

Flow Visualization of Cellulose Acetate–Acetone Solutions*

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Synopsis

The flow behavior of four cellulose acetate–acetone solutions varying from 20.00% to 28.18% solids concentration was observed using flow visualization techniques at 24°C with an industrial-type dry spinning system. Fibrous particulate matter entrained in the solutions and crossed polars revealed the streamlines and the birefringence due to flow in glass spinnerets. The streamlines of the four solutions were radially convergent at all obtainable throughputs. The crossed polars revealed a cone of birefringence with the order of the interference colors decreasing upstream of the capillary inlet. The intensity and extent of birefringence varied somewhat with concentration and throughput. Except for kneeing, the emerging extrudates were undeformed. The kneeing was not a result of flow instability at the capillary inlet but was due to another, undetermined instability. A separate study of the kneeing revealed both a concentration dependence and hysteresis. The birefringence of the emerging extrudates was a function of both concentration and throughput, and it yielded some information about the structure of the solutions.

INTRODUCTION

Polymer flow instability and the resultant extrudate distortion have been the subjects of many investigations in the past few decades. The literature contains much information on the relationships between extrusion and polymer variables and the inception of extrudate distortion on the one hand and the flow patterns of polymer fluids above the capillary inlet on the other. Several reviews of this literature have appeared in recent years.^{1–3} It is the prevailing opinion that extrudate distortion is the direct result of flow instabilities at the capillary inlet due to the elastic nature of polymer fluids. Although there is empirical information on how the severity of the distortion can be controlled by the proper choice of processing variables, there is still no general agreement on the exact cause or mechanism of the flow instabilities that lead to extrudate distortion.

Although it is thought that the flow behavior that results in instability is similar for both melts and solutions, most work on flow instability and extrudate distortion has been concerned with melts. As far back as 1961 Bagley⁴ proposed that the study of the flow properties of solutions may give information about melts, and Rama Murthy⁵ has certainly shown that there is a one-to-one correspondence between the flow patterns of melts and solutions. However, there has been very little investigation of the influence of extrusion and polymer variables on the flow instability of solutions and virtually no study of the emerging extrudates.

In this study the flow of four cellulose acetate–acetone (CA–acetone) solutions

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is examined at 24°C with an industrial-type dry spinning system. The motion of fibrous particulate matter entrained in the solutions is followed and the birefringence of the four solutions is observed with the aid of crossed polarizing sheets. The flow is examined both at the capillary inlet region of the spinneret and at the capillary exit. The rheological characterization of CA-acetone solutions has been the object of previous studies,^{6,7} and there has been brief mention of the melt fracture of cellulose acetate^{8,9} (probably tri-substituted), but there does not appear to be any reference to the flow patterns or the birefringence of CA-acetone solutions. It is the objective of this study to follow the flow of CA-acetone solutions through a capillary die and to note any transitions or instabilities in that flow and their effect on the emerging extrudate.

EXPERIMENTAL

Dry Spinning and Optical System

Extrusion was performed on an industrial-type dry spinning system.^{7,10} The system consists basically of four sections: (1) solution preparation, (2) solution extrusion, (3) spinning cabinet, and (4) yarn take-up. Only the first two sections were used in this investigation. Dope delivery is accomplished by a Zenith laboratory metering unit equipped with two gear pumps in a differential metering arrangement driven by a Graham variable-speed transmission. The gear pumps used in this study were rated at 1.168 cm³/rev.

Glass spinnerets were made by mounting a small disk of precision-bore glass tubing concentrically inside another piece of precision-bore glass tubing. The external tube has a length of about 5 in. and a 0.5-in. outside diameter. The inside diameter is approximately equal to the outside diameter of the disk. The tops of the external tubes were flanged to fit a modified spinneret holder. The heat treatment used in fabricating the spinnerets caused the capillary diameters to taper slightly. Table I lists the capillary dimensions of the spinnerettes used in this study.

A stereoscopic microscope with a 15× eyepiece and a 1/2× objective mounted on a floating action tripod was used for the visual and photographic observations of the flow patterns of the CA-acetone solutions. The glass spinnerets were placed between crossed polarizing sheets, and a full-wave retardation sheet was placed between the spinneret and the analyzer in order to boost the order of the interference colors. A high-intensity light source provided illumination for the microscopy and birefringence. A Mirax Laborec laboratory camera with a microscope adaptor was used with Kodak Tri-X-Pan ASA 400 black and white film to obtain photomicrographs. Figure 1 shows the optical system along with other features of the extrusion section of the dry spinner.

TABLE I
Capillary Dimensions of the Glass Spinnerettes

Capillary	Length, cm	Top diameter, cm	Bottom diameter, cm	Average diameter, cm
A	0.777	0.086	0.056	0.071
B	0.932	0.157	0.111	0.135
C	1.259	0.467	0.091	0.279

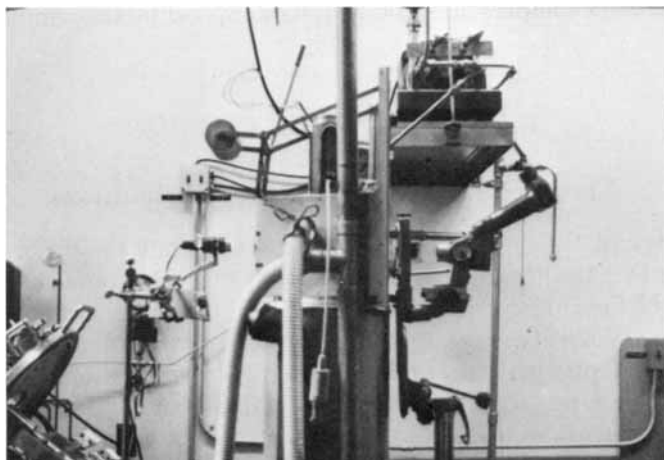


Fig. 1. Optical arrangement for flow visualization of cellulose acetate-acetone solutions.

Materials and Procedure

Conventional secondary cellulose acetate was obtained in flake form from Tennessee Eastman Co. (A-394 B60). The molecular weight of this polymer is 60,000 with a degree of polymerization of 225 and an intrinsic viscosity of 1.60 dl/g in 99.4% acetone. (These data were provided by the supplier.) Four CA-acetone solutions (20.00%, 24.93%, 25.12%, and 28.18% solids) were prepared in the manner described by Griswold and Cuculo,⁷ except that the filtration cycle was eliminated in this study. Partial filtration occurred in the candle filter/heat exchanger of the extrusion section of the dry spinner, and the particulate matter which remained in solution was used as an indicator for flow visualization. The solvent contained about 6% water, including the water in the cellulose acetate. The percent solids concentration of each solution was determined by the procedure given by Work and Cuculo.¹⁰

The experimental arrangement shown in Figure 1 was used to observe the flow of each CA-acetone solution. The crossed polars and full-wave retardation sheet allowed the birefringence to be observed. In addition, the crossed polars enhanced the visibility of the particulate matter entrained in the solutions. Photomicrographs were made to record the flow patterns above the capillary inlet of the glass spinnerets. Photomicrographs were also made of the emerging extrudate at the capillary exit. Observations and photographs of the flow patterns and emerging extrudates were made at various volumetric throughputs. Enough time was allowed at each throughput for steady-state flow patterns to develop before photomicrographs were taken.

The volumetric throughput was calculated by measuring the pinion speed of the Graham variable-speed transmission with a hand tachometer and multiplying that speed by the pump capacity after appropriate gear reduction. Pressure was measured about 5 in. above the capillary with a Dynisco pressure transducer (TPT 432-3M-6/18) mounted at the base of the candle filter/heat exchanger. The pressure was monitored on a Dynisco ER101 meter and recorded on a Hewlett-Packard 680-015 strip chart recorder. The flow data were converted to uncorrected shear rate and shear stress using the measured throughputs and pressures with the capillary dimensions of Table I. Extrusion was performed

at 24°C (ambient temperature); no heat was applied by the candle filter/heat exchanger.

RESULTS AND DISCUSSION

Flow Behavior of the CA-Acetone Solutions

Figure 2 is a plot of the uncorrected shear stress versus the uncorrected shear rate. Except for the low shear rate range where the flow approaches Newtonian character, the flow follows a power law model.¹¹ The consistency index, which is a function of viscosity, increases with solution concentration. The flow index remained constant with concentration, having an approximate value of 0.28. Since this value is less than 1, the flow is pseudoplastic. The pseudoplasticity of CA-acetone solutions has also been demonstrated by Chen⁶ and Griswold and Cuculo.⁷ Chen found that a 27.5% solids CA-acetone solution extruded at 50°C in an Instron capillary rheometer followed a power law with a flow behavior index of 0.37 at shear rates up to $1.7 \times 10^6 \text{ sec}^{-1}$.

Since the length-to-diameter ratios of the capillaries are fairly close, the data for the 24.93% solution fall approximately on the same line. Also, the data for the 25.12% solution are included on that line since the difference in concentration is relatively small. The 24–25% concentration was important in this study because Griswold and Cuculo⁷ had postulated that this concentration may represent the critical concentration for the inception of a long-range entanglement network structure for CA-acetone solutions. They plotted entrance pressure drop against concentration at constant shear rate and saw a rapid increase in slope at 24% solids. It was argued that the entrance pressure drop was a measure of the elastic nature of the solutions. The flow curves in Figure 2 do not suggest a change in elasticity with concentration, but the power law model does not describe the elastic properties of polymers anyway.¹²

Due to the short relaxation times of solutions, flow instability or elastic fracture and extrudate distortion are not usually observed in normal dry spinning.¹³ Capillary breakup at high throughputs is the usual form of instability. The

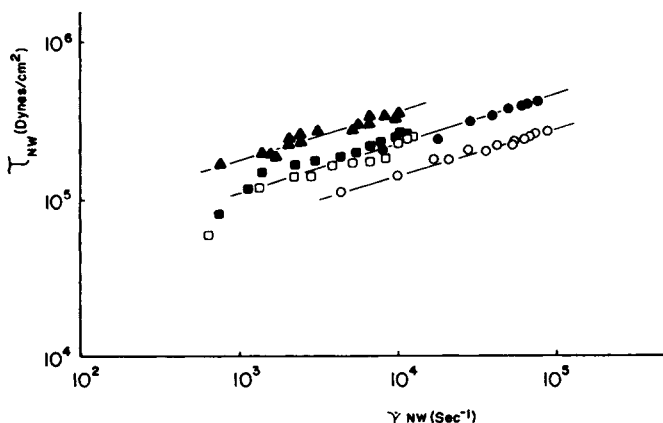


Fig. 2. Uncorrected shear stress vs. uncorrected shear rate for cellulose acetate-acetone solutions at 24°C: (○) cap A, 20%; (●) cap A, 24.93%; (□) cap B, 24.93%; (■) cap B, 25.12%; (▲) cap B, 28.18%.

absence of fracture and the short relaxation times are primarily due to the comparatively low molecular weights of the polymers coupled with the effects of dilution. Both aspects result in relatively low elasticity. Elastic fracture of solutions has been reported,^{5,14} but in these instances polymers of high molecular weight were employed. Rama Murthy⁵ used a dilute solution of polyacrylamide with a molecular weight between 2,000,000 and 3,000,000 and reported a surging of the exiting polymer jet. Southern and Paul¹⁴ used dilute solutions of polystyrene with a molecular weight of 2,000,000 and reported a screw-thread distortion of the emerging extrudate. Extrudate distortion in both cases was accompanied by unstable secondary flow patterns at the capillary inlet. Since the molecular weight of 60,000 for the cellulose acetate used in this study is low by comparison, it was not surprising that the usually described forms of extrudate distortion and flow instability were not observed. The polymer network of CA-acetone solutions is certainly extensive enough to affect spinnability,⁷ but it appears to be insufficient to provide the necessary elasticity for the onset of elastic fracture. Although the extrudate distortion and flow instability associated with elastic fracture did not appear, another form of instability, manifested by kneeling of the emerging extrudate, was observed. This aspect of the flow of CA-acetone solutions will be discussed after the flow at the capillary inlet and exit regions are described.

Flow Visualization at Capillary Inlet

Figure 3 shows the flow pattern of a 28.18% solids solution at a throughput of 1.153 cm³/sec. All solutions exhibited this radical convergence at all throughputs up to 3 cm³/sec. No secondary flow patterns developed. Most of the glare at the center of the spinneret above the capillary inlet in Figure 3 is due to the light source which can be seen at the left of the spinneret. Some of the glare is birefringence. The spots and streaks are not birefringence but are due to the particulate matter entrained in the dope. The lengths of the streaks indicate the relative velocities of different regions of the fluid. It is seen that the greatest

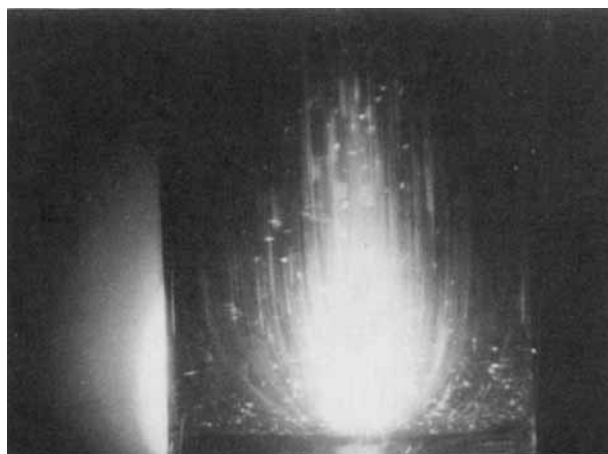


Fig. 3. Flow pattern at the capillary inlet for the 28.18% solids cellulose acetate-acetone solution at 24°C in capillary B. Throughput = 1.153 cm³/sec. Exposure time = 1 sec.

velocities develop along the centerline as the fluid approaches the capillary inlet. It is assumed that the motion of the particulate matter follows the streamlines of the flowing fluid.

When the solutions were placed between crossed polars, a dark cone of birefringence extending vertically above the capillary inlet was observed. A whitish-gray color encircled this central cone. With increasing throughput, the cone widened at the top and extended further up the spinneret. The size of the whitish area also increased by fanning out and extending further up the spinneret. The central cone remained dark at all throughputs and even at the highest throughputs no vorticity was evident. This pattern was basically the same for all concentrations. This description is similar to those given by other investigators studying the birefringence of both melts and solutions.¹⁴⁻¹⁶ The onset of flow instability is usually associated with a twisting distortion or vorticity of the central cone. In all cases the central cone has been described as a region of no stress. In analogy with the birefringence of channel flow, Vinogradov and Manin¹⁵ assigned a value of zero shear stress to the central cone. They also reported that interference colors were due to the photoelastic (shear stress) rather than orientation effect.

In the experiments cited above, the polars were oriented parallel (and perpendicular) to the flow direction. It was found in this study that if the crossed polars were rotated by 45° , the dark central cone above the capillary inlet changed to a whitish-gray color and the white color surrounding the cone vanished. The color of the cone was a first-order white as seen on a Michel-Levy color chart. Further, when a full-wave retardation sheet was added to the optical path, the cone exhibited a range of interference colors and a hazy whitish-yellow area surrounded the cone. Figure 4 is a diagrammatic representation of the birefringence pattern. Directly above the capillary inlet (a), the color was an intense white; above that (b), a yellow-green color was observed; and the top of the cone (c) was bluish violet. The intense white at the capillary inlet does not correspond to a recognized interference color but rather is due to the glare of the light source. The actual color is not known, but based on the preceding colors it was probably a second-order yellow or higher. With increasing throughput, the cone became larger and the lengths of the three colored regions increased. The whitish-yellow region surrounding the cone also became larger. The basic pattern described above did not appear to change with concentration; only the intensity of the colors changed. The more concentrated solutions developed more intense colors.

On the basis of this investigation, it is believed that, for CA-acetone solutions at least, the dark cone of birefringence observed above the capillary inlet is not a region of zero stress as has been reported for other polymer systems. If this were the case, changes in the orientation of the crossed polars would have no effect on the birefringence. On the contrary, these observations seem to indicate increasing stress as the fluid approaches the capillary inlet. Boles et al.¹⁷ have mentioned two occurrences which may account for the increase in order of birefringence: (1) the tendency for the shear stresses developed in channel flow to concentrate toward the centerline of the spinneret as the capillary inlet is approached, and (2) the increase in the difference in normal stresses as the inlet is approached. The second occurrence would seem to suggest that the polymer is becoming more oriented as the flow approaches the capillary inlet. This too would lead to increasing order of birefringence since the anisotropy of the fluid

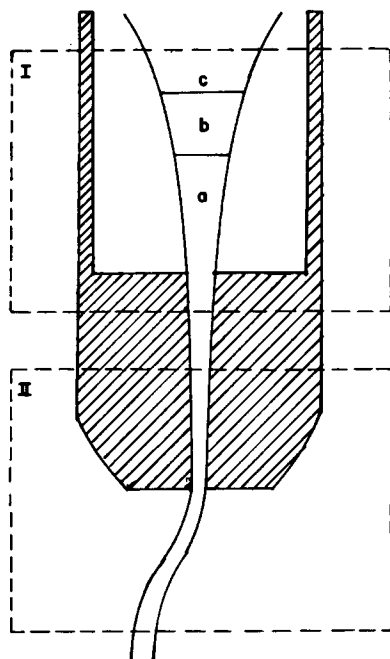


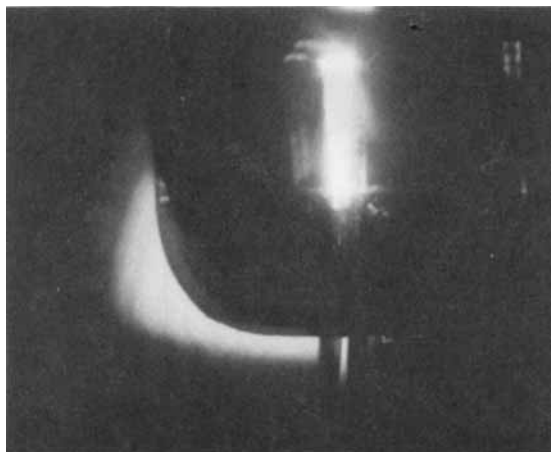
Fig. 4. Diagrammatic representation of birefringence pattern of cellulose acetate-acetone solutions at the capillary inlet.

would be increasing. Since the converging region at the capillary inlet is considered one of elongational flow, it seems likely that the birefringence patterns observed in this investigation are due to orientation effects. Observation of the emerging extrudates also seems to indicate that orientation effects may be important.

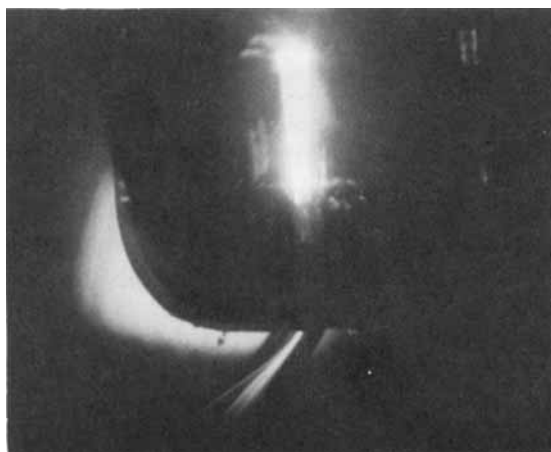
Flow Visualization at Capillary Exit

Figure 5 is a photographic sequence showing the variation of extrudate character with volumetric throughput for the 28.18% solids CA-acetone solution. The region being observed is that designated as II in Figure 4. Two things are obvious from the pictures. First, the die swell has passed through a maximum. The diameter of the extrudate at the $1.465 \text{ cm}^3/\text{sec}$ throughput is much smaller than that at the $0.209 \text{ cm}^3/\text{sec}$ throughput. Even though the die swell has passed through a maximum, the flow patterns above the capillary inlet remain radially convergent and stable. The second noticeable feature of Figure 5 is the occurrence of kneeing.

A number of investigators have reported that melt fracture occurs just past the maximum in a die swell-throughput plot.¹⁸⁻²⁰ At flow rates well past the maximum in die swell for CA-acetone solutions, the extrudate remained undeformed (except for kneeing) and the flow patterns above the capillary inlet exhibited no apparent instability. Rama Murthy²¹ also found no correlation between die swell and elastic fracture for a dilute polyacrylamide solution. In fact, he did not observe a maximum in die swell. Die swell continued to increase even past the point of extrudate distortion. Despite the large difference in elastic



(a)

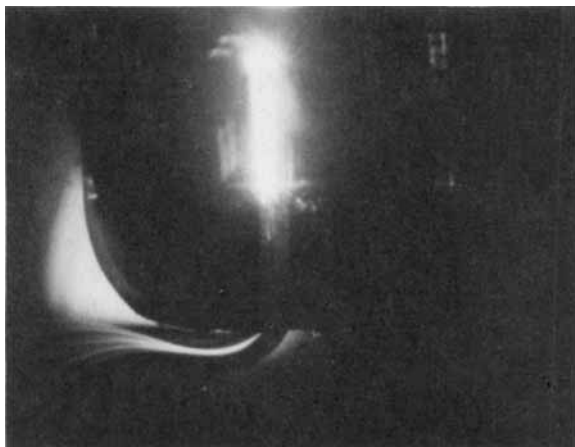


(b)

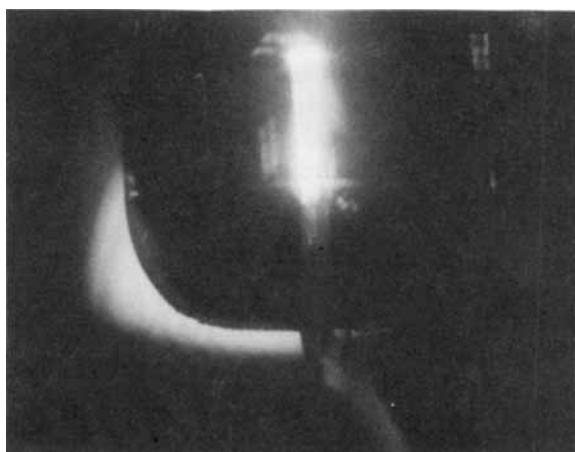
Fig. 5. Photographic sequence showing change in the emerging extrudate with volumetric throughput for 28.18% solids cellulose acetate-acetone solution at 24°C in capillary B. Exposure times = $\frac{1}{2}$ sec. Throughput: (a) 0.209 cm³/sec; (b) 0.438 cm³/sec; (c) 0.763 cm³/sec; (d) 1.046 cm³/sec; (e) 1.465 cm³/sec.

character of the solutions, this investigation corroborates the results of Rama Murthy. Previous investigations were concerned with polymer melts. It may be in this instance that melts and solutions behave differently, even though Rama Murthy⁵ had earlier demonstrated a one-to-one correspondence between melt flow instability and solution flow instability on the basis of inlet flow patterns.

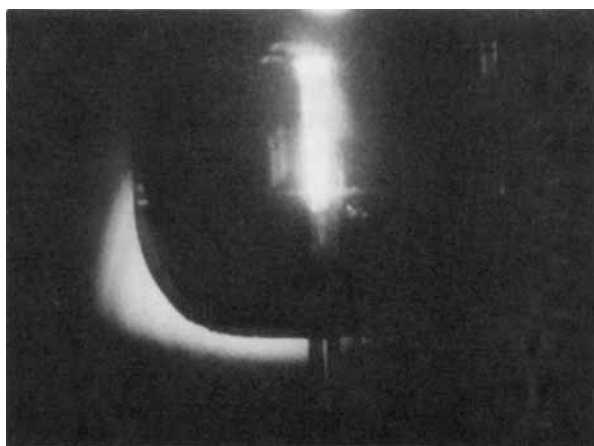
The birefringence of the emerging extrudate was observed in the same manner as above, i.e., the crossed polars were oriented 45° from the flow direction and a full-wave retardation sheet was placed in the optical path. Unlike the patterns at the capillary inlet, the birefringence of the emerging extrudate showed a strong concentration dependence. Each solution exhibited interference colors at the center of the extrudate, but the edges of the extrudate remained colorless and



(c)



(d)



(e)

Fig. 5. (Continued from previous page.)

dark. This can be seen in Figure 5. The interference colors changed with concentration. The center of the extrudate was whitish yellow in the 20.00% solids solution and it remained that color at all throughputs. The 25.12% solution exhibited a small blue spot directly at the capillary exit and the remainder of the extrudate was red yellow in color. This pattern did not change with throughput either. The 28.18% solution also had a blue spot at the capillary exit, but this was followed by a red color and then a yellow-white color further away from the exit. This time when the throughput was increased, the lengths of the regions of interference colors increased. At the highest throughputs the entire portion of the extrudate that was framed by the microscope was blue. It is believed that the blue interference color corresponds to a second-order blue and that the other colors are first order. However, the actual order was lower since a full-wave retardation sheet was in the optical path.

It was suggested earlier that the order of birefringence at the capillary inlet may be second-order yellow or higher. If the order of birefringence at the capillary exit in the 20.00% solids solution is first-order yellow and that of the 28.18% solids solution second-order blue, then the source of the birefringence definitely relaxes more quickly with lower concentration. If the birefringence is due to orientation effects, then it is reasonable that the deformation that produced the orientation would relax more quickly with lower concentration. With increasing concentration, the relaxation time of the solution and the order of birefringence seen in the emerging extrudate should increase. This is exactly what was observed, and thus there is nothing unusual about the birefringent behavior described in this investigation. What is unusual, however, is the change of birefringence with increasing throughput. For the 20.00%, 24.93%, and 25.12% solids solutions there was no change in pattern with throughput. However, as the throughput changed with the 28.18% solution, the lengths of the regions of interference colors of the extrudate increased. This observation seems to support the conclusion of Griswold and Cuculo⁷ that there is a change in the long-range network structure of CA in acetone at 24–25% solids concentration.

The change in order of birefringence with concentration is probably a reflection of the change in recovery time of the deformation that produces orientation, i.e., a viscosity-controlled relaxation. But the change in lengths of the interference colors with throughput for the higher concentration seems also to reflect the change in network structure suggested above. Thus, the relaxation time is increased not only by an increase in viscosity but also by a change in polymer aggregation. It is the latter change which affects the lengths of the regions of interference colors. This change in aggregation may be the formation of the long-range entanglement network suggested by Griswold and Cuculo. It is interesting that an indication of change in network structure does not appear in the flow patterns or birefringence at the capillary inlet.

Kneeing Behavior of the CA–Acetone Solutions

The phenomenon of kneeling has been briefly discussed by Pearson.²² In melts, kneeling is characterized by an asymmetrical bending of the emerging extrudate which tends to coincide with a preferential adherence to the spinneret face on one side of the capillary exit. It is not known whether kneeling is due to asymmetrical flow in the capillary channel or is related in some manner to the

physical properties of the spinneret face. The kneeling of polymer solutions does not appear to be mentioned in the literature.

Kneeing was the only form of instability observed in this investigation. Kneeing occurred with each CA-acetone solution. Moreover, kneeling commenced and ceased at fairly reproducible throughputs, and the range of throughputs in which kneeling was observed was concentration dependent. This aspect of the behavior was investigated further. First, the dry spinner was changed from the differential arrangement to a single-pump arrangement and a larger-capacity Zenith gear pump was used. This pump was related at 2.290 cm³/rev. Then, three CA-acetone solutions were prepared (22.72%, 23.23%, and 25.03% solids). The kneeling behavior was investigated at 24°C using capillary C.

Figure 6 is a plot of volumetric throughput versus solution concentration showing the range of kneeling. The lower solid curve denotes the throughputs at which kneeling commenced, and the upper solid curve indicates the throughputs at which kneeling ceased. The dashed curves have the same significance. The difference between the dashed and solid curves lies in the direction of change of throughput. The solid curves were generated with increasing throughput and the dashed curves represent the range of kneeling with decreasing throughput. The data points are the average of several determinations of throughput for commencement and cessation of kneeling. The brackets around each point indicate the standard deviation of the determinations. This plot shows not only a reproducible change in range of kneeling with concentration but also hysteresis.

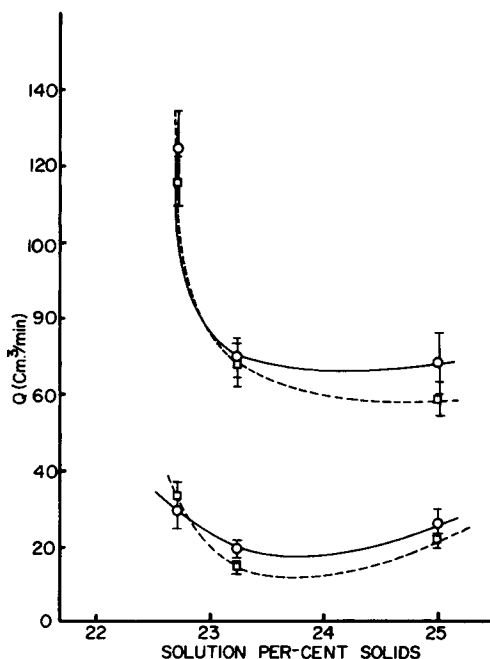


Fig. 6. Volumetric throughput vs. solution concentration showing the range of kneeling, its change with concentration, and hysteresis for cellulose acetate-acetone solutions at 24°C in capillary C: (—) increasing throughput; (---) decreasing throughput.

Kneeing in CA-acetone solutions is somewhat asymmetrical in that it generally began on the left side of the spinneret. The first indication of the kneeling was a slight kink in the emerging extrudate. With increasing throughput the kink changed to a definite bend which became more pronounced and at the same time moved closer to the capillary exit. Just before the throughput reached the point where kneeling ceased, the extrudate began to wag back and forth—at first gradually and then more violently. Then, almost as if the instability were sucked into the capillary, the kneeling ceased. This behavior occurred within the range of throughputs shown in Figure 6. With decreasing throughput the behavior was almost the reverse of that with increasing throughput. Figure 7 shows the kneeling at its earlier stage. The sequence of kneeling behavior is also illustrated in Figure 5.

When the kneeling extrudate began to rotate, it always moved first from left to right (counterclockwise as seen in Fig. 5) and then back to the left. The direction of first movement is opposite the direction of dope flow along a threaded dope guide in the candle filter/heat exchanger. The occurrence of kneeling was not affected by the presence of a distribution screen in the spinneret assembly. Kneeing occurred both with and without the screen. The distance the spinneret capillary was located from the candle filter/heat exchanger also had little effect on the occurrence of kneeling. Finally, there was no apparent relationship between the flow patterns above the capillary inlet and the occurrence of kneeling. The flow remained radially convergent and stable throughout the range of kneeling with each solution.

Kneeing is known in industry but is sometimes associated with the wetting

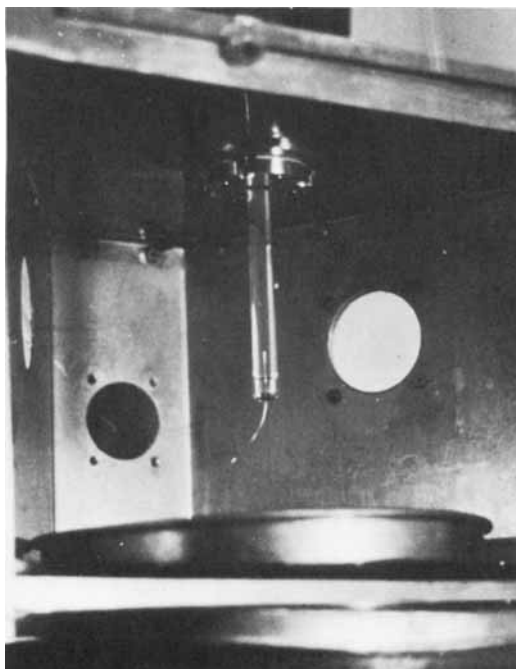


Fig. 7. Kneeing of a 25.03% solids cellulose acetate-acetone solution at 24°C in capillary C at throughput of 14.87 cm³/min.

of the spinneret face after a small deposit of material accumulates at the edge of the capillary exit. This is particularly true in the dry spinning industry. In this case kneeing is a surface tension or wetting effect which is influenced by the cleanliness of the spinneret face. Pearson²² has mentioned this cause of kneeing. In this investigation, the kneeing behavior was not a wetting phenomenon but a genuine rheological response to an instability in the flow. That it is a response is supported by the reproducibility of the phenomenon, the concentration dependence, and the hysteresis. At this point in the study, the exact nature of the instability that results in kneeing is not known. Further study is necessary for complete understanding of this aspect of the flow of CA-acetone solutions.

SUMMARY

The flow behavior of four cellulose acetate-acetone solutions was studied using a modified dry spinning system. Flow visualization at the capillary inlet revealed radially convergent streamlines at all throughputs for each solution. With the exception of kneeing, the extrudates were undeformed. These observations along with the birefringence patterns indicated no elastic fracture even though the shear rates and stresses were in the range of instability of other solutions mentioned in the literature. The absence of elastic fracture is due to low elasticity brought about by the comparatively low molecular weight of the cellulose acetate along with the dilution with acetone.

A dark cone of birefringence extending vertically above the capillary inlet was observed when the spinneret was placed between crossed polars. When the polars were rotated 45° and a full-wave retardation sheet added to the optical path, the central cone exhibited a range of interference colors. The highest order color appeared at the capillary inlet, and the order of the colors decreased with distance above the inlet. The pattern was not influenced by solution concentration; only the intensity of the interference colors changed. The pattern increased in length with increasing throughput. It is believed that the birefringence is an orientation effect.

The birefringence of the emerging extrudates was concentration dependent. Higher orders of interference colors appeared at the capillary exit, and the order decreased with distance away from the exit. The order of birefringence increased with concentration but was still of lower order than the birefringence at the capillary inlet. This observation can be related to the relaxation time of the polymer solution. The variation of the birefringence with throughput for the various solutions yielded some information about the polymer network.

A phenomenon known as kneeing was observed in the cellulose acetate-acetone solutions. The kneeing did not appear to be associated with unstable flow at the capillary inlet or with the cleanliness of the spinneret face. A separate investigation of the kneeing behavior revealed a reproducible range of kneeing which varied with concentration and exhibited hysteresis. This suggests a rheological response to an instability whose exact nature was undetermined in this investigation.

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References

1. J. L. White, *Appl. Polym. Sci. Symp.*, **20**, 155 (1973).
2. C. J. S. Petrie and M. M. Denn, *A.I.Ch.E.J.*, **22**, 209 (1976).
3. E. Boudreaux, Jr., and J. A. Cuculo, *J. Macromol. Sci.—Rev. Macromol. Chem.*, **C16**, 39 (1977–1978).
4. E. B. Bagley, *Trans. Soc. Rheol.*, **5**, 355 (1961).
5. A. V. Rama Murthy, *Trans. Soc. Rheol.*, **18**, 431 (1974).
6. F. C. Chen, *J. Appl. Polym. Sci.*, **16**, 2175 (1972).
7. P. D. Griswold and J. A. Cuculo, *J. Appl. Polym. Sci.*, **18**, 2887 (1974).
8. H. K. Nason, *J. Appl. Phys.*, **16**, 338 (1945).
9. E. T. Severs, *Rheology of Polymers*, Reinhold, New York, 1962, p. 22.
10. R. W. Work and J. A. Cuculo, Technical Report AFML-TR-67-173 Pt. III, Submitted to Air Force Materials Laboratory, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio, 1969.
11. J. A. Brydson, *Flow Properties of Polymer Melts*, Iliffe, London, 1970, pp. 12, 28.
12. C. D. Han, *Rheology in Polymer Processing*, Academic, New York, 1976, p. 129.
13. A. Ziabicki, *Fundamentals of Fibre Formation*, Wiley, New York, 1976, p. 264.
14. J. H. Southern and D. R. Paul, *Polym. Eng. Sci.*, **14**, 560 (1974).
15. G. V. Vinogradov and V. N. Manin, *Kolloid-Z. Z. Polym.*, **201**, 93 (1965).
16. H. Schott and W. S. Kaghan, *Ind. Eng. Chem.*, **51**, 844 (1959).
17. R. L. Boles, H. L. Davis, and D. C. Bogue, *Polym. Eng. Sci.*, **10**, 24 (1970).
18. D. L. T. Beynon and B. S. Glyde, *Br. Plast.*, **33**, 414 (1960).
19. P. L. Clegg, *Br. Plast.*, **39**, 96 (1966).
20. J. L. den Otter, *Plast. Polym.*, **38**, 155 (1970).
21. A. V. Rama Murthy, *Trans. Soc. Rheol.*, **20**, 503 (1976).
22. J. R. A. Pearson, *Mechanical Principles of Polymer Melt Processing*, Pergamon, New York, 1966, p. 114.

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