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Measurement of Extensional Viscosity by the Stretching of Viscoelastic Liquid Columns

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Recent work in the field of atomization has explored the breakup of viscoelastic liquids. Studies by Mansour[1] and by Lee and Sojka[2] have both observed that viscoelastic liquids are much more resistant to breakup in comparison with Newtonian liquids. This high resistance to breakup has been attributed to the high extensional viscosities that viscoelastic liquids possess. Although extensional viscosity has been identified as having a major influence on the mode of breakup, no studies have yet been successful in correlating spray quality with a parameter that is truly descriptive of the extensional viscosity of viscoelastic liquids. In this study, a new method is used to determine the extensional viscosities of viscoelastic liquids by the stretching of liquid columns. This method is used to show quantitative variations in extensional viscosity as a function of polymer concentration in aqueous solutions of polyacrylamide e10. Results suggest that as polymer concentration is increased, the liquids exhibit extension thickening behavior.

Introduction

When a liquid is subjected to an extensional flow or a stretching process, the viscous tensile stresses experienced by the liquid will be proportional to its extensional viscosity. The extensional viscosity is defined as the ratio of the first normal stress difference to the extension rate[3].

$$\eta_E^+ = \frac{\sigma_{11} - \sigma_{22}}{\dot{\varepsilon}}$$

In this expression, σ_{ii} are the normal components of the total stress tensor (pressure plus viscous stress) and $\dot{\varepsilon}$ is the rate of extension. The extensional viscosity, $\eta_E{}^+$, can be a function of time for viscoelastic liquids that have time-dependent material properties. The Trouton ratio is defined as the ratio of the extensional viscosity to the shear viscosity. For Newtonian liquids, the Trouton ratio is exactly three. In previous studies, the Trouton ratio has been reported to be as high as 30,000 for some viscoelastic polymer solutions[4]. Thus, viscoelastic liquids can have enormous resistance to extensional flows and stretching.

Past work by Goldin, et al., has investigated the effect of viscoelasticity on the breakup of low velocity laminar capillary jets[5]. It was found that the mode of breakup for viscoelastic liquid jets is significantly different than that of Newtonian jets. Breakup of viscoelastic jets is characterized by the formation of drops interconnected by liquid threads. As a drop's distance from the nozzle exit increases, the threads begin to thin until breakup is finally achieved. This is in contrast to the Newtonian jet in which disturbances result in the formation of drops that quickly break away from the intact length. Experiments conducted by Mansour on the air-assist atomization of viscoelastic polymer solutions of polyacrylamide have shown the same breakup phenomena documented in the work by

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Goldin et al. In Mansour's study, atomization of weakly viscoelastic liquids was achieved. However, as polymer concentration was increased, photographic evidence showed the formation of drops interconnected by liquid threads.

The presence of viscoelastic liquid threads indicates that elongational flow and stretching of the liquid is occurring from the time of drop formation until the time complete breakup occurs (no discernible liquid threads connected to drops). Evidence by Goldin et al. suggested that thinning of the liquid threads could be attributed to a combination of two distinct processes. If the position between two drops connected by a liquid thread remains relatively constant, elongational flow from the liquid thread to the drop occurs, thereby causing the liquid thread to thin. If the drops move relative to one and other, the thread may undergo stretching. In the breakup of a viscoelastic jet, both processes can occur as liquid threads thin leading to complete liquid breakup. This study focuses on the stretching of a liquid column of fixed identity by a moving drop.

Experimental Method

A new technique is used to determine the tensile stress in a liquid column undergoing simple uniaxial extension. Tests were conducted on aqueous solutions of polyacrylamide e10. The experimental set-up consists of a vertical capillary tube which is connected to a source of a viscoelastic liquid. As liquid is supplied to the capillary tube at a very low flow rate, a liquid drop forms at the tube exit. When the drop can no longer be suspended from the tube exit, it falls under its own weight. Because of viscoelastic forces, the drop does not break away from the liquid that is inside the capillary tube as would be expected with a low viscosity Newtonian liquid. Instead, a liquid column is formed that connects the drop with the liquid inside the capillary tube. Once the liquid column has developed, it is stretched by the drop's motion. A time sequence of these events can be seen in Fig.1 for a 0.10% solution of e10. The diameter of the liquid column is uniform along its length and grows smaller as the length of the column increases. Because of viscoelastic and surface tension in the liquid column, the drop acceleration is smaller than that of a free drop. This effect is seen in Fig. 2 in which the trajectory of a water drop is compared to that of a drop formed from the 0.10% e10 solution. When the viscoelastic and surface tensile forces in the column become small in comparison to the weight force, the drop acceleration approaches the gravitational acceleration.

To determine the magnitude of the tensile stress in the liquid column during stretching, the momentum equation is applied to the descending drop accounting for weight, drag, surface tensile, and viscoelastic tensile forces. The resulting equation for the viscoelastic tensile stress is given by the following:

$$T = \frac{4\,m\,(g - a_{drop})}{\pi\,D_{column}^{2}} - \frac{4\,\sigma_{surface}}{D_{column}} - \frac{C_D\,\rho_{air}\,V_{drop}^{2}}{2}\left(\frac{D_{drop}}{D_{column}}\right)^{2}$$

where m is the drop mass, a_{drop} is the drop acceleration, and g is the gravitational acceleration. Drag was calculated using the following expression for the drag coefficient [6] where $Re = v_{drop}D_{drop}/\nu_{air}$.

$$C_D = \frac{24}{Re}\left(1 + 0.167\ Re^{0.667}\right)$$

The stretch rate of the liquid column can be expressed as the ratio of the velocity of the drop to the length of the liquid column. Using continuity, the stretch rate can alternatively be expressed in terms of the column diameter and its time derivative.

$$\dot{\varepsilon} = \frac{V_{drop}}{L_{column}} = -\frac{\dot{D}_{column}}{2D_{column}}$$

Calculation of the tensile stress requires knowledge of the drop mass and the volume of the liquid column. This information was obtained by analyzing enlarged video images of the drop and column within a distance of 0.5 cm of the capillary tube exit. Measurements of the column diameter were performed using a laser-refraction method. As laser light is cast into the liquid column, it is refracted by the column's cylindrical lens shape. The current in a receiving photodiode circuit varies in proportion to the amount of light reaching the active surface of the photodiode. Hence, measurement of a load voltage in the photodiode circuit is related to the liquid column diameter. The column diameter was calibrated with the load voltage by using a series of glass columns ranging in size from 120 - 450 μm. The signal from the load voltage was then analyzed to determined the stretch rate, drop acceleration, and velocity as a function of time assuming the column volume remains constant. Tests were conducted on 0.10%, 0.20%, and 0.30% aqueous solutions of polyacrylamide e10 at 22 °C. Calculations of the tensile stress and extension rate were terminated once the drop acceleration neared 9.6 m/s^2. As the drop acceleration nears the gravitational acceleration, the forces developed by the column have very little effect on the drops motion and the column is observed to break free from the drop shortly after this point in time.

Results

Graphs of the viscoelastic tensile stress and extension rate vs time are shown in Fig. 3,4, and 5 for 0.10%, 0.20%, and 0.30% solutions, respectively. In all three cases, the extension rate increases almost linearly as the column is stretched by the accelerating drop. The extension rates tend to decreases as polymer concentration is increased. As polymer concentration increases, the viscoelastic tensile stress grows to larger values. However, a point is reached at which the viscoelastic tensile stress reaches a maximum and then decreases rapidly. The stretch duration also increases with increasing concentration because the higher viscoelastic force affects a drop's motion over a longer time period until the viscoelastic force has little effect on the drop's acceleration. The following table summarizes maximum tensile stress and critical column diameters for the three solutions.

Concentration (%)	Maximum Tensile Tensile Stress (Pa)	Column Diameter (μm) at Max. Tensile Stress	Column Diameter (μm) at Maximum Acceleration
0.10	2805	201	142
0.20	3709	180	130
0.30	5262	154	101

In the third column of the table, the column diameter is reported at the point in time when the tensile stress reaches a maximum. The fourth column of the table reports the column diameter at the point in time when the acceleration of the drop is close to 9.81 m/s^2. As would be expected, a smaller column diameter can influence the drop's motion if the polymer concentration is increased because of the greater viscoelastic force.

The forces determining the drop's acceleration are the weight, drag, surface and viscoelastic tensile forces. The relative magnitudes of these forces can be shown by decomposing the drop's acceleration to show the separate contributions. Fig. 6 shows the decomposition of the drop acceleration for the 0.10% e10 solution. The graph shows that the drag force is very small which is expected over the relatively short time period stretching occurs. Throughout most of the stretching period the viscoelastic force is approximately twice the magnitude of the surface tension forces. As the surface tension force and viscoelastic force approach the same magnitude, the tensile stress begins to decreases rapidly as seen in Fig. 3. Although the viscoelastic tensile stress is seen to decrease, caution should be used in drawing conclusions after the diameter of the column nears 120 microns which is the limit of the diameter/voltage calibration. As the column diameter becomes much smaller than 120 microns, the assumptions upon which the calculations are based are not accurate because the column lengths become unrealistically long. As the diameter decreases beyond approximately 40-80 microns, the column may break away from the drop and hence the motion of the drop is unknown and the tensile stress in the column cannot be determined.

The extensional viscosities for the 0.10%, 0.20%, and 0.30% solutions are shown in Fig. 7 plotted as a function of extension rate. The graph shows that as the extension rate increases during stretching, so does the extensional viscosity. It is also seen that, as polymer concentration is increased, the extensional viscosity increases with a larger slope. However, it should be stressed that, for a given extension rate, the steady state value of the extensional viscosity is not necessarily reached because the tensile stress in a viscoelastic liquid undergoes a transient period once the deformation rate is changed. Nevertheless, the data quantitatively show a tendency for extension thickening.

Discussion

The method presented in this study provides a quantitative description of the effect of polymer concentration on the tensile strength of liquid columns. Work continues on performing experiments with different size capillary tubes which allow for the development of larger drops and liquid columns. Parametric studies will allow more general conclusions to be drawn about the strength of viscoelastic liquid columns undergoing breakup. Fitting of experimental data with calculations based on viscoelastic constituitive models will allow the extensional viscosity to be described more simply in terms of a relaxation time that is characteristic of a polymer solution. Once the strength of a viscoelastic liquid column can be suitably described in terms of a single parameter, strong correlations may be possible between atomization quality and extensional viscosity.

References

[1] Mansour, A.B., Carnegie Institute of Technology, Thesis, 1994.
[2] Lee,W.Y, and P.E. Sojka, "Effervescent Atomization at Low Mass Flow Rates. Part IV: The Influence of Fluid Viscoelasticity", ASME FED Vol. 178/HTD 270 Vol., pp. 129-135 (December 1993).
[3] Gupta, R.K, Sridhar,T., Rheological Measurements, Elongational Rheometers, 1988.
[4] Metzner, A.P., and Metzner, A.B., Rheol. Acta, 1970.
[5] Goldin, N., Yerushalmi, J., Pfeffer, R., Shinnar, R.,"Breakup of a Laminar Capillary Jet of a Viscoelastic Fluid," Journal of Fluid Mechanics, vol. 38, part 4, pp. 689-711, 1969.
[6] Lefebvre, A.H., Atomization & Sprays, Hemisphere Publishing, 1989.

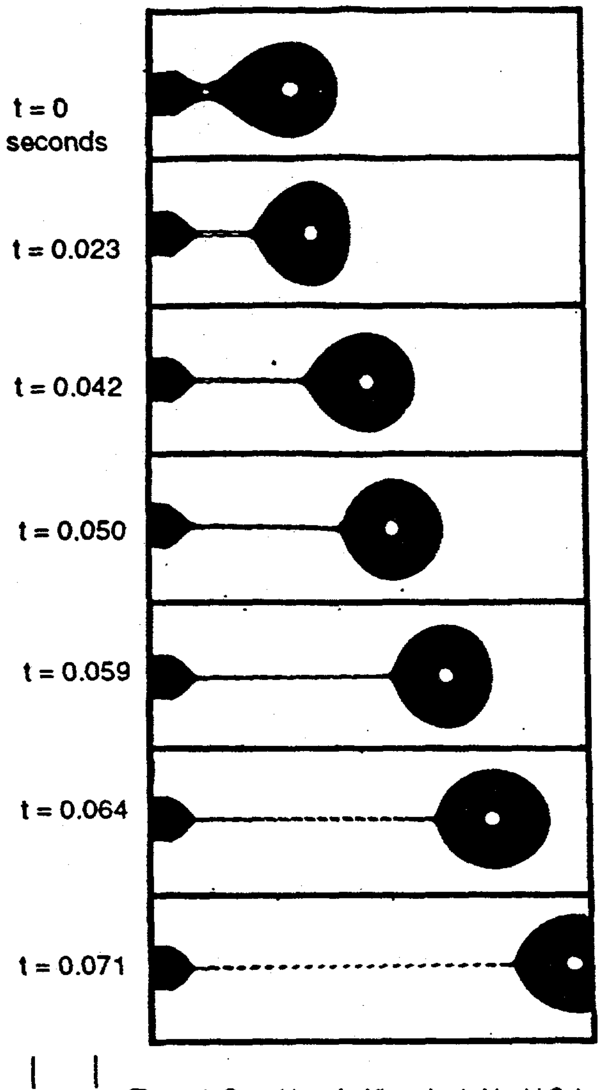

Figure 1: Stretching of a Viscoelastic Liquid Column by a Falling Drop - 0.10% Polyacrylamide e10 Time Sequence

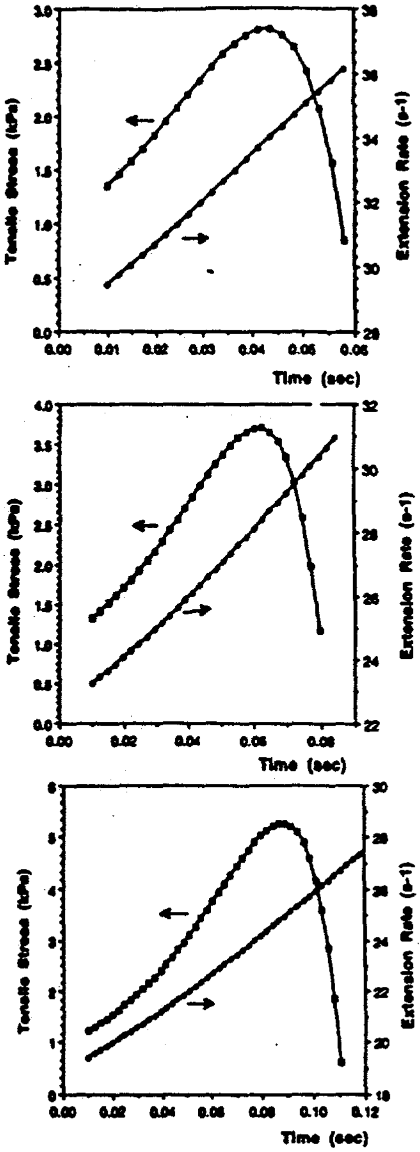

Fig. 3,4,5. Tensile stress and extension rate vs time for 0.10%, 0.20%, 0.30% Polyacrylamide e10 solutions (figures are from top to bottom in order of concentration).

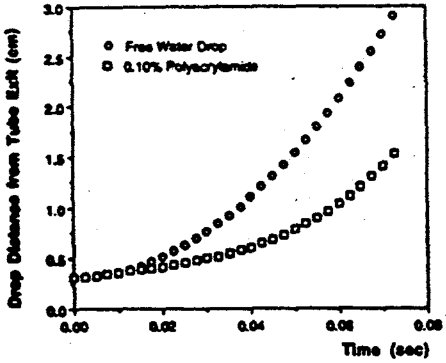

Fig. 2. Trajectory comparison of a water drop and a viscoelastic drop descending from the capillary tube exit.

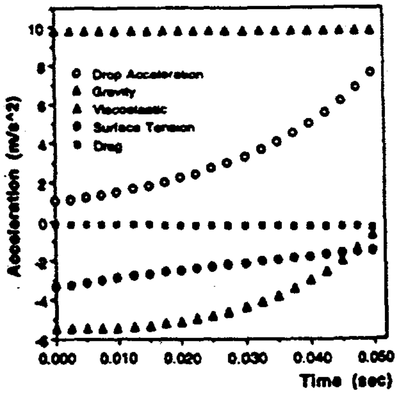

Fig.6. Decomposition of drop acceleration showing relative contributions of viscoelastic, suface tension, drag, and gravitational forces over time.

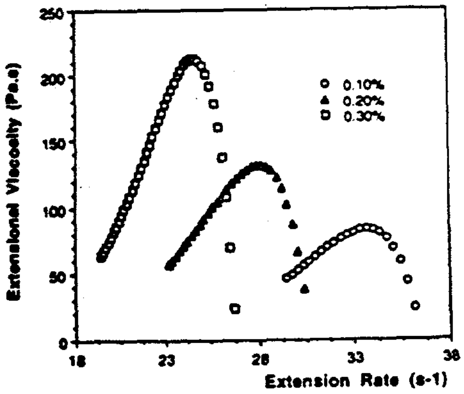

Fig. 7. Extensional viscosity shown as a function of extension rate indicating the effect of increasing polymer concention.