

STABILITY OF A LAYER OF VISCOELASTIC FLUID HEATED  
FROM BELOW

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The polymerization of methyl methacrylate is accompanied by liberation of heat; this results in overheating of the reaction mass during production of plastics. The temperature distribution in the polymerizing layer is complicated by convection, which disrupts the natural temperature field. Thus, in addition to the stress along the sheet, local internal stresses appear that show up in operation of the product. Product quality and intensification of the polymerization process depend on the critical temperature gradient, which determines the stability threshold of the layer of polymerizing methyl methacrylate. The Rayleigh-Jeffrey problem is considered for a weak viscoelastic fluid described by an integral rheological constitutive relationship. The critical Rayleigh numbers are determined for stationary and oscillatory instabilities with free and ideally heat-conducting rigid boundaries.

1. We consider the Rayleigh-Jeffrey problem for the viscoelastic fluid described by the model of [1] and referred to there as a B'-type fluid:

$$T^{ik} = -\pi g^{ik} + \tau^{ik} \quad (1.1)$$

$$\tau^{ik} = 2 \int_{-\infty}^t \psi(t-t') \frac{\partial x^i}{\partial x'^m} \frac{\partial x^k}{\partial x'^n} e^{mr}(x', t') dt' \quad (1.2)$$

$$\psi(t-t') = \int_0^{\infty} \frac{N(\tau)}{\tau} e^{-(t-t')/\tau} d\tau \quad (1.3)$$

where  $e^{mr}$  is the strain-rate tensor,  $T^{ik}$  is the stress tensor,  $\pi$  is the isotropic pressure,  $g^{ik}$  is the conjugate metric tensor of the fixed coordinate system  $x^i$ ,  $N(\tau)$  is the relaxation-time distribution function,  $x'^i = x'^i(x^i, t, t')$  is a fixed Lagrangian coordinate system (displacement function), and  $t$  is the current time,  $t > t'$ . Equation (1.2) may be treated as the equation of media whose behavior at low velocities may be characterized by the relaxation-time spectrum. The B' fluid exhibits a positive Weissenberg effect when there is shear between rotating cylinders, and it has a normal-stress distribution analogous to that found in [2]. In addition, the values of  $N(\tau)$  have been determined for several real materials [3].

The (1.2) form of the model of the B' fluid has a drawback: it is not suited to the description of non-Newtonian viscosity. For our problem the strain rates are small, so that we may neglect anomalies of viscosity. Besides the generalization of "contravalent" type, there is the A' fluid, which is the "covariant" analog.

2. In the Boussinesq approximation the equations of the perturbed state are written as

$$\frac{\partial u_i}{\partial t} = -\frac{1}{\rho} \frac{\partial \pi'}{\partial x_i} + \lambda_i \alpha g \theta + \frac{1}{\rho} \frac{\partial \tau_{ij}}{\partial x_j} \quad (2.1)$$

$$\left( \frac{\partial}{\partial t} - \kappa \Delta \right) \theta = \beta u_i \lambda_i \quad (2.3)$$

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$$\frac{\partial u_i}{\partial x_i} = 0 \quad (2.3)$$

where  $\lambda_i = (0, 0, 1)$ ,  $\beta$  is the negative temperature gradient,  $\kappa$  is the thermal-conductivity coefficient of the fluid,  $\pi'$ ,  $\theta$ ,  $u_i$  are the perturbations in the pressure, temperature, and velocity,  $\rho$  is the density of the fluid,  $\alpha$  is the coefficient of thermal expansion,  $g$  is the gravitational acceleration, and  $\tau_{ij}$  is the tensor representing the stress due to perturbed motion.

The perturbation and temperature coefficients are written as

$$u_j(x_i) = u_j^0(x_3) \exp[i(a_1 x_1 + a_2 x_2) + pt] \quad (2.4)$$

$$\theta(x_i) = \theta^0(x_3) \exp[i(a_1 x_1 + a_2 x_2) + pt] \quad (2.5)$$

Here  $a_1, a_2$  are the wave numbers, and  $p$  is the complex damping.

The displacement functions of perturbed motion have the form

$$\begin{aligned} x_1' &= x_1 + \alpha(x_i, x_i', t, t') \\ x_2' &= x_2 + \zeta(x_i, x_i', t, t') \\ x_3' &= x_3 + \gamma(x_i, x_i', t, t') \end{aligned} \quad (2.6)$$

where  $\alpha, \zeta, \gamma$  satisfy the initial conditions

$$\alpha|_{t=t'} = 0, \quad \zeta|_{t=t'} = 0, \quad \gamma|_{t=t'} = 0 \quad (2.7)$$

We have the system of equations

$$\frac{\partial x_i'}{\partial t} + u_j \frac{\partial x_i'}{\partial x_j} = 0 \quad (2.8)$$

for determining  $x_i'$ .

If we restrict the discussion to fluids having "short" memory, we may write

$$x_i' = x_i - (t - t') u_i \quad (2.9)$$

After determining the components of the perturbed-motion stress tensor from (1.2) and eliminating  $\pi'$ , following [4], we arrive at the amplitude equations

$$(D^2 - \gamma^2) [\sigma \text{Pr}^{-1} - (1 - \sigma \Gamma)(D^2 - \gamma^2)] W = R \gamma^2 T \quad (2.10)$$

$$[\sigma - (D^2 - \gamma^2)] T = W \quad (2.11)$$

Here we employ the following dimensionless variables:

$$\begin{aligned} (x, y, z) &= \left( \frac{x_1}{d}, \frac{x_2}{d}, \frac{x_3}{d} \right), \quad \text{Pr} = \frac{\eta_0}{\rho \kappa} \\ R &= \frac{\alpha g \beta d^4 \rho}{\eta_0 \kappa}, \quad \tau = \frac{\kappa t}{d^2}, \quad W = \frac{u_3 d}{\kappa} \\ T &= \frac{\theta}{\beta d}, \quad \sigma = \frac{p d^2}{\kappa}, \quad D = \frac{\partial}{\partial z} \\ a_i^* &= a_i d, \quad \gamma^2 = a_1^{*2} + a_2^{*2} \end{aligned} \quad (2.12)$$

where  $\text{Pr}$  is the Prandtl number,  $R$  is the Rayleigh number,  $d$  is the thickness of the fluid layer,  $\Gamma = \kappa \eta_0^{-1} d^{-2} \int_0^\infty \tau N(\tau) d\tau$  is the elasticity parameter, and  $\eta_0$  is the Newtonian viscosity.

3. A monotonic or oscillatory instability will arise, depending on the properties of the fluid. It has been established for a Newtonian fluid that the "principle of monotonic perturbations" is always satisfied and that the stability threshold is determined for zero damping. This principle is violated when there is elasticity in non-Newtonian viscoelastic media.

Following [5], for the given case we may show that

$$\text{Im} \sigma \left\{ \int_0^1 |F|^2 dz + R \gamma^2 \int_0^1 (|DM|^2 + \gamma^2 |W|^2) dz - R \text{Pr} \gamma^2 \Gamma \int_0^1 |J|^2 dz \right\} = 0 \quad (3.1)$$

$$J = (D^2 - \gamma^2) W, \quad F = [\sigma \text{Pr}^{-1} - (1 - \sigma \Gamma)(D^2 - \gamma^2)] J$$

The integral (3.1) is not of fixed sign, and for  $R > 0$  the perturbations are only monotonic for small  $\Gamma$ .

4. Let us consider the solution of the problem for two free boundaries. In this case the boundary conditions are written as

$$T = W = D^2W = 0 \quad (z = 0, 1) \quad (4.1)$$

The solution satisfying (4.1) is written as

$$W = W_0 \sin n\pi z \quad (n = 1, 2, \dots) \quad (4.2)$$

where  $W_0$  is a constant.

For the fundamental instability mode the damping equations are found from (2.10), (2.11):

$$\sigma^2 + \sigma^2 \kappa^2 \left( 1 + \frac{\text{Pr}}{1 - \Gamma \text{Pr} x^2} \right) + \frac{\text{Pr} x^4}{(1 - \Gamma \text{Pr} x^2)} - \frac{R \text{Pr} x^2}{x^2 (1 - \Gamma \text{Pr} x^2)} = 0$$

$$x^2 = \pi^2 + \gamma^2 \quad (4.3)$$

For a Newtonian fluid, where  $\Gamma = 0$ , the values of the critical Rayleigh number and wave number coincide with the known values. A detailed discussion of the damping rates for this case is given in [4].

Assume  $\text{Re } \sigma = 0$ , on the instability boundary; then letting  $\sigma = i\omega_+$ , we may obtain the conditions for appearance of an oscillatory instability. We find the dimensionless frequency of neutral oscillation

$$\omega_+^2 = -\frac{\gamma^2}{x^2} [R_*^{(s)} - R] \quad (4.4)$$

from (4.3); here  $R_*^{(s)} = \chi^6 / \gamma^2$  is the critical Rayleigh number for steady-state instability. The neutral-oscillation frequency is a real quantity, so that (4.4) is only valid for  $R > R_*^{(s)}$ . The oscillatory instability appears later, so that the stability threshold is determined by  $R_*^{(s)}$ .

5. The two-rigid boundary problem cannot be solved in elementary form. Various approaches have been suggested. Here we use the Galerkin method, as was done in [6]. Shifting the origin to the middle of the layer, we write the boundary conditions

$$T = W = DW = 0 \quad (z = \pm 1/2) \quad (5.1)$$

for ideally heat-conducting boundaries.

We seek solutions of (2.10) and (2.11) in the form

$$W(z) = \sum_{m=1}^M a_m W_m(z) \quad (5.2)$$

$$T(z) = \sum_{m=1}^M b_m T_m(z) \quad (5.3)$$

where  $a_m$  and  $b_m$  are unknown coefficients. The basic functions  $W_m(z)$  and  $T_m(z)$  must satisfy the boundary conditions (5.1). Here we shall only consider even solutions, as being the least stable.

We take as the basic functions [7]

$$W_m(z) = \frac{\text{ch}(\mu_m z)}{\text{ch} \mu_m / 2} - \frac{\cos(\mu_m z)}{\cos \mu_m / 2} \quad (5.4)$$

$$T_m(z) = A_m \cos(2m - 1) \pi z \quad (5.5)$$

where  $\mu_m$  are positive roots of the equation

$$\text{th} \frac{\mu}{2} + \text{tg} \frac{\mu}{2} = 0 \quad (5.6)$$

The amplitude coefficient  $A_m$  is chosen on the basis of the normalization condition.

TABLE 1

$\Gamma$	Pr	$\gamma_*$	$R_* \cdot 10^{-5}$
0.001	10	33.01	12.608
	100	31.62	10.646
0.005	0.1	46.77	50.000
	1.0	19.71	1.697
	10	14.48	0.535
	100	13.86	0.455
0.01	0.1	32.98	12.556
	1.0	13.71	0.438
	10	9.98	0.145
	100	9.54	0.126
0.02	0.1	23.18	3.167
	1.0	9.34	0.117
	10	6.67	0.045
	100	6.38	0.0404
0.05	0.1	14.37	0.519
	1.0	5.01	0.025
	10	3.35	0.0174
	100	3.20	0.0171
	0.1	9.74	0.134
	1.0	1.93	0.022
	10	0.62	0.139
	100	0.39	0.333

After certain manipulations we arrive at a system of equations in the unknown coefficients  $a_m$ ,

$$\sum_{m=1}^M a_m \left[ E_{mn} + Rc_4 \sum_{i=1}^M \frac{F_{im} F_{in}}{J_{ii}} \right] = 0 \quad (5.7)$$

This reduces to algebraic equations of degree M for R,

$$\left\| E_{mn} + Rc_4 \sum_{i=1}^M \frac{F_{im} F_{in}}{J_{ii}} \right\| = 0 \quad (5.8)$$

Here we let

$$\begin{aligned} E_{mn} &= c_1 [D^4 W_m, W_n] - c_2 [D^2 M_m, W_n] + c_3 [W_m, W_n] \\ F_{mn} &= [T_m, W_n], \quad J_{mn} = [D^2 T_m, T_n] - [T_m, T_n] c_5 \\ c_1 &= (1 - \sigma\Gamma), \quad c_2 = \sigma Pr^{-1} + 2(1 - \sigma\Gamma) \gamma^2 \\ c_3 &= \sigma Pr^{-1} \gamma^2 + (1 - \sigma\Gamma) \gamma^4 \\ c_4 &= \gamma^2, \quad c_5 = \gamma^2 + \int_{-0.5}^{+0.5} UV dz \end{aligned} \quad (5.9)$$

A Nairi computer was used for numerical determination of the critical Rayleigh number for the first and second approximations. For the range of parameters studied no oscillatory instability was found to appear. The critical Ray-

leigh numbers found on the assumption of oscillatory instability were large. Table 1 shows the calculated data. A check on the critical Rayleigh number for  $\Gamma = 0$  showed full agreement of our values with [5].

6. Knowing the specific form of the function  $N(\tau)$  we may go over to relationships valid for different models.

Newtonian fluid [5]:

$$N(\tau) = \eta_0 \delta(\tau) \quad (6.1)$$

Maxwellian fluid [8]:

$$N(\tau) = \eta_0 \delta(\tau - \lambda_1) \quad (6.2)$$

$$\Gamma = \lambda_1 \kappa / d^2 \quad (6.3)$$

Oldroyd fluid (type B) [6]:

$$N(\tau) = \eta_0 \frac{\lambda_2}{\lambda_1} \delta(\tau) + \eta_0 \frac{\lambda_1 - \lambda_2}{\lambda_1} \delta(\tau - \lambda_1) \quad (6.4)$$

$$\Gamma = \frac{\lambda_1 \kappa}{d^2} \left( 1 - \frac{\lambda_2}{\lambda_1} \right) \quad (6.5)$$

where  $\lambda_1$  is the relaxation time,  $\lambda_2$  is the retardation time, and  $\delta(\tau)$  is the Dirac delta function.

Linear viscoelastic fluid [9]:

$$(1 - \sigma\Gamma) = \frac{1}{\eta_0} \int_0^\infty N(\tau) e^{-p\tau} d\tau = \frac{\eta(p)}{\eta_0} \quad (6.6)$$

For  $p = 0$ ,  $\eta_0 = \eta(0)$  is the maximum Newtonian viscosity.

If  $p = i\omega$ , then  $\eta(i\omega) = \eta'(\omega) - i\eta''$  is the complex viscosity, defined in the linear theory of viscoelasticity.

There is a relationship between a "second-order" fluid and a B' fluid [10].

We write the equation describing the second-order fluid [11] as

$$T_{ij} = -\pi \delta_{ij} + \Phi_1 e_{ij} + \Phi_2 a_{ij} + \Phi_3 e_{ik} e_{kj} \quad (6.7)$$

$$a_{ij} = \frac{D e_{ij}}{Dt} + e_{ik} \frac{\partial v_k}{\partial x_j} + e_{kj} \frac{\partial v_k}{\partial x_i} \quad (6.8)$$

TABLE 2

Pr	$\lambda_{+1}$	$R_*$			$\gamma_*$			$\omega_{+*}$		
		[°]	[°]	from (5.8)	[°]	[°]	from (5.8)	[°]	[°]	from (5.8)
1.0	0.1	877.8	894.9	893.1	4.917	4.869	4.9	15.07	14.92	14.95
1.0	1.0	51.58	51.28	51.18	3.696	3.621	3.6	6.061	6.061	6.031
10	0.1	230.0	235.2	234.6	7.309	7.198	7.3	76.68	75.64	76.6
10	1.0	7.496	7.521	7.507	4.72	4.658	4.7	20.77	20.71	20.74
100	0.1	130.1	133.9	133.8	11.96	11.75	11.7	385.8	376.8	377.0
100	1.0	2.203	2.237	2.232	7.297	7.145	7.2	83.45	82.24	82.73
1000	0.1	108.0	112.0	111.2	20.46	19.99	20.1	2052.0	2006	2017
1000	1.0	1.289	1.329	1.322	12.76	11.74	11.8	418.8	389.0	390.9

where  $\delta_{ij}$  is the Kronecker symbol, and  $\varphi_i$  is the viscometric function.

The formal series of solutions of (2.8) for the displacement function is

$$x_i' = \sum_{n=0}^{\infty} \frac{1}{n!} (t' - t)^n \frac{D^n x_i}{Dt^n} \quad (6.9)$$

and takes the form of the Taylor-series expansion in the reciprocal time.

In like manner we may write

$$e_{ij}(x', t') = \sum_{n=0}^{\infty} \frac{1}{n!} (t' - t)^n \frac{D^n e_{ij}(x, t)}{Dt^n} \quad (6.10)$$

Using (6.9) and (6.10), taking into account first-order terms we may obtain

$$T_{ik} = -\pi \delta_{ik} + \int_{-\infty}^t \psi(t-t') [e_{ik} + (t'-t)a_{ik}] dt' + \dots \quad (6.11)$$

from (1.2).

Thus,

$$T_{ik} = -\pi \delta_{ik} + \varphi_1 e_{ik} + \varphi_2 a_{ik} + \dots \quad (6.12)$$

$$\varphi_1 = \int_{-\infty}^t \psi(t-t') dt' \quad (6.13)$$

$$\varphi_2 = \int_{-\infty}^t (t'-t) \psi(t-t') dt' \quad (6.14)$$

Using (1.3) we find

$$\eta_0 = \varphi_1 = \int_0^{\infty} N(\tau) d\tau \quad (6.15)$$

$$\varphi_2 = - \int_0^{\infty} \tau N(\tau) d\tau \quad (6.16)$$

$$\Gamma = -\varphi_2 \kappa / \eta_0 d^2 \quad (6.17)$$

Equation (2.10) may be reduced to the familiar form for Maxwell and Oldroyd (type B) fluids provided we neglect the term  $(\sigma\Gamma)^2$ . When the initial equation (2.10) was obtained, only the linear terms were considered in the expansion of the expression occurring in the stress-tensor components,

$$\eta = \int_0^{\infty} \frac{N(\tau)}{1+p\tau} d\tau \quad (6.18)$$

TABLE 3.

Fluid model	Form of boundary conditions		Source
	both bound- aries free	both bound- aries rigid	
Fluid B Oldroyd	— Oscillatory	Steady-state Oscillatory	[17] [8, 12]
Maxwellian fluid	Oscillatory	Oscillatory	[8, 9]
"Second-order" fluid	—	Steady-state	[18-20]
Integral model	—	Steady-state	[19, 20]

This corresponds to a fluid having weak elasticity, where  $N(\tau)$  decreases rapidly. As was shown in [6], if the elasticity of the fluid is less than the critical value, the principle of monotonic perturbation will hold. Replacing (6.18) by the expression for the complex viscosity brings out the characteristic features of viscoelastic fluids [9]. In this case the problem has been solved for two rigid boundaries and realized in an Odra-1204 computer. Table 2 gives the data for the critical parameters. It was assumed that the dimensionless quantities satisfy the relationships  $\lambda_{+1}\omega_{+} = \lambda_1\omega$ ,  $\lambda_{+1} = \lambda_1(x/d^2)$  [9].

The presence of relaxation time reduces stability; retardation time stabilizes the layer. Vest and Arpace [8] took the constitutive Maxwell equation to be sufficient for detection of the fundamental effects produced by the influence of viscoelasticity on thermal instability. There is a sharp difference in the behavior of Maxwellian and Oldroyd fluids [6]. Thus, adequacy of the rheological model adopted is essential for detection of an oscillatory instability.

Although oscillatory instability is theoretically possible, it cannot be observed experimentally as has been shown in the publications cited [12]. On the assumption of steady-state instability it has been possible to make an experimental determination of the maximum Newtonian viscosity, which is a fundamental rheological characteristic of a fluid [13-16].

Table 3 gives data on the appearance of a convective instability for various models of viscoelastic fluids heated from below.

Fluids having nonlinear viscosity have not been considered here. This property of a fluid is a destabilizing factor [14-16]. No investigation was made with simultaneous allowance for elasticity and nonlinear viscosity.

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