

## ANOMALOUS ELONGATIONAL FLOWS AND CHANGE OF TYPE

DANIEL D. JOSEPH, KANGPING CHEN

*Department of Aerospace Engineering and Mechanics, University of Minnesota, Minneapolis, MN 55444 (U.S.A.)*

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### Summary

Anomalous effects on elongational flows at high rates of elongation reported by Ferguson et al. [1] are here treated as a change of type. Analysis predicts that the vorticity near the drum is hyperbolic, elliptic away from the drum, under the supercritical conditions in the experiment when the shear-wave speeds have the values which we measured with our wave-speed meter.

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### 1. Introduction

Recent experiments of Ferguson et al. [1] on elongational flows of polymer solutions at high rates of elongation have revealed anomalous effects which appear to arise from a qualitative change to more elastic responses in the neighborhood of certain critical drum speeds. They suggested that these changes are brought about by a stress-induced phase change called complexation, and they gave evidence to back up their idea. The drum speeds at which these anomalous effects were reported are of the order of critical extrusion velocities in similar solutions reported by Joseph et al. [2] for the delay in delayed die swell. Delayed die swell can also be connected to phase changes in some cases (Giesekus, [3]) but another explanation based on a change of type of the vorticity equation from elliptic to hyperbolic seems to be in better agreement with the experiments. In this paper, we do what we can to establish the change of type explanation for the anomalous effects observed by Ferguson et al. Our effort is two-fold: (1) On our wave-speed meter, we measured the shear-wave speeds for the liquids used by Ferguson et al. The measured speeds range from about forty to eighty per cent of the post critical drum speeds reported in their experiments. This means that the Mach number  $M = W/c$ , where  $W$  is the drum speed and  $c$  the shear-wave speed, is between 1.20 and 2.5, roughly. (2) We

did an analysis of an idealized problem for the experiment, using different models. The analysis shows that the vorticity near the drum in the idealized problem changes type when the wave speeds have the measured values and the idealized thread line is longer than a model dependent critical length ranging between one and twenty centimeters.

## 2. Experiments I

We shall consider the experiments of Ferguson et al. [1] and the measurements of wave speeds for the same fluids which we did on the wave-speed meter [4] in our laboratory.

TABLE 1

Some typical flow phenomena observed during elongational flow (after Ferguson, Hudson and Warren [1])

Polymer solution	Drum speed ( $\text{m} \cdot \text{s}^{-1}$ )	Total strain at the drum	Phenomena observed
Silicone Oil ( $\eta_0 = 1.3 \text{ Pa} \cdot \text{s}$ )	0.0	0.8	Pure extensional flow
with $\text{Al}_2\text{O}_3$ particles	0.35	2.2	Pure extensional flow
0.5% PAM in ethylene glycol ( $\eta_0 = 1.4 \text{ Pa} \cdot \text{s}$ )	0.0	0.8	Pure extensional flow
with $\text{Al}_2\text{O}_3$ particles	0.35	2.2	Anomalous flow. Particles moving at different speeds dependent upon their radial position. Faster particles in the body overtaking slower ones on the outside of the filament
5% PEO in water	0.05	0.9	Tack high. Fluid adheres to syringe needle touching filament.
( $\eta_0 = 45 \text{ Pa} \cdot \text{s}$ )	1.50	3.4	No obvious tack. Syringe needle passes through filament without fluid adhering
4.5% in water ( $\eta_0 = 31 \text{ Pa} \cdot \text{s}$ )	0.05	0.9	Filament deflected by air jet, returns to equilibrium position when the disturbance is removed
	2.00	3.5	When the deflecting air jet is removed, the filament oscillates laterally. The vibrations die out slowly
2% PAA in ethylene glycol	0.80	1.8	Fluid adheres to and is drawn by the rotating drum.
( $\eta_0 = 98 \text{ Pa} \cdot \text{s}$ )	1.00	3.3	Filament bounces off drum surface. Temporary adherence is followed by ligaments being thrown off the drum

Ferguson et al., did experiments on elongation flows at high rates of extension using their extensional rheometer. With their device, they extended a thread of liquid by winding it onto a drum. They report the winder speed and the zero-shear viscosity of the liquids used, but they do not give the rate of extension or the length of the extending thread. They describe anomalous effects as follows:

“During elongational flow experiments on polymer solutions certain anomalous examples of flow behavior have been noted. These are,

(1) a non-uniform velocity profile in the radial direction in strongly non-Newtonian fluids being elongated at high rates.

(2) the loss of tack in the filament as rate of elongation increased

(3) the bouncing of the filament off a rotating drum at high elongation rates, and

(4) the development of sustained rubber-like lateral vibrations in the fluids elongated at high rate.”

The anomalous behavior is described more precisely, with experimental details, in Table 1 which is reproduced above. We are going to assume that anomalous flow occurs as a critical phenomenon, at a certain speed, though this may be masked by the action of a large effective viscosity as was true for some cases of delayed die swell.

### 3. Theory

In the theory, we assume that the flow is a pure cylindrical extension,  $r$  is the radial distance from the axis of the jet,  $z$  is the axial distance (see Fig. 1) and the components of velocity are  $\mathbf{u} = (u, v, w) = (-sr, 0, 2sz)$  in cylindrical coordinates  $(r, \theta, z)$  and  $s$  is the extensional rate. The analysis will proceed along the lines introduced by Joseph et al. [5] and Joseph and Saut [6]. We choose a constitutive equation and find the stresses corresponding to pure cylindrical extension. Then we perturb the system with a small but otherwise arbitrary perturbation and linearize. We can find the regions of hyperbolicity and the characteristics from the linearized equations. Universally, the same regions of hyperbolicity and characteristics apply to each and every small perturbation of uniform extension. This information is cheap. To go further, we would need to solve a linearized problem perturbing our already idealized problem and the form that such a problem should take is not clear to us. Since we do not solve the problem, or even a linearized version of it, the best we can say is that it appears that a change of type is probable in the set of parameters for which anomalous effects are observed. The physical implications of a change of type are left obscure.

Let  $\mathbf{T} = -p\mathbf{I} + \boldsymbol{\tau}$  be the stress. We assume that the evolution of  $\boldsymbol{\tau}$  is

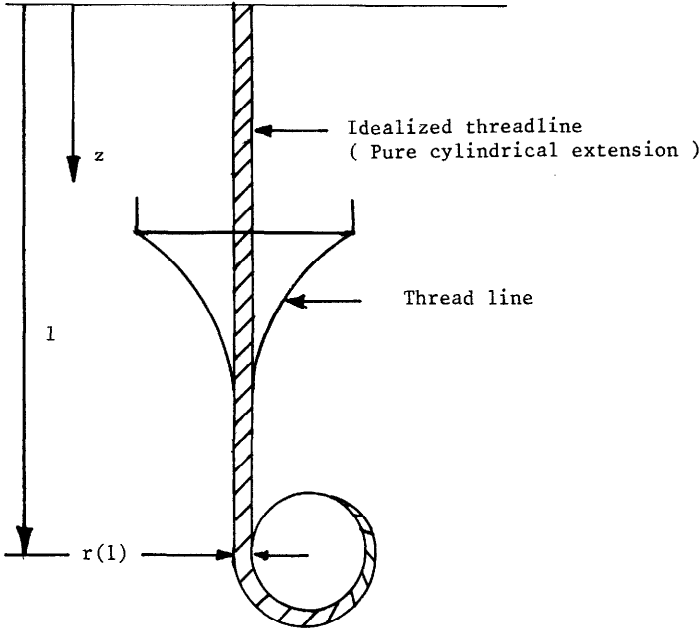


Fig. 1. A liquid thread is elongated by winding it on a rotating drum with peripheral speed  $W$ . The idealized threadline is in pure cylindrical extension with the same speed ( $u, W$ ) =  $(-sr(l), 2sl)$  as the real thread. We imagine measuring  $u, W$  and  $r$  in an experiment, fixing  $l$  and  $s$ . Only  $W$  was given by Ferguson et al. [1].

governed by an upper convected Maxwell model (UCMM)

$$\lambda \frac{D\tau}{Dt} + \tau = 2\eta D[\mathbf{u}], \quad (1)$$

where  $D[\mathbf{u}]$  is the symmetric part of  $\nabla \mathbf{u}$ ,  $\mathbf{u}$  is the velocity,  $\lambda$  a relaxation time,  $\eta$  the viscosity,  $\eta/\lambda$  the rigidity and

$$\frac{D\tau}{Dt} = \frac{\partial \tau}{\partial t} + (\mathbf{u} \cdot \nabla) \tau - \Omega \tau + \tau \Omega - (D\tau + \tau D) \quad (2)$$

where  $\Omega[\mathbf{u}]$  is the skew-symmetric part of  $\nabla \mathbf{u}$  and  $D/Dt$  is an invariant derivative. Pure cylindrical extension is possible if

$$[\tau_{rr}, \tau_{\theta\theta}, \tau_{zz}, \tau_{rz}] \stackrel{\text{def}}{=} [\sigma, \gamma, \beta, \tau]$$

have constant values and  $p$  is a quadratic polynomial,

$$\begin{bmatrix} \sigma \\ \gamma \\ \beta \\ \tau \\ p \end{bmatrix} = \begin{bmatrix} -2\eta s/(1+2\lambda s) \\ -2\eta s/(1+2\lambda s) \\ 4\eta s/(1-4\lambda s) \\ 0 \\ -\frac{1}{2}\rho s^2 r^2 - 2\rho \sigma^2 z^2 + \text{const.} \end{bmatrix}. \quad (3)$$

The stresses are bounded if

$$s < 1/4\lambda. \quad (4)$$

In the idealized flow  $W = 2sl$  and (4) implies that

$$l > 2\lambda W. \quad (5)$$

Now we consider the linearized problem for flows which perturb pure cylindrical extension. We can frame our theory in terms of the vorticity. We shall assume that the disturbances of pure cylindrical extension are axisymmetric. Then the disturbance vorticity  $\zeta$  is equal to the total vorticity. For the moment, let us think that  $\zeta$  is the total vorticity, so we are looking at axisymmetric problems for the UCMM without linearization. Then,

$$A \frac{\partial^2 \zeta}{\partial r^2} + 2B \frac{\partial^2 \zeta}{\partial r \partial z} + C \frac{\partial^2 \zeta}{\partial z^2} = \text{l.o.t.} \quad (6)$$

where l.o.t. means lower order terms, 3rd derivatives of the velocity components at most,

$$A = \rho u^2 - \left( \frac{\eta}{\lambda} + \sigma \right), \quad B = \rho u w - \tau, \quad C = \rho w^2 - \left( \frac{\eta}{\lambda} + \beta \right).$$

It is well known from the theory of 2nd order partial differential equations (PDE's) that (6) is hyperbolic wherever  $B^2 - AC > 0$ . In those regions, the first order differential equations which determine the characteristics are

$$\frac{dz}{dr} = \frac{B \pm \sqrt{B^2 - AC}}{A} \quad (7)$$

In the linearized case, the characteristics are determined a priori by (7) with  $A$ ,  $B$  and  $C$  evaluated on pure cylindrical extension. We find that

$$B^2 - AC = c^4 \rho^2 \left\{ \frac{M_1^2(r)}{1 - 4\lambda s} + \frac{M_2^2(z)}{1 + 2\lambda s} - \frac{1}{(1 - 4\lambda s)(1 + 2\lambda s)} \right\}, \quad (8)$$

where

$$M_1(r) = -u(r)/c = sr/c,$$

$$M_2(z) = w(z)/c = 2sz/c,$$

and

$$c = \sqrt{\eta/\rho\lambda}$$

is the speed of vorticity waves (shear waves) into rest for the Maxwell model. Then the region in the thread in which the vorticity is hyperbolic is defined by

$$(1 + 2\lambda s)M_1^2(r) + (1 - 4\lambda s)M_2^2(z) > 1 \quad (9)$$

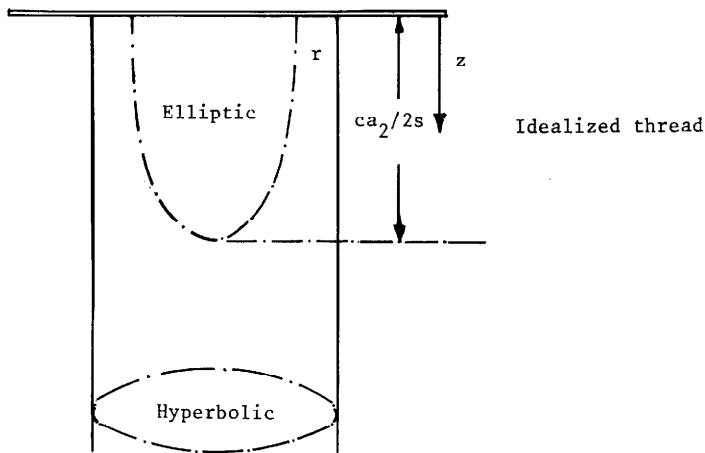


Fig. 2. The vorticity outside the ellipsoid is hyperbolic; inside, it is elliptic.

where  $s < 1/4\lambda$ . The border of the hyperbolic region is defined by an equality in (9) and the inequality says that the vorticity is hyperbolic outside an ellipsoid.

In Fig. 2 we have sketched an ellipsoid

$$\frac{M_1^2(r)}{a_1^2} + \frac{M_2^2(z)}{a_2^2} = 1 \quad (10)$$

centered on the origin of the idealized extending thread. For the UCMM, the semimajor axis is  $ca_2/2s$ , where  $a_2 = (1 - 4\lambda s)^{-1/2}$ . Every point of the extending thread with  $z > ca_2/2s$  is in the hyperbolic region. The portion of the thread which is hyperbolic touches the drum when

$$M_2(l) = W/c = 2sl/c > a_2 \quad (11)$$

and

$$l > cg(\lambda, s) \quad (12)$$

where

$$g(\lambda, s) = 1/2\sqrt{s^2 - 4\lambda s^3}$$

is a concave function such that  $g(\lambda, 0) = g(\lambda, 1/4\lambda) = \infty$ . There is a minimum thread length in the idealized theory

$$l > l_{\min} = c \min g(\lambda, s) = cg(\lambda, \lambda/6) = 3\sqrt{3}\lambda c. \quad (13)$$

The idealized theory says that we may expect a hyperbolic transition for  $0 < s < 1/6\lambda$  and  $l > l_{\min} = 3\sqrt{3}\lambda c$ .

TABLE 2  
Model parameters for pure cylindrical extension calculated from theory

Model	$a_2$	$g(\lambda, s)$	$l_{\min}$	Restrictions for model
UCMM	$\frac{1}{\sqrt{1-4\lambda s}}$	$\frac{1}{2 \cdot \sqrt{s^2 - 4\lambda s^3}}$	$3\sqrt{3}\lambda c$	$0 < s < \frac{1}{4\lambda}$
LCMM	$\frac{1}{\sqrt{1-2\lambda s}}$	$\frac{1}{2 \cdot \sqrt{s^2 - 2\lambda s^3}}$	$\frac{3\sqrt{3}}{2}\lambda c$	$0 < s < \frac{1}{2\lambda}$
CMM	$\sqrt{1+3\lambda s}$	$\frac{\sqrt{1+3\lambda s}}{2s}$	$\frac{3}{\sqrt{2}}\lambda c$	$0 < s < \frac{1}{3\lambda}$
Giesekus	$\sqrt{1 + \frac{1}{2 \cdot \alpha} [4\lambda s - 1 + \sqrt{(1-4\lambda s)^2 + 16\alpha\lambda s}]}$	$\frac{1}{\sqrt{1 + \frac{1}{2\alpha} [4\lambda s - 1 + \sqrt{(1-a\lambda s)^2 + 16\alpha\lambda s}]}}$	$\frac{2s}{2s}$	$0 < s < \frac{1}{4\lambda}$
Leonov-like	$\sqrt{4\lambda s + \sqrt{1 + 16\lambda^2 s^2}}$	$\frac{\sqrt{4\lambda s + \sqrt{1 + 16\lambda^2 s^2}}}{2s}$	$2 \cdot \sqrt{1 + \sqrt{2}} \lambda c$	$0 < s < \frac{1}{4\lambda}$

The restrictions for model validity are associated with singular extensional stresses or with stability. \* We have, in addition, the restrictions of inequality (5) for the UCMM,  $l > \lambda W$  for the LCMM,  $l > 3\lambda W/2$  for the CMM and  $l > 2\lambda W$  for the Leonov model.

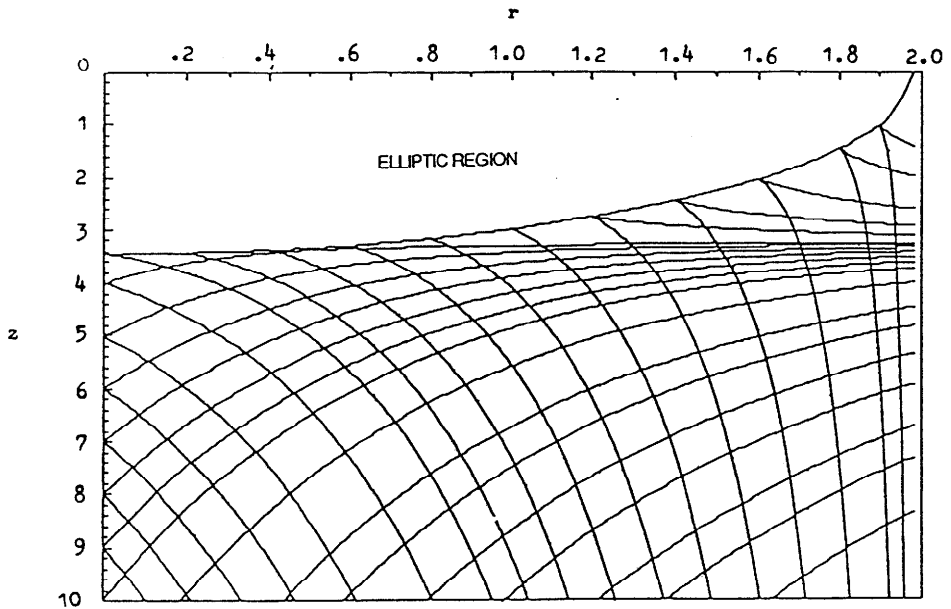


Fig. 3. Characteristics on the cross-section of characteristic surfaces of revolution covering the portion of the thread in which the vorticity is hyperbolic. The computation is for 0.5% PAM in ethylene glycol:  $\lambda = 0.022$  s,  $s = 10.0$  s<sup>-1</sup>,  $\bar{\mu} = 1.4$  Pa s,  $c = 23.9$  cm s<sup>-1</sup> and  $\rho = 1.117$  g cm<sup>-3</sup>.

In Fig. 3, we have exhibited the cross-sections of the characteristic surfaces of revolution for the vorticity generated by (7).

The results just given depend on two false assumptions: that the unperturbed problem is a pure cylindrical extension and the fluid is an UCMM. We tested the results by using other models. We looked at Oldroyd–Maxwell models with a parameter  $a$ ,  $-1 \leq a \leq 1$ , multiplying the last bracket of (2). This alters the principal part of the rate equation. Some Phan Thien–Tanner (Ph.TT) models use this altered principal part and adjust the lower order terms, replacing  $\tau$  with  $\tau(1 + \text{tr } \tau)$ . When  $a = 0$ ,  $\text{tr } \tau = 0$  in pure cylindrical extension, there is no difference between the Ph.TT models and the Oldroyd–Maxwell models. A Giesekus model can be obtained from (2) by replacing  $\tau$  with  $\tau + (\alpha\lambda/\eta)\tau^2$  where  $\alpha = 1/2$  for a Leonov-like model. The general form of the change of type results are similar for all the models but  $a_2$ ,  $g(\lambda, s)$  and  $I_{\min}$  differ from model to model, as can be seen in Table 2.

Joseph and Saut [6] showed that the initial value problem for plane extensional flow of all Oldroyd–Maxwell models with  $a^2 \neq 1$  become ill-posed for sufficiently large values of  $s$  smaller than a yet larger value of  $s$  for which one of the stresses becomes unbounded. Ill-posed problems are badly unstable. In the present case, the corotational model becomes unstable



when  $s > 1/3\lambda$ , but the stresses are all finite at finite  $s$ . The value  $s = 1/3\lambda$  is used as a limit of validity for the corotational Maxwell model in Table 2, whereas the values  $s = 1/4\lambda$  and  $s = 1/2\lambda$  for  $a = 1, -1$  are obtained from singular stresses in the two cases in which there is no loss of well-posedness. The restriction  $l > 2\lambda W$ , arising from the requirement (5) that stresses remain bounded, may also be formulated for the other entries in Table 2:  $l > 2 > \bar{W}$  for the lower convected Maxwell Model (LCMM),  $l > 3\lambda W/2$  for the CMM and  $l > 2\lambda \bar{W}$  for the Leonev model. This  $l$  is not larger than  $l_{\min}$  for the fluids used in the experiments (Table 4). If the idealized flow is hyperbolic at the drum, the stresses will stay bounded.

Equation (11) and the column of values of  $a_2$  in Table 2 show that a necessary condition for a hyperbolic transition is that  $M_2(l) > 1$  or  $W > c$ . In the idealized problem, it is also necessary that  $l > l_{\min}$  where  $l_{\min}$  is achieved as an interior minimum for some models and at the end points of model validity in others.

#### 4. Experiments II

We say that anomalous effects observed by Ferguson et al. are consistent with an explanation based on change of type if  $W > c$  and  $l_{\min}$  is of the

TABLE 3

Wave speeds and rigidities. The columns contain the following data:  $\eta_0$  zero-shear viscosity,  $\bar{c}$  average shear-wave speed,  $G_c$  effective shear modulus,  $\lambda$  relaxation time,  $h$  gap size,  $c$  shear-wave speed for each gap size,  $T$  temperature, and  $\rho$  density.

Fluid	$\eta_0$ (Pa·s)	$\bar{c}$ ( $10^{-2}$ m/s)	$G_c$ (Pa)	$\lambda$ (s)	$h$ ( $10^{-3}$ m)	$c$ ( $10^{-2}$ m/s)	$T$ ( $^{\circ}$ C)	$\rho$ (kg/m <sup>3</sup> )
4.5% PEO <sup>a</sup>	31	86.40	943	0.033	1.0	87.01	25	1263
					3.0	83.03	25	
					6.75	87.30	24	
5.0% PEO	45	125.7	1841	0.024	2.0	119.62	24	1165
					3.0	131.86	25	
0.5% PAM <sup>b</sup>	1.4	23.9	63.8	0.022	1.0	27.21	24	1117
					2.0	24.49	24	
					3.0	19.99	24	
2.0% PAA <sup>c</sup>	98	167.9	3707	0.026	2.0	168.03	24	1315
					3.0	167.92	24	

<sup>a</sup> PEO = poly(ethylene oxide) WSR-301, solution in water,  $M_n = 4,000,000$ .

<sup>b</sup> PAM = poly(acrylamide), solution in ethylene glycol with  $Al_2O_3$ .

<sup>c</sup> PAA = poly(acrylic acid), solution in ethylene glycol,  $M_n = 4,000,000$ .

TABLE 4

Calculated values  $l_{\min}$  for various models using the model parameters of Table 2 and the data from Table 3.  $l_{\min}$  is larger than the  $l$  required for model validity (see caption to Table 3) for the upper and lower convected models and is of the same magnitude for the other two cases

Fluid	$l_{\min}$ (cm)			
	UCMM	LCMM	CMM	Leonov-like
4.5% PEO in water	14.8153	7.4076	6.0483	8.8602
5% PEO in water	15.6758	7.8379	6.3996	9.3749
0.5% PAM in ethylene glycol with Al <sub>2</sub> O <sub>3</sub>	2.7321	1.3661	1.1154	1.6339
2% PAA in ethylene glycol	22.6833	11.3416	9.2604	13.5657

order one might expect to observe in the experiments. We have  $W$  from their experiments and we measured  $c$  in ours. Thread lengths were not reported in their paper. We may compute  $l_{\min}$ , however, from measured data remembering that  $l$  is an idealized quantity defined on a purely cylindrical extensional flow. To compute  $l_{\min}$ , we need the wave speed and a relaxation time  $\lambda$ . A relaxation time  $\lambda = \eta_0/G_c$  may be found from the zero shear viscosity  $\eta_0$  given in Table 1 and the rigidity  $G_c = \rho c^2$ .

TABLE 5

Viscoelastic Mach numbers for supercritical elongational flow

Fluid	$W$ (cm/s)	$c$ (cm/s)	$M$
4.5% PEO	200	86.4	2.31
5.0% PEO	150	125.7	1.19
0.5% PAM	35	23.9	1.46
2.0% PAA <sup>a</sup>	200	167.9	1.19

<sup>a</sup> The bouncing filament phenomenon was described by Ferguson et al. as follows:

The drum speed was set at 200 cm/s. When the filament touched the drum, it was, at first, taken up in the usual manner. However, it very quickly changed its behaviour. It began to bounce off the drum, with a definite and sharp angle of deflection, very reminiscent of an elastic collision of an object with a wall. Its velocity on meeting the drum was approximately 160 cm/s. Occasionally, it attached itself to the drum, but was then thrown off in coherent ligaments before returning to the bouncing behaviour. The same phenomenon occurred when the drum was started from rest with the filament already falling, the bouncing effect starting from a drum speed of about 180 cm/s.

We measured wave speeds in the four solutions used in the experiments reported in Table 1. These are reported in the standard way (Joseph et al. [5] and Riccius et al. [7]) in Table 3 where  $c$  is the average wave speed over different size gaps  $h$  and  $\mu = \eta_0$ . The computation of  $l_{\min}$  for different models, using the data in Table 3, is given in Table 4. These values appear to be reasonable. In Table 5, we compute  $M_2(l) = W/c$  where  $W$  is from Table 1 and  $c$  from Table 3. Since  $c < W$  in all the experiments exhibiting anomalous behavior, we cannot discount the possibility that this behavior is associated with a change of type.

## 5. Conclusions

The analysis just given is to see if the anomalous effects in the experiments could be framed as a change of type and the answer is affirmative. We certainly do not claim that the analysis establishes anything, the flow is not pure cylindrical extension and the fluids used in the experiments are not perfectly described by the models used in the analysis. However, the same general conclusions do follow from each and every one of the models used and the calculated values of the thread lengths which might be expected seems reasonable. It is of interest that all of the aforementioned models do exhibit some kind of hyperbolic transition for high  $s$  when  $W/c > 1$ . The fact that  $W$  and  $c$  are measured quantities, independent of constitutive assumptions, in completely different and unrelated experiments with the universal result that  $W/c > 1$  is a fact which cannot be easily attributed to error or accident.

The results given in this paper should be added to the growing list of transitions observed in experiments which seem to be associated with a change of type. The first entry on this list is the attempt of Ultman and Denn [8] to explain anomalous heat transfer results which were observed by James [9]. In the experiment, there is a rather sharp change in the slope of the graph of the Nusselt number vs. free-stream velocity in the heat transfer from cylindrical wires, for three different size wires, at a speed

$$U = 2.4 \text{ cm/s.}$$

Ultman and Denn [8] calculated what this speed must be, using the formula  $c^2 = \eta/\rho\lambda$ , estimating  $\lambda$  for dilute solutions of Rouse and Bueche, and found agreement with experiments. The solutions from theories used in James' experiment was very dilute, aqueous polyox, 50.2 ppm, almost water. Many rheologists seem reluctant to believe in such low speeds; they think that these waterlike solutions are like water and are dominated by diffusion. However, Joseph et al. [4] measured  $c = 2.48 \text{ cm/s}$  for 50 ppm aqueous polyox WSR-301, and the scatter for different size cylinders was not that

bad. While being sympathetic to the objections about low shear-wave speeds in water-like solutions, we think that it is premature, or worse, to dismiss the results and claims of Ultman and Denn [8].

We have already mentioned delayed die swell (Joseph et al. [2]) in which the delay always occurs when  $u/c > 1$  with  $u/c < 1$  after the swell is completed where  $u$  is the centerline velocity. This close association of an experimentally observed critical transition with the speed of propagation of shear waves occurred in all eighteen liquids studied and for hundreds of measurements.

Another case is the anomalous vorticity distribution which was observed by Metzner et al. [10] in their experiments on flow into a hole. They found potential flow with a zero vorticity inside a conical region with a vortex of about  $20^\circ$  and non-zero vorticity, eddying, outside the cone. This implies a discontinuity in some derivative of the vorticity, suggesting that the vorticity satisfies a hyperbolic equation which will allow such discontinuous derivatives across characteristic surfaces of the type required if the theory is to agree with the experiments. In fact, the observations of Metzner et al., are compatible with an analysis of the type just given when the wave speeds are assumed to be at values which we have measured in solutions similar to those used in the experiment (see Yoo et al. [11]).

Supercritical flow over a flat plane is yet another case in which analysis of flow in two dimensions leads to regions of zero and non-zero vorticity across characteristics which form a Mach wedge centered at the leading edge (see Joseph [12]). The recent elegant Wiener–Hopf solution of this problem by Fraenkel [13] agrees with many of the features of flow observed in the [1967] experiments by Hermes and Fredrickson [14] when  $c$  has the values which we measured on our wave-speed meter for the solutions used in the experiments.

The explanation of anomalous flow as a change of type does not necessarily exclude explanations based on stress induced phase changes (complexation) which was given by Ferguson et al. [1]. Such changes might be expected to occur precisely at points at which the flow speed outruns the speed at which the fluid can propagate shear waves.

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## Appendix

*Governing equations for convected Maxwell model in axisymmetric flow*

$$-1 \leq a \leq 1, \quad \mathbf{u} = u\mathbf{e}_r + w\mathbf{e}_z,$$

$$[\tau_{rr}, \tau_{\theta\theta}, \tau_{zz}, \tau_{rz}] = [\sigma, \gamma, \beta, \tau], \quad \mathbf{T} = -p\mathbf{I} + \tau.$$

Quasilinear system:

$$A \frac{\partial \mathbf{q}}{\partial r} + B \frac{\partial \mathbf{q}}{\partial z} = \mathbf{f}$$

where

$$\mathbf{q} = [u, w, p, \sigma, \gamma, \beta, \tau]$$

$$\mathbf{f} = \left[ -\frac{u}{r}, \frac{\sigma - \gamma}{r}, \frac{\tau}{r}, -\frac{\sigma}{\lambda}, 2\mu \frac{u}{r} + \left(2a \frac{u}{r} - \frac{1}{\lambda}\right) \gamma, -\frac{\beta}{\lambda}, -\frac{\tau}{\lambda} \right]$$

and  $A, B$  are  $7 \times 7$  matrices given by

$$A = \begin{vmatrix} 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ \rho u & 0 & 1 & -1 & 0 & 0 & 0 \\ 0 & \rho u & 0 & 0 & 0 & 0 & -1 \\ -2(\mu + a\sigma) & (1-a)\tau & 0 & u & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & u & 0 & 0 \\ 0 & -(1+a)\tau & 0 & 0 & 0 & u & 0 \\ -a\tau & -\frac{1+a}{2}\sigma + \frac{1-a}{2}\beta - \mu & 0 & 0 & 0 & 0 & u \end{vmatrix}$$

$$B = \begin{vmatrix} 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ \rho w & 0 & 0 & 0 & 0 & 0 & -1 \\ 0 & \rho w & 1 & 0 & 0 & -1 & 0 \\ -(1+a)\tau & 0 & 0 & w & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & w & 0 & 0 \\ (1-a)\tau & -2(\mu + a\beta) & 0 & 0 & 0 & w & 0 \\ \frac{1-a}{2}\sigma - \frac{1+a}{2}\beta - \mu & -a\tau & 0 & 0 & 0 & 0 & w \end{vmatrix}$$