10 Calculated (by Eq. (6c)) vs Measured SMD

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