Viscoelastic Breakup in a High Velocity Airstream

J. E. MATTA and R. P. TYTUS, Chemical Systems Laboratory, Aberdeen, Maryland

SYNOPSIS

Viscoelastic fluids were injected into a high velocity airstream (200 m/s) to investigate how the addition of small polymer quantities to fluids significantly increase the resultant disseminated drop size. For each liquid tested several hundred resultant drops were sampled and measured using an automated image analyzer. The resultant mass median diameter (MMD) for a viscoelastic fluid was an order of magnitude larger than a comparable viscous Newtonian fluid. A relaxation time measured from a die swell experiment correlates the dissemination results suggesting, an elongational rather than shear breakup mechanism.

INTRODUCTION

The aerodynamic breakup of liquids is an important mechanism in many processes, e.g., paint spraying, fuel atomization, explosive dissemination, aerial dissemination of insecticides, fire retardants, etc. Although many have studied the breakup of Newtonian fluids, aside from low Reynolds's number jet breakup (Rayleigh), empirical models mainly exist. In fact, uncertainty exists even about the mechanism of high velocity liquid jet disruptions.¹ In any case, the drop size, the usual dependent variable of interest is correlated to the liquid density, surface tension, viscosity, and the relative air velocity.

A high relative air velocity normally causes Newtonian liquids to breakup into extremely small particles. For many applications this is undesirable since the drops remain air-borne for long times and fail to impact on the intended locations. To generate larger drops and thus reduce the settling time, polymers are often added.

Various investigators^{2,3} have shown that the slight addition of polymer to a solution may significantly alter a dissemination process. Wilcox et al.² demonstrated using a shock tube technique that polymer modifiers produce viscoelastic properties in solutions which retard their breakup. Hoyts'³ photos of jet discharges from a nozzle demonstrate that spray droplet formation is inhibited by low concentration polymer solutions. Although qualitatively these studies show that a slight addition of polymer significantly alters the deformation process, a quantitative description is not provided.

To investigate the breakup of viscoelastic fluids small quantities were injected into a high velocity airstream. Rheological properties were varied to study their influence on the resultant measured drop sizes and to determine which variables are responsible for the increased particle size of disseminated viscoelastic fluids.

EXPERIMENTAL

Test Fluids

The solvent used in this study was diethylmalonate, DEM, with various concentrations and grades of polymethyl methacrylate additives: (a) a high molecular weight polymer referred to in this report as PMMA was obtained from Rohm and Haas Co. with a viscosity molecular weight average of 6×10^6 , (b) copolymer, a medium molecular weight, 1.9×10^6 , polymer of 80% PMMA and 20% poly(ET/BU acrylate), also from Rohm and Haas, (c) DuPont's low molecular weight, 4×10^5 , powder, Elvacite 2041. 2% Calco Oil Blue ZV was added to all solutions to enhance drop stain measurements.

The viscosity η and first normal stress difference N_1 of the test fluids were measured using a Weissenberg Model R-18 Rheogoniometer (Figs. 1 and 2). The polymer solutions are obviously viscoelastic. Glycerol, also tested, was Newtonian over the same shear rate range with an approximate viscosity of 9 poise.

Wind Tunnel Test Procedure

To generate a sustained high velocity airstream a blow downwind tunnel was fabricated (Fig. 3). The tunnel consisted essentially of a 30 m³ compression tank connected to a cylindrical test section (3 m long with a 6.3 cm ID). The downwind end joined an expansion section which tapered up in 2 m to a 7.25 m² cross-sectional sampling section about 3 m in length.

The standard test procedure was first to establish the high velocity airflow and then inject costream into the 200 m/s wind a 2 s pulse of liquid. The fluid was injected into the test section 3 m downwind of the compression tank either through a 0.30 or 0.46 cm ID stainless steel tube both with length to diameter



Fig. 1. Viscosity vs. shear rate as measured with a rheogoniometer.



Fig. 2. First normal stress difference vs. shear rate as measured with a rheogoniometer.

ratios greater than 80. The fluid flow rate was regulated using a timer controlled solenoid to 3.5 and 7.0 g/s for the small and large diameter nozzles, respectively. The disseminated drops were collected on a vertical paper coated grid located about 2 m downwind in the sampling section. The cross-sectional area of the grid was about 10% of the wind tunnel at the sampling location. After drying, the collected stains were measured on a Quantimet 720 Image Analyzer and were



Fig. 3. Schematic of wind tunnel test facility.



Fig. 4. Log normal probability plot of the disseminated glycerol: drop diameter vs. cumulative mass.

converted to actual drop diameters, using a previously determined spread factor. Stain measurements were made for all the fluids tested except for glycerol, where drops were collected on teflon slides and later measured. Normally a few hundred stains were sampled for each test and several replications with each fluid were conducted.

RESULTS

Newtonian

Figure 4 is a log probability curve of the measured particle diameter for the disseminated glycerol in the high velocity wind. The plot indicates that the fluid is essentially log normally distributed, although the slight inflection hints that the distribution is slightly bimodal.⁴ Table I shows that the measured mass median diameter (MMD) agrees well with the empirical prediction of Weiss and Worsham obtained for low viscosity fluids. Occasionally photos using a high intensity strobe light were taken of the fluid ejecting from the nozzle. Figure 5 clearly shows the rapid atomization of glycerol in the high velocity wind near the nozzle exit.

Weiss and Worsham's⁵ results were correlated empirically by

$$(X\rho_a V^2/\sigma) = 0.61(V\eta/\sigma)^{2/3}(1+10^3\rho_a/\rho)(W\rho\sigma\eta_a/\eta^4)^{1/12}$$

where mass median diameter X, air density (ρ_a) , relative velocity (V), liquid viscosity (η) , and mass injection rate (W) were changed over 4-25-fold range. Surface tension (σ) , liquid density (ρ) , and air viscosity (η_a) were not varied significantly.

Fluid	$MMD (\mu m)$	
	Weiss and Worsham ^a	Measured
2.1% PMMA/DEM	210	1980 ± 250^{b}
1.5% PMMA/DEM	110	1780 ± 200
1.0% PMMA/DEM	60	1480 ± 150
0.5% PMMA/DEM	30	1050 ± 150
5.2% Copolymer/DEM	100	1300 ± 150
9.8% Elvacite/DEM	100	1300 ± 150
Glycerol	130	160 ± 20

TABLE I Comparison of Measured MMD Test Results with Weiss and Worsham Predictions for Various Viscoelastic and Newtonian Fluids Disseminated Using 0.30-cm ID Nozzle

^a Zero shear viscosity was used in the viscoelastic calculations.

^b Standard error of the mean.

Viscoelastic Results

The resultant normal drop distribution is clearly evident from the linear probability fit of four combined replica 9.8% Elvacite tests (Fig. 6). Similar distributions were observed for the other polymer solutions as well. The breakup behavior of the viscoelastic fluid as it is ejected from nozzle (Fig. 7) obviously differs from that of the Newtonian fluid. The fluid appears stretched and does not atomize rapidly as does the Newtonian liquid which eventually results in different final drop sizes.

Shown in Table I are the measured MMD for the polymer solutions. The measured particle sizes are an order of magnitude larger than that predicted from the Weiss and Worsham equation. Actually the effect is possibly even larger since the zero shear viscosities were used in the calculation rather than the reduced values within the injection tube. A correlation of resultant particle size with viscosity was not observed and a comparison of the viscoelastic with the glycerol results indicates that viscosity does not account for the increased particle size.

The fluid shear rate in the nozzle apparently has no effect on the resultant drop size. Similar MMD measurements were found for copolymer tests where only the fluid injection rate was doubled.



Fig. 5. Glycerol injected into a 200 m/s wind.



Fig. 6. Normal probability plot of the disseminated 9.8% Elvacite: drop diameter vs. cumulative mass.

DISCUSSION

Both the ejection photos of the stretched viscoelastic ligament and the observation that shear viscosity does not correlate the dissemination results suggests that the breakup is an elongational process. Using the convected Maxwell constitutive relationship,⁶ one can express the steady elongational viscosity as

$$\eta_e = 3\eta \lambda_e / (\theta \dot{\gamma}_e + 1)(1 - 2\theta \dot{\gamma}_e)$$

For this model the maximum elongation rate $\dot{\gamma}_{e \max}$ is bound by $\frac{1}{2}$ the fluid relation time, θ . This relationship suggests a possible correlation between particle size and θ . From steady state shear measurements θ is calculated using the following expression,

$$\theta = N_1 / \eta \dot{\gamma}^2.$$

An appropriate shear rate for the breakup process is required since both N_1 and η are shear rate dependent. Estimating the process time from the breakup distance and air speed, and then assuming the deformation rate is inversely proportional to this time, results in a deformation rate of 10^2-10^3 s^{-1} . Corre-



Fig. 7. 9.8% Elvacite injected into a 200 m/s wind.



Fig. 8. Attempted correlation of MMD test results vs. relaxation time measured with rheogoniometer data (shear rate, 500 s⁻¹). Test conditions: relative wind velocity, 200 m/s; nozzle ID, 0.30 cm; (O) 0.5% PMMA; (\Box) 1.0% PMMA; (\diamond) 1.5% PMMA; (Δ) 2.1% PMMA; (O) 5.2% copolymer; (∇) 9.8% Elvacite.

lations were thus attempted with rheological properties measured at 500 s⁻¹. The MMD results do not correlate with this θ (Fig. 8). However, since the breakup is viewed as an elongational process, a correlation may exist, if θ is measured in an extensional rather than shear experiment. Recently, Tanner⁷ has associated the die swell phenomena of viscoelastic liquids with the outer sheath extension of the ejected fluid and treats die swell as an elongational process. θ determined from a die swell measurement was thus considered a possible relaxation time for correlating dissemination results.

The die swell ratio was measured for the viscoelastic fluids over a shear range of 500–5000 s⁻¹ (Fig. 9). The swell ratio D_J/D_N was measured from the largest diameter along the jet, D_J , and the inner diameter of the nozzle, D_N . The fluid



Fig. 9. Die swell ratio vs. shear rate measurements. Test conditions: (/) indicates 1.27 mm. ID nozzle otherwise 1.78 mm; (O) 0.5% PMMA; (\Box) 1.0% PMMA; (\diamond) 1.5% PMMA; (Δ) 2.1% PMMA; (O) 5.2% copolymer; (∇) 9.8% Elvacite.



Fig. 10. Correlation of MMD vs. relative relaxation time, $\theta/\theta_{0.5\%}$. Test fluids: (O) 0.5% PMMA; (D) 1.0% PMMA; (\diamond) 1.5% PMMA; (\diamond) 2.1% PMMA; (O) 5.2% copolymer; (∇) 9.8% Elvacite. The dash line for the 0.30 cm ID results is the least square power fit, MMD = 1100 ($\theta/\theta_{0.5\%}$)^{0.19} μ m.

was ejected vertically downward from either a 1.78- or 1.27-mm nozzle with L/D ratios of 85 and 120, respectively. No swell dependence was observed on nozzle diameter. The swell-shear behavior is similar for all the fluids except for the most elastic fluid (2.1% PMMA), which appeared to exhibit some sort of instability towards the higher shear rates. Assuming the swell is dependent only on Weissenberg number ($\sim \theta \cdot \dot{\gamma}$), one can calculate a relative θ by the amount of shift required to superimpose all the curves. The 0.5% PMMA fluid was chosen as the relative fluid to which all the curves were shifted. The essentially parallel swell curves result in relaxation times independent of shear rate.

A correlation obviously exists between the MMD results and the relative relaxation time $\theta/\theta_{0.5\%}$ (Fig. 10). Surprisingly, the least square fit shown is ex-



Fig. 11. Correlation of MMD versus first normal stress difference measured at 500 s^{-1} shear rate. Test conditions: nozzle ID, 0.30 cm; (0) 0.5% PMMA; (\Box) 1.0% PMMA; (\diamond) 1.5% PMMA; (Δ) 2.1% PMMA; (O) 5.2% copolymer; (∇) 9.8% Elvacite.

tremely consistent with a linear stability analysis,⁸ using the convected Maxwell model for extensional viscosity. Although the linear stability analysis is not really applicable to the later stages of drop formation, it was used to provide some insight into the nonlinear effects and results in a MMD ~ $\theta^{1/6}$ relationship. Furthermore, the linear behavior between resultant drop size and initial ligament diameter, also a stability analysis prediction, was observed.

In addition to θ , N_1 measured at 500 s⁻¹ correlates MMD results (Fig. 11). However, since considerable uncertainty exists over the breakup deformation rate, the shear rate independent θ is preferred over N_1 for resultant particle size predictions. The consistency of breakup results with the linearized stability analysis also tends one to favor θ . Planned tests, however, with heated fluids may clarify the appropriate variable. Preliminary measurements indicate that N_1 decreases much more rapidly with temperature than θ ; thus dissemination of heated fluids should provide a distinction between the two correlating variables.

References

1. R. D. Reitz, "Atomization and Other Breakup Regimes of a Liquid Jet," Ph.D. dissertation, Princeton University, 1978.

2. J. D.Wilcox, R. K. June, and H. A. Brown, J. Appl. Polym. Sci., 13, 1-6 (1961).

3. J. W. Hoyt, J. J. Taylor, and C. D. Runge, J. Fluid Mech., 63, 635-640 (1974).

4. G. Gerdan, Small Particle Statistics, 2nd ed., Academic, New York, 1960, pp. 96-97.

5. M. A. Weiss and C. H. Worsham, ARS J., 29, 252-259 (1959).

6. M. M. Denn and G. Marrucci, Stretching of Viscoelastic Liquids, AIChE J., 17, 101-103 (1971).

7. R. I. Tanner, J. Non-Newtonian Fluid Mech., 6, 289-302 (1980).

8. J. E. Matta, "Non-linear Viscoelastic Breakup in a High Velocity Airstream," paper presented at the annual Chemical Systems Laboratory Technical Conference, Edgewood, Maryland, 1980.

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