$Q$ = mass flow rate in axial direction inside the filter hose, kg/s
$r$ = radial distance from center of the filter hose to the surface of cake, $m$
$R$ = radius of the filter hose, $m$
$R_m$ = filter medium resistance, $m^{-1}$
$t$ = time, s
$t_c$ = cycle time, s
$\omega$ = amount of dry cake per area, kg/m$^2$
$W'$ = average mass flux (or rate) of filtrate from a segment of the hose between filtrate collars, kg/m$^2 \cdot$ s
$\overline{W'}$ = time average filtration flux over a segment of hose, kg/m$^2 \cdot$ s
$X$ = weight fraction of solid particle in slurry, dimensionless
$Z$ = axial distance along the filter hose, $m$
$\alpha$ = specific cake resistance, m/kg
$\alpha_i$ = constant specific cake resistance when $\Delta P \leqq \Delta P_i$, m/kg
$\mu$ = viscosity, kN $\cdot$ s/m$^2$
$\rho$ = density, kg/m$^3$

**Subscripts**

avg = average
DFF = dual functional filtration
$v$ = filtrate
$I$ = region I (slurry phase)
$II$ = region II (cake phase)
$F$ = external feed concentration
$\phi$ = actual feed concentration fed to the hose

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# Onset of Draw Resonance During Isothermal Melt Spinning: A Comparison Between Measurements and Predictions

**Various polymer melts were spun isothermally to determine the critical extensional strain at which draw resonance occurs. Simultaneously, the variation of apparent spinning viscosity with extensional strain was measured. Experimental results failed to confirm the theoretical prediction that strain thickening behavior leads to enhanced stability.**

**C. B. WEINBERGER**
**G. F. CRUZ-SAENZ**
**and**
**G. J. DONNELLY**
**Department of Chemical Engineering**
**Drexel University**
**Philadelphia, Pennsylvania 19104**

## SCOPE

The phenomenon of draw resonance is of significant importance in polymer processing. Such resonance appears as a cyclic pulsation in extrudate thickness as polymeric material is continuously extruded and drawn, for example, film coating, fiber spinning, etc. The pulsation, although constant in terms of thickness amplitudes for fixed extrusion and take-up speeds, becomes more severe as take-up speed is increased.

The dynamics of typical drawing operations can frequently be expressed in terms of tensile force which is constant with respect to position but variable with respect to time. Such dynamics obtain when the viscosity is sufficiently large to render negligible the contributions from gravity, inertia, and surface tension to the tensile force. For these dynamics, linearized perturbation analyses (Kase et al., 1966; Pearson and Matovich, 1969; Gelder, 1971) of the spinning of Newtonian fluids have shown that the onset of such resonance occurs at a critical draw ratio, $E_{\text{crit}}$ = take-up speed/extrusion speed at onset, of about 20. This theoretical result has recently been corroborated by spinning experiments with a Newtonian silicone oil (Donnelly and Weinberger, 1975). For the constant-

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G. F. Cruz-Saenz and G. J. Donnelly are with E. I. du Pont de Nemours, Inc., Wilmington, Delaware.

**force (with position) spinning of a Newtonian fluid, then, only $E$, a measure of the total extensional strain, determines the onset of resonance; such variables as extrusion speed, length of spinning zone, or fluid viscosity do not affect $E_{crit}$.**

**For non-Newtonian fluids, the situation is more complex, since theoretical stability analyses indicate that $E_{crit}$ depends upon nonlinear viscous response (Shah and Pearson, 1972) or elastic response (Denn, 1975). For inelastic, power law fluids, $E_{crit}$ is predicted to increase with the power law exponent. This prediction is equivalent to the prediction that $E_{crit}$ increases with extensional strain; thus extension thickening (strain thickening) fluids should be more stable than extension thinning ones. For elastic fluids, the presence of the elastic response is predicted to be stabilizing, that is, higher values of $E_{crit}$. Since elastic response would be manifest by an apparent extension thickening behavior, the conclusion regarding such behavior and stability is the same as that for inelastic fluids.**

**Previous experimental studies of the relationship between draw resonance and rheological response have been limited to such extension thinning materials as polypropylene (Han et al., 1972) and polystyrene (Zeichner, 1973). Thus, the principal theoretical results described above for non-Newtonian fluids have not been tested. The present work describes the results of isothermal spinning experiments with a variety of polymer melts, some of which are extension thickening. The work was undertaken to determine, experimentally, whether the onset of draw resonance is controlled by the degree of extension thickening behavior or the elastic response of the polymer melt. The melts included low-density polyethylene, high-density polyethylene (two), polystyrene, and polypropylene.**

## CONCLUSIONS AND SIGNIFICANCE

**Quantitative measures of the dependence of the apparent spinning viscosity upon extension rate were obtained in the form of a power law exponent by fitting data for the threadline kinematics to a power law model. Such an exponent lumps together nonlinear viscous and elastic effects. Extension thickening behavior, as indicated by exponents greater than unity, for a low-density and a high-density polyethylene melt did not correlate with enhanced stability. The critical draw ratios for these samples were less than 5, roughly the same as those for the extension thinning materials, polystyrene and polypropylene, and much smaller than the value of 20 for a Newtonian fluid. Approximate measures of the elasticity of the melts likewise revealed no enhanced stability due to melt elasticity. The highest value for the critical draw ratio was recorded for the melt which appeared closest to a Newtonian fluid, that is, least elastic and exponent closest to unity.**

**The principal conclusion to be drawn from the present work is that extension thickening behavior, arising from nonlinear viscous or elastic response or both, does not lead to enhanced stability. For typical thermoplastic polymers, the analysis for purely viscous power law fluids of Shah and Pearson cannot be used to predict the onset of resonance. For extension thickening melts, theoretical predictions obtained from the analysis of Fisher and Denn (1976) do agree approximately with experimental results when shear flow data are used to characterize the fluid. However, these melt characterization data do not describe the observed spinning kinematics, and, when the spinning kinematics are used to obtain melt characterization data, the theoretical values of $E_{crit}$ from Fisher and Denn do not agree with experimental values. For the theoretical stability analyses to represent more closely the behavior of actual polymer melts, the analyses apparently must be modified, for example, by using more accurate fluid models and by using an initial velocity which, like the velocity at the point of maximum die swell, can oscillate as the tensile force oscillates.**

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When polymeric material is extruded and drawn, a continuous increase in take-up speed usually causes a continuous decrease in extrudate cross-sectional area. However, at a critical take-up speed, a cyclic pulsation in the extrudate thickness appears. This pulsation, sometimes called draw resonance (Miller, 1963), becomes more severe as the speed is increased further. The word resonance is appropriate here, since the period of pulsation or oscillation appears to depend closely upon the residence time of a material particle in the region where the stretching takes place (Donnelly and Weinberger, 1975). Furthermore, for fixed take-up and extrusion velocities, the pulsation is stable with respect to amplitude of thickness variation; that is, the system exhibits characteristics of a stable limit cycle.

Draw resonance is a different sort of instability from the so-called *melt-fracture* and *capillary* instabilities, both of which also cause thickness variations in extruded liquid columns. Melt fracture, which has been widely studied (Ballenger et al., 1971; Bialas and White, 1969) is usually associated with excessive shear stresses within the die and occasionally with conduit configuration upstream of the die. Capillary instabilities (Middleman, 1965; Goldin et al., 1969) are caused by surface tension forces which tend to neck down, and eventually to break up, fluid columns.

Draw resonance is associated only with stretching or drawing motions. For example, previous linearized perturbation analyses (Kase, 1966; Pearson and Matovich, 1969; Gelder, 1971) of the stability of drawing Newtonian fluids have shown that the fundamental variable which controls the onset of instability is the total extensional strain (isothermal spinning). This strain variable has been replaced by an equivalent strain measure, the draw ratio $E$, which is the ratio of take-up speed to extrusion speed. The critical draw ratio $E_{crit}$ then defines the strain at which the onset of draw resonance appears. For Newtonian fluids, the theoretical value for $E_{crit}$ is 20.21; this value has recently been corroborated experimentally within 10% for a Newtonian silicone oil (Donnelly and Weinberger, 1975).

For non-Newtonian fluids, the situation is more complex, since $E_{crit}$ is then predicted to depend upon nonlinear viscous response (Shah and Pearson, 1972) or elastic response (Zeichner, 1973; Denn, 1975). For inelastic, power law fluids whose apparent tensile viscosity increases with extension rate, $E_{crit}$ is predicted to increase with the power law exponent $q$; that is, $E_{crit}$ increases with increasing extension thickening behavior. (For this fluid, rate thickening behavior implies extension thickening behavior.) For elastic liquids, the presence of the elastic response is predicted to be stabilizing; that is, elasticity should lead to higher values for $E_{crit}$.

The practical motivation for these theoretical analyses has been to develop means to predict the onset of resonance; presumably such predictive capability can thus afford the processor means to control or avoid such resonance. For Newtonian liquids, then, the result is rather simple; the processor merely must avoid draw ratios which approach or exceed 20. For non-Newtonian fluids, however, the theoretical analyses show that the onset of resonance can be strongly affected by the fluid's rheological response. Our predictive capability regarding resonance with such fluids is then no better than the capability of the particular fluid model chosen to describe accurately the fluid's rheological response in a spinning motion. However, to test the accuracy of such descriptions is no mean task. For example, assuming that one wishes to use integral or rate type fluid models, one can conduct shearing experiments and thus obtain appropriate forms for the material functions (presumably of the invariants of the rate-of-deformation tensor) which are included in the fluid model chosen. One can then measure the kinematics and tensile forces in spinning experiments to attempt to determine whether the observed rheological response conforms to that predicted from the results of shearing tests. Subsequent agreement or discrepancy does not constitute a critical test, however, since the third invariant, while zero in the shearing tests, is nonzero in the spinning motion. If the fluid is modeled as a simple fluid (Coleman and Noll, 1964), the material functions depend, in general, upon all three invariants of the rate-of-deformation or strain tensor. Therefore, shearing tests, in general, are insufficient to predict the rheological response of a fluid in a spinning motion. The practical implication of this is that resonance, in turn, cannot, in general, be predicted from such shearing tests.

To improve our fundamental understanding of draw resonance, it is important to attempt to relate the onset of this instability to the rheological response of the fluid (in a spinning motion). In terms of the theoretical analyses, this amounts to testing the notion that increasing apparent viscosity and elasticity (with strain) leads to enhanced stability, higher critical draw ratio. In steady state spinning experiments it is difficult, if not impossible, to distinguish between elastic and nonlinear viscous response. Typically, spinning experiments are conducted under conditions such that gravity, inertial, and surface tension forces are negligible, thereby yielding the dynamical condition of a tensile force which is constant with axial position. Under such conditions, all spinning tests belong to a monogenic family of extensions, that is, extensions all having the same history of extension rate (Weinberger and Goddard, 1974). It is possible, however, to ignore the distinction between elastic and viscous response and to consider only extension thickening or extension thinning behavior; such behavior can be assumed to arise from some elasticoviscous property of the fluid. Based upon physical arguments for a general Oldroyd fluid, and for the theoretical analysis cited above, the elastic response should always contribute to extension thickening behavior. Therefore, extension thickening behavior, whether it arises from nonlinear viscous effects or elastic effects, is predicted to be associated with more stable threadlines.

Previous draw resonance experiments with molten polypropylene, and extension thinning material, yielded $E_{crit}$ less than that of a Newtonian fluid, thereby tending to confirm the qualitative notion that extension thinning behavior leads to diminished stability (Bergonzoni and DiCresce, 1966; Han et al., 1972). Similar findings have also been reported for molten polystyrene, likewise an extension thinning material. Experiments with extension thickening fluids and with fluids of varying degrees of elastic response have not yet been reported, however. The principal objectives of the present study were to conduct such experiments and therefore to test, at least qualitatively, the relationship between extension thickening behavior and onset of draw resonance.

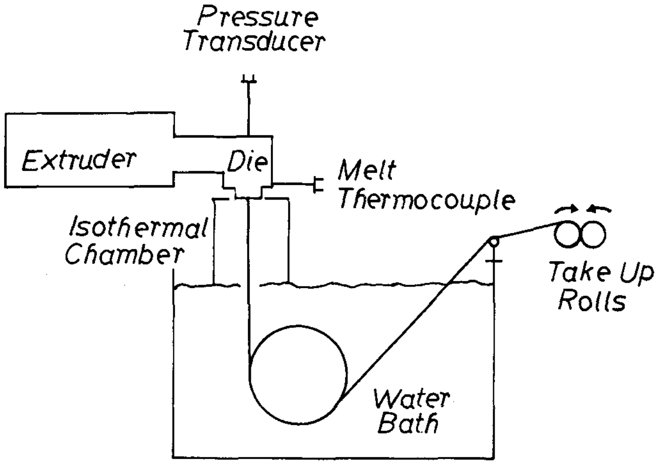

Fig. 1. Sketch of apparatus.

## APPARATUS

Experiments were conducted with a melt spinning apparatus which included a polymer melt extruder, an isothermal chamber, a quench bath, and a variable speed take-up system, Figure 1. Solid polymer pellets were fed to a 19.1 mm Brabender extruder, which served to melt and heat the polymer and to develop the pressure required to extrude the polymer through a 1.59 mm circular die (L/D = 3). The isothermal chamber, 165 mm long, extended from the die to the quench bath, so that all of the drawing took place within the chamber and at a constant temperature. The temperature for all tests was maintained at 218°C. The isothermal spinning chamber was employed to allow rheological effects to be isolated from those of threadline cooling. The chamber had two opposing glass sides to permit visual and photographic observation of the threadline. The quench bath provided rapid quenching, so that extensional deformation beneath the bath surface appeared negligible. The take-up system, consisting of a pair of contacting rolls, could be driven at linear speeds ranging from 0.05 to 0.92 m/s.

## METHOD

The polymer throughput for all the tests was maintained at 6.0 $cm^3/s$ (based upon ambient density) by suitable adjustment of the extruder speed. This throughput corresponds to a die velocity of 5 cm/s. The speed of the take-up system was then gradually increased until a steady pulsation of the threadline thickness could be observed. The speed was then reduced until no pulsation occurred, and the threadline profile was photographed. The take-up speed was then increased in steps, and fiber samples were collected at each speed. Measurement of the fiber diameter as a function of axial fiber distance permitted determination of the ratio of maximum to minimum fiber thickness (D.R.). This ratio was plotted vs. $E$, the ratio of take-up speed to average speed at the die; Figure 2 illustrates such a plot. The intercept of this line with that of D.R. = 1 defines the onset of draw resonance. Such a method provides a less ambiguous means of defining $E_{crit}$ than that of visual observation of the onset of pulsating behavior.

For the sake of convenience, average speed at the die was computed from mass flow rates and density at room

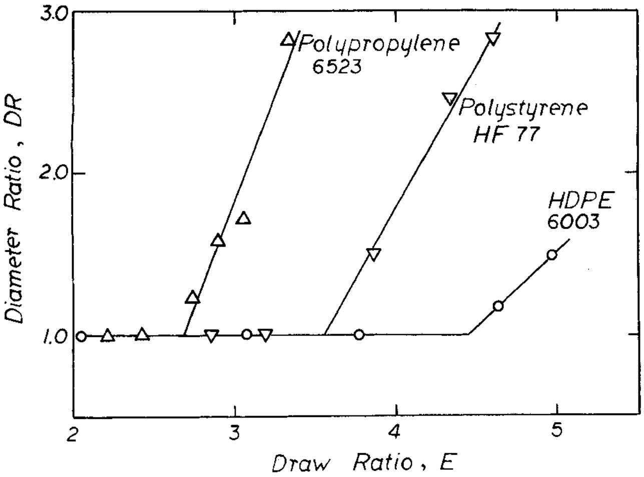

Fig. 2. Determination of critical draw ratio.

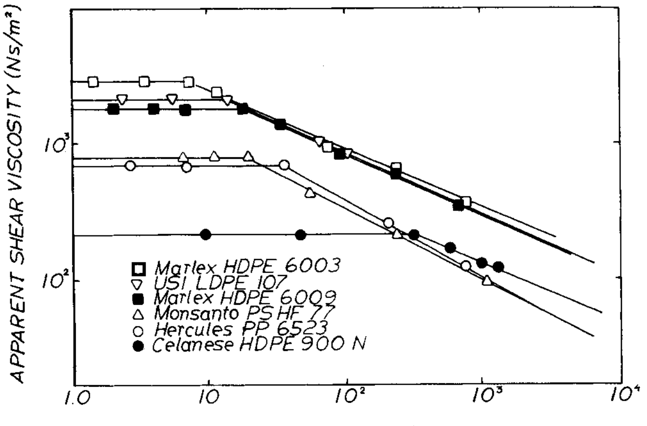

Fig. 3. Dependence of shear viscosity upon shear rate.

temperature. Also, these computed velocities do not include any correction for the swell that such materials experience as they exit from a die. The results of previous experiments with a Newtonian fluid (Donnelly and Weinberger, 1975) suggest that either velocity, die or swell, can be used, since each yield $E_{\text{crit}}$ which differs by approximately the same amount, although in opposite directions, from the theoretical value. In any case, since we were more concerned with changes in $E_{\text{crit}}$ with changes in rheological response than in absolute values of $E_{\text{crit}}$, the particular choice of the basis for $E_{\text{crit}}$ was not so important as the use of a consistent basis. The reproducibility of $E_{\text{crit}}$, as estimated from repeat tests, was better than 10%.

One crucial assumption which required experimental confirmation was that of immediate (nearly) quenching of the polymer filament at the bath surface. This assumption corresponds to the boundary condition of constant take-up speed; such a boundary condition is usually employed in theoretical analyses of the process. If the filament undergoes significant stretching beneath the bath surface, we might expect the measured value of $E_{\text{crit}}$ to be high, based upon predicted stabilizing effects of cooling (Pearson and Shah, 1974) The assumption was checked by using a chilled roll instead of a quench bath and by comparing the two measured values for $E_{\text{crit}}$. For each material, the two values for $E_{\text{crit}}$ agreed within 20% with one exception. The exception was a low-density polyethylene sample, U.S.I. LDPE NA 107; its critical draw ratio was almost three times larger for the quench bath (20°C bath) test than for the chilled roll test. Since this sample also possessed a relatively high shear viscosity and was quite extension thickening, the tensile stress at bath entry was undoubtedly high, and therefore such indirect evidence of stretching beneath the bath surface for this material was not surprising. In any case, one must interpret with care the results of those draw resonance experiments which employ quench baths.

TABLE 1. LIST OF MATERIALS USED

| Material | Manufacturer | Melt Index (g/10 min) | Zero shear viscosity (N-s/m²) |
|---|---|---|---|
| LDPE NA107 | USI | — | $2.0 \times 10^3$ |
| HDPE 900N | Celanese | 9.0 | $2.1 \times 10^2$ |
| HDPE 6003 | Phillips | 0.3 | $2.8 \times 10^3$ |
| HDPE 6009 | Phillips | 0.9 | $1.8 \times 10^3$ |
| Polystyrene HF77 | Monsanto | 4.5 | $7.8 \times 10^2$ |
| Polypropylene 6523 | Hercules | 4 | $7.0 \times 10^2$ |

## MATERIAL CHARACTERIZATION

The materials used in this study included commercial grades of polystyrene, atactic polypropylene, and high- and low-density polyethylene. Table 1 lists the manufacturers and melt index of each material.

### Shear Tests

The shear viscosity of the polymer melts was measured by capillary viscometry, with suitable corrections for end effects. Figure 3 illustrates these data; all the polymer melts were pseudoplastic, with power law exponents in shearing flow ranging from 0.44 to 0.59. The onset of nonlinear viscous behavior occurred at rates of shear ranging from 7.69 to 313 reciprocal s. The onset of nonlinear behavior can be viewed qualitatively as an elastic response, and the inverse of the shear rate at such an onset provides a measure of the material's relaxation time. Dynamic experiments were also conducted with the materials, by using a Rheometric Mechanical Spectrometer. Astarita and Marucci (1974) suggest the following relationship for the relaxation time

$$\lambda_o = \lim_{\omega \to 0} \frac{G_1}{\eta_1 \omega^2} \tag{1}$$

Here $G_1$ and $\eta_1$ are the in-phase components of the complex storage modulus and viscosity, respectively, and $\omega$ is the frequency of oscillation. Both measures of relaxation time yielded nearly the same ranking of the materials in terms of relative elastic response, Table 2.

### Spinning Viscosity

The dynamical equation for the spinning threadline (Matovich and Pearson, 1969) is

$$\frac{1}{\rho}\frac{d}{dx}(\tau/u) = -\frac{g}{u} + \left[1 + \frac{\sigma}{\rho}\sqrt{\frac{\pi}{Qu}}\,\right]\frac{du}{dx} \tag{2}$$

TABLE 2. RANKING OF MATERIALS WITH RESPECT TO ELASTICITY

| Material | $\lambda_R$ (s) | $\lambda_o$ (s) |
|---|---|---|
| HDPE 6003 | 0.13 | 1.64 |
| LDPE NA107 | 0.075 | 1.31 |
| HDPE 6009 | 0.059 | — |
| Polystyrene HF77 | 0.053 | 0.65 |
| Polypropylene 6523 | 0.028 | 0.68 |
| HDPE 900N | 0.0032 | 0.358 |

$\lambda_R$ = calculated from onset of nonlinear viscous behavior.
$\lambda_o = G_1/\eta_1\omega^2$ at $\omega = 0.63\ s^{-1}$ from oscillatory tests.

where $\tau$ is the (axial-radial) principal stress difference, $u$ is the velocity component in the axial $x$ direction, $\sigma$ is the surface tension, and $Q$ is the volumetric flow rate. For highly viscous polymer melts spun at moderate speeds, with sufficiently large threadline diameter and over sufficiently short distances, the terms corresponding to contributions of inertia, surface tension, and gravity, respectively, are negligible, and Equation (2) reduces to

$$\frac{d}{dx}(\tau/u) = 0 \tag{3}$$

which is equivalent to constant threadline tension. In our experiments, gravity contributed to a slight decline in threadline tension in the direction of increasing $x$. Since tensile forces were not measured directly, it was not possible to determine precisely how much gravity contributed to a fractional variation in tension. Estimates of the tensile force, based upon the zero shear viscosity measurements, along with the integrated form of Equation (2), indicated that such tension variation was less than 30% for all the polymer samples except Polypropylene 6523, which was markedly extension thinning. For the extension thickening melts, the materials of most interest in the present study, the tension variation was less than 10%. Accordingly, for the purposes of melt characterization, the use of Equation (3) appears justifiable for the present work.

To fit the observed kinematical data, a simple power law fluid model

$$\tau = K\left(\frac{du}{dx}\right)^q \tag{4}$$

was employed. The assumption of power law behavior is consistent with previous findings with polymer solutions (Weinberger and Goddard, 1974) and perhaps represents less an assumption than a means to characterize the material's rheological response in a spinning motion. Since the local extension rate $du/dx$ can vary along the threadline, we are using Equation (4) to describe a rheological response which may be elastic, as well as nonlinearly viscous. This implies that the power law exponent $q$ does not match the corresponding exponent in shear and that the conditions postulated in the theoretical analyses of spinning stability of purely viscous power law fluids (for example, Shah and Pearson, 1972) are not strictly adhered to. In any case, the present analysis provides a means to quantify extension thickening (or thinning) behavior, and the distinction between nonlinear viscous and elastic response is not strictly required. The rheological response of the melts in a spinning motion was characterized in terms of a relative spinning viscosity, defined as the ratio of the apparent viscosity (as a function of position, extensional strain, or strain rate) to the apparent viscosity near the die:

$$\eta_{\text{spin(rel)}} = \frac{\eta_{\text{spin}}(x)}{\eta_{\text{spin}}(0)} \tag{5}$$

Applying the definition of apparent spinning viscosity

$$\eta_{\text{spin}}(x) = \tau \bigg/ \left(\frac{du}{dx}\right) \tag{6}$$

to the fluid model, one has

$$\eta_{\text{spin(rel)}} = \frac{\left(\frac{du}{dx}\right)^{q-1}}{\left[\left(\frac{du}{dx}\right)^{q-1}\right]_{x=0}} \tag{7}$$

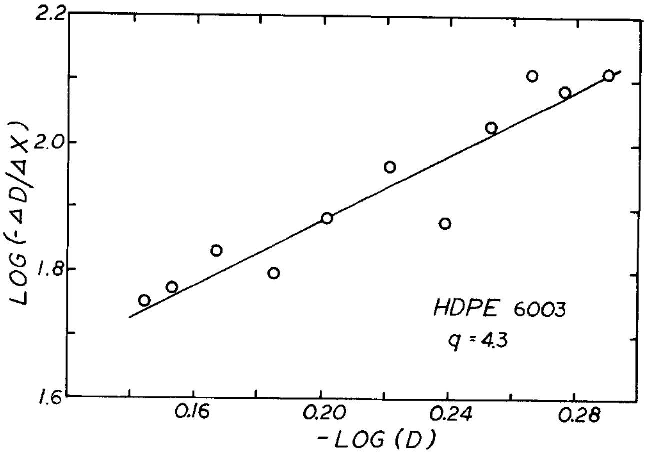

**Fig. 4. Graphical determination of $q$ for HDPE 6003.**

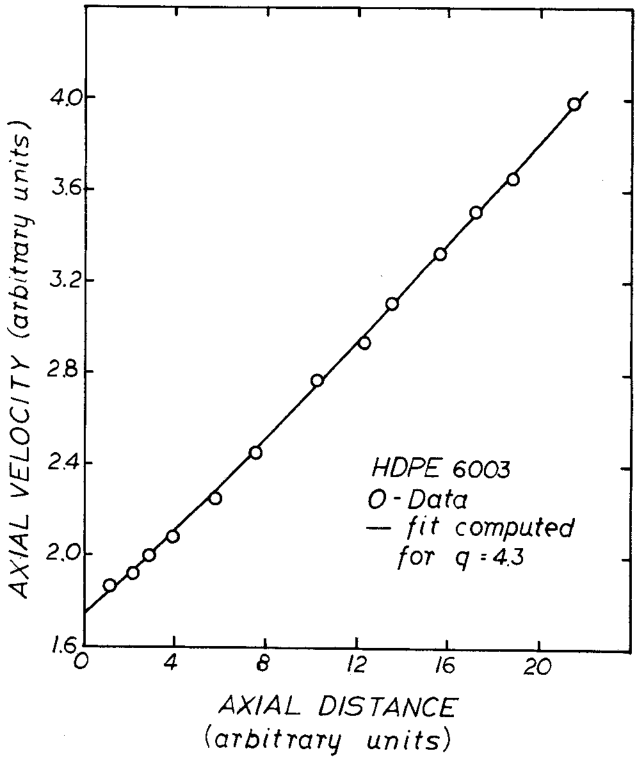

**Fig. 5. Comparison of velocity-distance data with curve computed for $q = 4.3$.**

For the fluids of the present study, $du/dx$ increased with $x$ (and strain); therefore, from Equation (7), $q > 1$ implies extension thickening behavior, and $q < 1$ implies thinning behavior. Absolute values for the spinning viscosity were not required, since the only quantity of interest was the power law exponent $q$. It was obtained solely from kinematic data computed from filament profile measurements; inherent in the calculations were assumptions of constant threadline tension and power law behavior.

When the fluid model, Equation (4), is applied to the dynamical equation corresponding to constant (with space) threadline tension, Equation (3), one has

$$\frac{d}{dx}\left[\frac{(du/dx)^q}{u}\right] = 0 \tag{8}$$

Upon integration and expression of $u$ in terms of threadline diameter $D(x)$, Equation (8) becomes

$$C D^{\left(\frac{3q-2}{q}\right)} = -\frac{dD}{dx} \tag{9}$$

where $C$ is a constant. Thus, $q$ can be obtained from

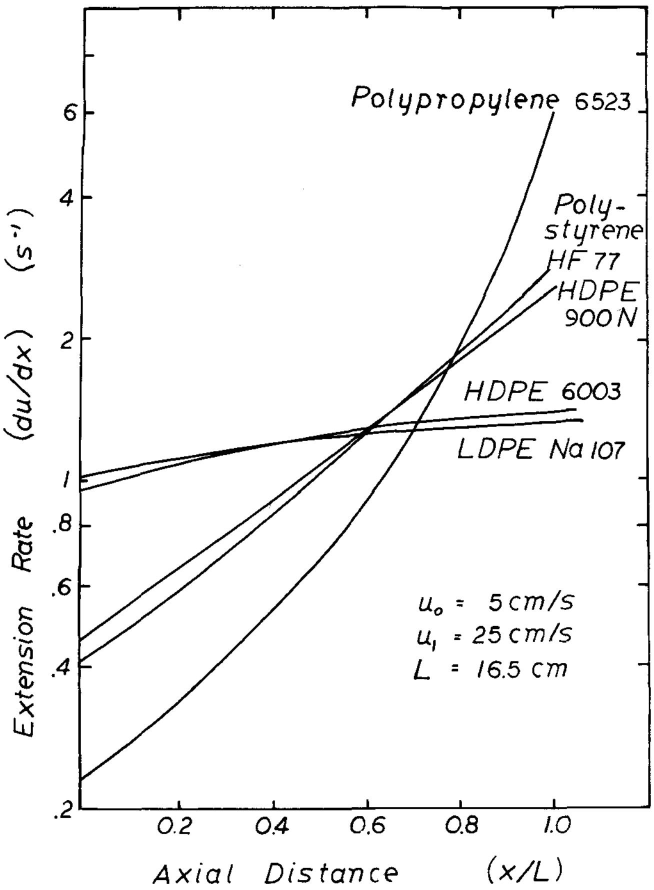

Fig. 6. Variation of extension rate *du/dx* along drawdown zone.

TABLE 3. COMPARISON OF RESONANCE PERIODS TO RESIDENCE AND RELAXATION TIMES

| Material | Period of resonance ($s$) | Residence time ($s$) | $\lambda_R$ ($s$) | $\frac{dDR}{dE}$ |
|---|---|---|---|---|
| HDPE 6003 | 2.1 | 2.6 | 0.13 | 0.9 |
| LDPE NA107 | 1.60 | — | 0.075 | — |
| HDPE 6009 | 3.9 | — | 0.059 | 0.4 |
| HDPE 900N | 2.2 | 1.8 | 0.0032 | 0.5 |
| Polystyrene HF77 | 5.3 | 3.1 | 0.053 | 1.8 |
| Polypropylene 6523 | 5.2 | 3.7 | 0.028 | 2.5 |

TABLE 4. EFFECT OF SPINNING POWER LAW EXPONENT UPON $E_{crit}$ AND COMPARISON WITH PREDICTIONS FROM SHAH AND PEARSON

| Material | $q$ | $E_{crit}$ (observed) | $E_{crit}$ (predicted) |
|---|---|---|---|
| LDPE NA107 | 5.70 | 4.6* | (∞) |
| HDPE 6003 | 4.25 | 3.8 | (∞) |
| HDPE 900N | 0.94 | 18.5 | 17.6 |
| Polystyrene HF77 | 0.83 | 3.6 | 13.7 |
| Polypropylene 6523 | 0.49 | 2.7 | 4.8 |

* Nonisothermal test.

graphical analysis of $D(x)$ data. Threadline diameters as a function of axial position are obtained from photographs of the threadline profile. Approximately fourteen measurements are taken from each photograph. The photographs are taken at take-up speeds slightly lower than those which cause draw resonance.

The finite difference data are then plotted directly, with no smoothing of the data by previous curve fitting. In this sense, then, the data are free of analytical artifact. A typical plot is that of HDPE 6003, Figure 4; the degree of data scatter is approximately the same for the other melts. The fit to the velocity data for the resultant $q$ value for this sample is shown in Figure 5. For this particular sample, the velocity profile was nearly linear; Denn et al. (1975) recently pointed out that such kinematics obtain in the limit of highly elastic materials which can be described by a Maxwell type of model.

The value of $q$ strongly influences the variation of the local extension rate *du/dx* along the threadline. From Equation (8), *du/dx* is

$$\frac{du}{dx} = \left[\frac{u_1^{\beta} - u_0^{\beta}}{\beta L}\right] u^{1/q} \qquad (10)$$

where $u_1$ and $u_0$ are the take-up and die velocities, respectively, $\beta = (q - 1)/q$, and $L$ is the length of the drawdown zone. Figure 6 illustrates the computed variation of *du/dx* for a representative set of experimental conditions for the materials used in the present study. *du/dx* for Polypropylene 6523, which is highly extension thinning ($q = 0.49$), varies tenfold over the threadline, whereas that of HDPE 6003, which is extension thickening ($q = 4.3$), varies only 30% over the same distance.

## RESULTS AND DISCUSSION

The relationship between period of resonance, computed from take-up speed and distance between diameter maxima, and residence time of fluid particles in the drawdown zone is shown in Table 3. The residence times, calculated from the velocity profile as determined from Equation (8), do not include the contribution from die swell and are undoubtedly low. Therefore, the actual residence times can be expected to show even closer agreement with period of resonance. These findings were expected, since earlier tests with a Newtonian fluid (Donnelly and Weinberger, 1975) exhibited the same behavior.

Conversely, approximate estimates of the materials relaxation time, defined as the inverse of the rate of shear at the onset on nonlinear viscous behavior, were always less than one-fourth the resonance period. Further, the period of resonance did not appear to depend upon such relaxation times, Table 3. This does not imply that melt elasticity does not affect the onset of resonance, only that the existence of the resonance does not appear to require the presence of an elastic response in the fluid.

Another reason to monitor the periods of resonance was to assure ourselves that the phenomenon was not initiated by some artifact of the apparatus. One potential such artifact is that associated with small pressure surges (less than 2%) at the extruder exit; such surges, when observed, coincided exactly with the rotational period of the screw. These rotational periods were usually at least twice those of resonance and did not correlate with the resonance periods. When such variables as drawdown zone length or die velocity were changed (keeping rotational speeds constant), the period of resonance always changed in the same direction as would be computed for the residence time from Equation (8). Accordingly, these small pressure variations were neglected.

Table 3 also contains data related to the severity of resonance, characterized by *dDR/dE*, the slope of the diameter ratio vs. draw ratio for $E > E_{crit}$. The high value for polypropylene perhaps suggests the reason that earlier work on draw resonance focused attention on this polymer. Earlier work with a Newtonian silicone oil (Donnelly and Weinberger, 1975) revealed a value of 0.5 for *dDR/dE* for conditions where the calculated tensile force was nearly independent of axial position. It is perhaps significant that Celanese HDPE 900N exhibits approximately the same severity of resonance as the silicone oil.

The principal experimental results are those of critical draw ratio $E_{crit}$ for the various materials. These data, along with the power law exponent $q$ and predicted $E_{crit}$ based upon Shah and Pearson's (1972) theoretical analysis for power law fluids, are given in Table 4. [The critical draw ratio for the low-density polyethylene sample, LDPE 107, although obtained by spinning nonisothermally onto a chilled roll, has been added for the sake of completeness. Values of $E_{crit}$ for the remaining samples spun under nonisothermal conditions differed only slightly ($< 10\%$) from the isothermal values, so that we felt justified including this single nonisothermal datum point in the results.] Although the behavior of extension thinning melts, polypropylene and polystyrene, exhibits qualitative agreement with that predicted theoretically, that is, $E_{crit}$ is much less than that of a Newtonian fluid, the predicted enhanced stability ($E_{crit} > 20$) for extension thickening melts, LDPE Na 107 and HDPE 6003, certainly was not observed. Quantitative agreement was obtained only for the melt which exhibited nearly Newtonian behavior, HDPE 900N with $q = 0.94$.

These results do not constitute a refutation of the Shah and Pearson analysis, since that analysis was completed for inelastic power law fluids, and the polymer melts of this study surely possess some elastic or time dependent response. Our intention was primarily to determine whether the theoretical analysis could be applied to representative polymer melts. A critical test of the Shah and Pearson analysis would obviously require the use of an inelastic power law fluid; however, whether such a fluid exists is open to question.

The results of Table 4 also suggest that elastic properties of the polymer melts do not contribute to enhanced stability. If the extension thickening behavior is attributable to elasticity, we should have observed higher values of $E_{crit}$ for the LDPE and HDPE 6003 samples. A direct comparison between the approximate measure of melt elasticity $\lambda_r$ and the observed $E_{crit}$, Table 5, reveals no stabilizing effect of melt elasticity. A fivefold increase in $\lambda_r$ corresponded to negligible change in $E_{crit}$. If anything, elasticity appears to be destabilizing. Sample HDPE 900N exhibits behavior closest to Newtonian (least elastic), that is, $q$ closest to unity and the lowest value of $\lambda_r$, and yet exhibits the most stable (highest $E_{crit}$) behavior.

One possible interpretation of these findings is that the proper power law exponent with which to predict the onset of draw resonance is that observed in shear, not in spinning. One would then obtain closer agreement between the predicted and experimental values for $E_{crit}$ for all samples except HDPE 900N. However, for the fluids studied here, such an interpretation would require the use of a fluid model with a very strong memory function, so that the wide range of observed extension thickening and thinning behavior for a small range of relaxation times (within a factor of 5) could be properly accounted for. A strong memory function would then be expected, at least qualitatively, to lead to enhanced stability, which was not observed.

TABLE 5. RELATIONSHIP BETWEEN RELAXATION TIME $\lambda_r$ AND $E_{crit}$

| Material | $\lambda_r$ (s) | $E_{crit}$ |
|---|---|---|
| HDPE 6003 | 0.13 | 3.8 |
| LDPE NA107 | 0.075 | 4.6 |
| Polystyrene HF77 | 0.053 | 3.6 |
| Polypropylene 6523 | 0.028 | 2.7 |
| HDPE 900N | 0.0032 | 18.5 |

TABLE 6. COMPARISON OF EXPERIMENTAL RESULTS WITH ANALYSIS OF FISHER AND DENN (1976) FOR EXTENSION THICKENING MELTS

| | HDPE 6003 | LDPE NA107 |
|---|---|---|
| $\alpha^{1/n}$ (from shear data) | 0.0394 | 0.0227 |
| $n$ (from shear data) | 0.58 | 0.56 |
| $E_{crit}^*$ (predicted from shear data) | 8 | 5.2 |
| $E_{crit}$ (experimental) | 3.8 | 4.6 |
| Shear rate at onset of nonlinear viscous response in shear ($s^{-1}$) | 7.7 | 13.3 |
| Average rate of extension in spinning experiments ($s^{-1}$) | 0.834 | 1.07 |
| $\alpha^{1/n}$ (from spinning data) | 0.364 | 0.284 |
| $E_{crit}^*$ (predicted from shear and spinning data) | stable | stable |
| $E_{max}$ | 3.75 | 4.52 |

* From Figure 5 of Fisher and Denn (1976).

The recent analysis by Fisher and Denn (1976) of the spinning stability of a Maxwell type of fluid incorporating a single relaxation time and a power law viscosity can also be used to predict $E_{crit}$. Their analysis yields a plot (Figure 5 of F-D) describing

$$E_{crit}\ (n, \alpha^{1/n})$$

where $n$ is the power law exponent in shear, and

$$\alpha^{1/n} = \frac{\lambda u_o}{L} \tag{11}$$

where $\lambda$ is the single relaxation time of the material. $\lambda$ can be based upon either shear flow data or spinning flow data. In the former case, $\lambda = \lambda_r$, and, from Figure 5 of F-D, the predicted values of $E_{crit}$ for our extension thickening melts are obtained, Table 6. The comparison with observed $E_{crit}$ in this table appears quite good, qualitatively. This agreement is perhaps insufficient to constitute confirmation of the Fisher-Denn analysis for our fluids, though, owing to the question of how best to define $\lambda$. Also included in Table 6 are values for the shear rate at the onset of nonlinear viscous behavior and the range of extension rates in the spinning experiments. Non-Newtonian behavior for these materials occurs at much lower strain rates in a spinning motion than in a shearing motion (roughly $< 1\ s^{-1}$ vs. 7 to 300 $s^{-1}$). This type of behavior has been observed previously with polymer solutions (Weinberger and Goddard, 1974). It would therefore seem more appropriate to use the kinematical response of the polymer melts in a spinning motion, rather than a shearing one, to define $\lambda$. Fisher and Denn point out that, for their fluid model, in the limit of dominant elastic stresses

$$u = u_o + \frac{u_o x}{L \lambda^{1/n}} \tag{12}$$

or one obtains a nearly linear velocity profile. For large $q$, such a linear profile is obtained, for example, Figure 5. Differentiating Equation (12) with respect to $x$ and comparing it to Equation (10), one can obtain estimates of $\alpha^{1/n}$ based upon spinning data. Table 6 also shows such estimates of $\alpha^{1/n}$ and corresponding predicted values for $E_{crit}$. In both cases, the flow is predicted to be stable, in contradiction with experimental results. The F-D analysis also yields, in this limit of high stresses

$$E_{\max} = 1 + \frac{1}{\alpha^{1/n}}$$

which is a consequence of their particular fluid model which is reminiscent of the infinite tensile viscosity at finite extension rates (Weinberger and Goddard, 1974). Although these values for $E_{\max}$ are not too different from $E_{\mathrm{crit}}$, oscillatory behavior for these melts was observed at $E_{\mathrm{crit}}$, and one would not expect that $E_{\max}$ should define another region of oscillatory instability.

In terms of evaluating the predictive capability of the Fisher-Denn analysis for thermoplastic polymers, we are left with the rather perplexing finding that predictions of $E_{\mathrm{crit}}$ based upon shearing flow data do agree with experiment, whereas those based upon spinning flow data do not. This difference is equivalent to the fact that the observed spinning kinematics cannot be predicted by the fluid model used by F-D, using our shear-flow data. Since the Maxwell type of model of the F-D analysis does not include any dependence upon the third invariant of the rate-of-deformation tensor, we should perhaps not be surprised by such lack of agreement. However, this inability of a single Maxwell type of model (with given material constants) to describe rheological response in both shear and spinning for the same material raises the question of whether the earlier agreement between theory and experiment for $E_{\mathrm{crit}}$ is fortuitous. In any case, additional experimental tests with fluids possessing widely varying degrees of elastic response should permit a more complete evaluation of the predictive capability of the Fisher-Denn analysis.

The principal conclusion to be drawn from the present work is that extension thickening behavior does not contribute to enhanced spinning stability. Such a finding provides some insight into the nature of the instability, since it suggests that the instability does not occur as the growth of local (applying to a fluid particle) disturbances. If enhanced stability had been observed, then the analysis of Chang and Lodge (1971) of the growth of local diameter disturbances with fluids exhibiting extension thickening behavior could also have been used to explain such a result. Apparently, therefore, the instability arises from a global (that is, the entire flow domain) effect rather than from a local one. This concept is embodied in previous limit cycle analyses of the phenomenon (Ishihara and Kase, 1975; Fisher and Denn, 1975).

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## NOTATION

$D$ = filament diameter
$DR$ = filament diameter ratio
$E$ = draw ratio
$L$ = filament length
$q$ = power law exponent
$u$ = axial velocity
$x$ = axial distance
$\eta_{\mathrm{spin}}$ = apparent spinning viscosity
$\lambda$ = relaxation time
$\tau$ = (axial-radial) principal stress difference

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