

ON OSCILLATORY NECKING IN POLYMERS*

By

B. J. MATKOWSKY AND G. I. SIVASHINSKY**

Northwestern University

Abstract. The phenomenon of oscillatory necking in the stretching of polyethylene films is described. We propose an extension of a model of Barenblatt for isothermic necking in polymers, and we show that oscillatory necking may arise, for example, in polymers for which the effect of diffusion of stresses exceeds that of diffusion of oriented material. In such polymers, at sufficiently low temperatures, uniform necking is no longer stable and self-oscillations are observed.

1. Introduction. It is well known that when polyethylene films are stretched by external stresses, uniform necking is observed. That is, an orientation wave of the polymer material propagates. This phenomenon was first observed and described in 1932 by Carothers and Hill [1] (see also Nadai [2]). A theory of the phenomenon was proposed by Barenblatt [3, 4], using an analogy between necking and the propagation of a flame in a premixed gaseous combustible mixture. It is known (see, e.g., Zeldovich [5]) that the uniform propagation of a flame is maintained by the interaction of several effects including diffusion of the component limiting the combustion reaction, heat release in the reaction zone, and the flow of heat from the reaction zone into the region of the cold fresh mixture.

The necking analogue of the concentration of combustion products is the concentration n of oriented material, which is assumed to be zero ($n = 0$) far ahead of the conversion zone (the necking front) and to reach its maximum ($n = 1$) behind this zone. The principal mechanism of flame propagation is conduction of heat; the analogous factor in polymer stretching is the diffusion of stresses from the narrower region (a) of oriented material to the region (b) of nonoriented material (cf. Fig. 1).

Necking is often accompanied by heating, and the orientation of the polymer may be regarded as an exothermic reaction. Such is the case e.g. in the model of Barenblatt, Entov and Segalov [6] who considered necking caused by the transfer of heat generated in the reorientation zone. However, in the experiments of Lazurkin [6] and Vincent [7], the drawing of the polymer was carried out so slowly that the temperature could not have varied significantly at any point in the sample. Nevertheless, necking was clearly observed. Here, heat conduction cannot have been the decisive mechanism in necking. Therefore, in Barenblatt [3, 4], no attention was paid to the nonuniform temperature distribution. That is, heat release was ignored and the temperature of the polymer being stretched was treated as constant.

* Received November 26, 1977; revised version received May 4, 1978. This research was supported in part by N.S.F.-MCS77-25660, A.R.O.-DAAG29-77-G-0222, and D.O.E.-ER78-S-02-4650.

** Permanent address: Tel-Aviv University, Ramat-Aviv, Israel.

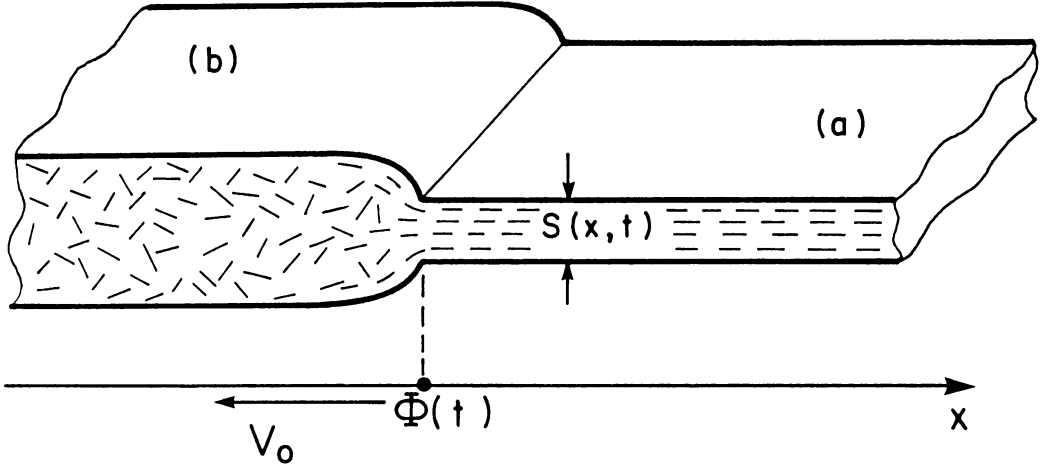


FIG. 1.

Another premise of the Barenblatt theory is that the propagation of the polymer orientation wave is a quasistationary and quasi-one-dimensional process. Thus

$$\frac{\partial(\sigma S)}{\partial x} = 0 \quad \text{or} \quad \sigma S = \sigma_{\infty} S_{\infty} \quad (1)$$

where $\sigma = \sigma(x, t)$ is the stress acting on the cross-section of the film at a point x at time t (see Fig. 1); $S = S(x, t)$ is the thickness of the film at (x, t) ; σ_{∞} , S_{∞} are the stress and thickness in the region corresponding to the final, oriented state of the polymer.

In the region of the initial, nonoriented state ($x \rightarrow -\infty$),

$$S = S_0 = S_{\infty}/\alpha, \quad \sigma = \alpha\sigma_{\infty} \quad (\alpha < 1) \quad (2)$$

where α is a given constant determined by the change in the structure of the polymer material in the process of stretching. The rate of the orientation reaction is *strongly* dependent on the local stress (the dependence involves an exponential, like the Arrhenius law). The necking process may therefore be fully described even when there is a small stress drop, i.e., when the change in S is small. In that case we may write Eq. (1) approximately as

$$\sigma \simeq 2\sigma_{\infty} - S\sigma_{\infty}/S_{\infty}. \quad (3)$$

The change in the density of the polymer material during orientation is not essential for an understanding of necking. For this reason, the next assumption of the Barenblatt theory [3, 4] is that the density of the material is constant.

Since the density is assumed constant and the change in thickness small, it follows that the motion of the neck induces almost no motion of the polymer material. That is to say, to a first approximation, the orientation wave propagates in motionless material. In view of the assumptions made, the variation in the concentration n is described by the equation of diffusion in a motionless medium:

$$(\partial n/\partial t) + (\partial/\partial x)j_n = W \quad (4)$$

where j_n is the flow of oriented material due to transport effects, and W the rate of the orientation "reaction," which is a function of n , σ and the temperature T .

Barenblatt suggests the Arrhenius law as the function describing the behavior of W

$$W = z(1 - n) \exp \left(- \frac{U - \gamma\sigma}{KT} \right) \quad (5)$$

where U is the activation energy of bonds broken during the conversion process, k is Boltzmann's constant, T the absolute temperature, γ a constant depending on the structure of the material, and z a constant (the "pre-exponential").

The strong stress-dependence of W is due to the assumption that the quantity $\gamma\sigma_\infty/kT$ is large (typically about twenty-thirty or so.) It should be noted that this quantity appears in the exponent.

The flow of oriented material j_n is assumed to have the classical form¹

$$j_n = -\mu(\partial n/\partial x). \quad (6)$$

The coefficient of diffusion μ is assumed to be a known function of σ and T . It is natural to assume that the transfer coefficient is a monotonically increasing function of σ , vanishing at $\sigma = 0$. We also assume that this dependence is weaker than that of the conversion rate, say a power function. Then in view of the assumption that σ varies only slightly, we may put

$$\mu(\sigma, T) = \mu(\sigma_\infty, T) \quad (T = \text{const.}). \quad (7)$$

To complete the system of equations (3), (4), Barenblatt [3, 4] proposes a simple relationship between the cross-sectional thickness S and the concentration of oriented matter, given by

$$S/S_0 = 1 - (1 - \alpha)n. \quad (8)$$

From (2), (3) and (8), we obtain

$$\sigma/\sigma_\infty = \alpha + (1 - \alpha)n. \quad (9)$$

Thus, condition (8) is equivalent to the assumption that the distribution of stress and the concentration of oriented material are *similar*. It is this assumption which we shall later relax.

We note that the system (3), (4), (8), (9) admits a simple analytical solution—a wave propagating at constant velocity:

$$n = n(x - V_n t), \quad n(+\infty) = 1, \quad n(-\infty) = 0. \quad (10)$$

This solution describes normal necking. The necking rate V_n is uniquely determined by the parameters of the problem.

2. Instability of normal necking. In 1970, Andrianova, Kargin and Kechekyan [8], studying stretching of polyethylene terephthalate films, observed that under certain conditions the necking process is not uniform but proceeds in regular jumps. The onset of oscillatory necking was found to be highly sensitive to the temperature maintained during stretching and to the heat transfer conditions. Fig. 2 is a photograph of a film being stretched at temperatures of 85°C (a) and 15°C (b) respectively. The photographs are the result of an informal experiment performed by the authors (B.M. and G.S.). In the first

¹ We note that Barenblatt uses a slightly more general form.

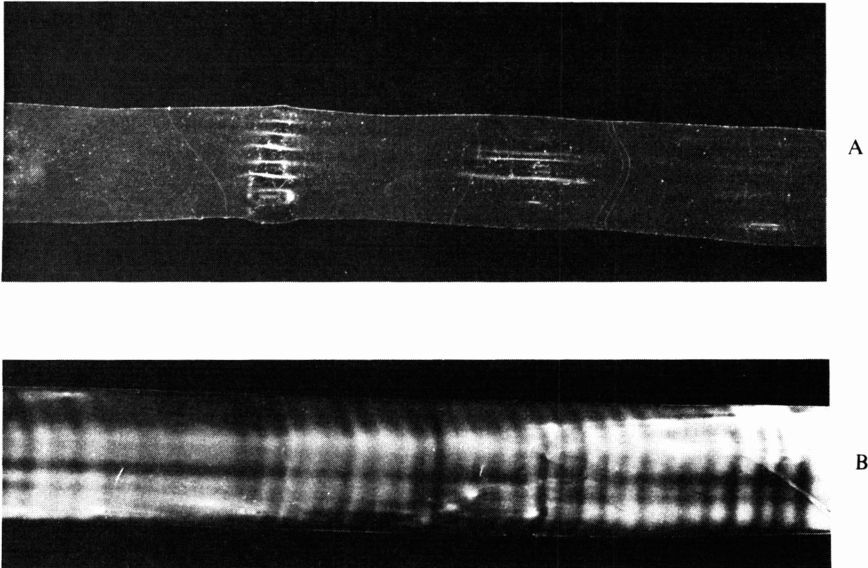


FIG. 2.

case the neck propagates uniformly, leaving behind it homogeneous oriented material. In the second case the necking is oscillatory, as evidenced by the regularly alternating transparent and dull strips in the region of oriented material.

On the basis of the isothermic model of Barenblatt, wavelike necking as described by Eq. (10) is known to be stable. (A mathematically equivalent problem of flame theory was considered by Barenblatt and Zeldovich [9].) Barenblatt, therefore, in his theoretical description of oscillatory necking [10], discarded the isothermic model, and proposed taking into consideration thermal effects, transfer of heat to and from the external medium, and the elastic properties of the material. He constructed a zero-dimensional model of the necking process, which disregards the spatial distribution of stress and temperature, considering only their mean values. He showed that in a certain parameter interval self-oscillation was indeed possible.

In this paper, we show that if the similarity condition (9) is dropped and replaced by a somewhat more general condition, oscillatory necking may also take place in the framework of the *isothermic* model. Moreover, in accordance with the observed situation (Fig. 2), we find that, in polymers which in principle admit self-oscillations, the effect may be suppressed by keeping the polymer at sufficiently high temperatures, or re-induced by keeping it at sufficiently low temperatures.

3. Modification of the Barenblatt model. Fundamental equations. It follows from assumption (9) that the change in stress concentration is described by an equation of type (4):

$$\frac{\partial \sigma}{\partial t} + \frac{\partial}{\partial x} j_{\sigma} = \sigma_{\infty}(1 - \alpha)W \quad (11)$$

where

$$j_{\sigma} = (1 - \alpha)\sigma_{\infty}j_n. \quad (12)$$

In fact, conditions (9) and (12) are equivalent. In combustion theory, a condition of type (9) represents the similarity of the concentration and temperature fields, which occurs at a Lewis number (L) of unity. (Recall that the Lewis number defines the ratio of the conductivity of the gaseous mixture to the diffusivity of the limiting component.) Though the Lewis number is usually close to unity, even a small departure from similarity ($L = 1$) may in some cases lead to a number of nontrivial effects (see e.g. [11, 12, 14]). In this sense, the case of ideal similarity ($L = 1$) is quite exceptional.

Therefore, in the necking problem it seems quite natural to replace condition (9) by the more general equation

$$J_\sigma = -\sigma_\infty \lambda (\partial \sigma / \partial x), \quad \lambda = \lambda(\sigma_\infty, \Upsilon). \quad (13)$$

We note that it is no longer mandatory that $j_\sigma = \sigma_\infty(1 - \alpha)j_n$ which occurs for $\lambda = \mu$. The adoption of (13) in place of (12) considerably enlarges the number of possible situations with stable necking.

We now show that the isothermic model described by Eqs. (4), (11) and (13) does indeed admit oscillatory necking. The system of equations (4), (11) becomes

$$\partial \theta / \partial t = \lambda (\partial^2 \theta) / \partial x^2 + W(n, \theta), \quad (14)$$

$$\partial n / \partial t = \mu (\partial^2 n) / \partial x^2 + W(n, \theta), \quad (15)$$

where $\sigma / \sigma_\infty = \alpha + (1 - \alpha)\theta$. The case of equality, $\lambda = \mu$, corresponds to similarity, when n and θ are linearly related by Eq. (9). We assume that the temperature dependencies of λ and μ are similar. Then λ / μ is a temperature-independent quantity, which we denote by L :

$$\lambda / \mu = L. \quad (16)$$

For the sequel, it is convenient to transform to dimensionless variables:

$$x' = xV_n / \lambda, \quad \tau = tV_n^2 / \lambda, \quad w = W\lambda / V_n^2. \quad (17)$$

As stated previously, because of the strong stress-dependence of the orientation-reaction rate W , the width of the reaction zone is very narrow, concentrated in the vicinity of a certain moving surface $x' = \phi(\tau)$ which we call the necking front. Normal necking corresponds to $\phi(\tau) = -\tau$ (the necking front moves with unit velocity).

We introduce a coordinate system attached to the front, setting

$$\xi = x' - \Phi(\tau) \quad (18)$$

in terms of which Eqs. (14) and (15) become (using Eqs. (16), (17))

$$\frac{\partial \theta}{\partial \tau} - \Phi_\tau \frac{\partial \theta}{\partial \xi} = \frac{\partial^2 \theta}{\partial \xi^2} + w(\theta, n), \quad (19)$$

$$\frac{\partial n}{\partial \tau} - \Phi_\tau \frac{\partial n}{\partial \xi} = \frac{1}{L} \frac{\partial^2 n}{\partial \xi^2} + w(\theta, n). \quad (20)$$

As remarked above, Barenblatt considered the case $L = 1$. We shall consider the case of L close to but not equal to one. At the same time we consider the case in which $N \equiv (\gamma\sigma_\infty(1 - \alpha)/kT) \gg 1$, and $\nu \equiv N(1 - L) = O(1)$. In this case we now show that the orientation rate w may be approximated by a point source at $\xi = 0$ (i.e., $x' = \phi(\tau)$), given by

$$w \simeq \exp\left(\frac{1}{2}N(\theta - 1)\right) \delta(\xi) \quad (21)$$

(where $\delta(\xi)$ is a Dirac delta function and the term $N[\theta(0, \tau) - 1]$ is $O(1)$). First, however, we note that for $\xi > 0$ the material reaches the state of complete orientation:

$$n(\xi, \tau) \equiv 1 \quad \text{at} \quad \xi > 0. \quad (22)$$

Far ahead of the necking front, the concentration of oriented material is zero, while the stress is equal to its initial value. Far behind the front, the film contracts and the stress reaches a certain maximum mean value. In the event of oscillatory necking, this mean value will differ from σ_∞ , the latter value corresponding to normal necking. This implies the following boundary conditions:

$$\theta(-\infty, \tau) = 0, \quad n(-\infty, \tau) = 0, \quad \theta(+\infty, \tau) < \infty. \quad (23)$$

To show that for $N \gg 1$ and $\nu = N(1 - L) = 0$ (1), the reaction rate may be approximated by the point source given by (21), we note that in terms of parameters defined above, the reaction rate is given by

$$w = AN^2(1 - n) \exp N(\theta - 1) \quad (24)$$

with A defined as

$$A = \frac{\lambda z}{V_n^2} \exp \frac{\gamma \sigma_\infty - U}{NkT}. \quad (25)$$

The width of the reaction zone is very narrow, due to the strong stress dependence of w as evidenced by the term $\exp N(\theta - 1)$ in (24). In our analysis we treat this zone as an internal boundary layer, of width $O(1/N)$, about the moving front. In the limit as $N \rightarrow \infty$, the reaction rate is approximated by a concentrated source on the front. From the form of the reaction rate (24), it is clear that the concentrated source may be represented in the limit by a Dirac delta function. The strength Q of that source will be derived by an asymptotic analysis in the parameter $1/N$. We consider the regions both within and outside of the boundary layer by the method of matched asymptotic expansions.

In the region outside of the reaction layer, we seek expansions, which we refer to as outer expansions, of the form

$$\theta_\pm \sim \sum_{j=0} \theta_\pm^j \left(\frac{1}{N}\right)^j, \quad (26)$$

$$n_\pm \sim \sum_{j=0} n_\pm^j \left(\frac{1}{N}\right)^j, \quad (27)$$

$$A \sim \sum_{j=0} A_j \left(\frac{1}{N}\right)^j, \quad (28)$$

$$Q \sim \sum_{j=0} Q_j \left(\frac{1}{N}\right)^j, \quad (29)$$

$$\Phi \sim \sum_{j=0} \Phi_j \left(\frac{1}{N}\right)^j, \quad (30)$$

where \pm refers to the regions $\xi > < 0$ respectively. We recall that L is given in terms of N by

$$L = 1 - (\nu/N), \quad (31)$$

and employ (26)–(31) in Eqs. (19) and (20). We note that $n_\pm^j \equiv \delta_{0j}$, where δ_{0j} is the Kronecker delta since the region $\xi > 0$ is the region of complete orientation. Thus, in the

region $\xi > 0$ the reaction term vanishes. Similarly in the region $\xi < 0$, the reaction term is transcendentally small since $\theta_-^0 < 1$. This follows from the fact that $\theta_-^0 \geq 1$ implies $n_-^0 \equiv 1$ which does not satisfy the boundary condition that $n(-\infty, \tau) = 0$.

Thus as far as the outer regions are concerned, the reaction term may be replaced by the concentrated source on the front. The strength Q of the source will be computed from an analysis of the boundary layer. In the outer regions therefore, the resulting equations for $\theta_{\pm}^0, n_{\pm}^0$ are given by

$$\frac{\partial \theta^0}{\partial \tau} - (\Phi_0)_\tau \frac{\partial \theta^0}{\partial \xi} = \frac{\partial^2 \theta^0}{\partial \xi^2} + Q_0 \delta(\xi), \quad (32)$$

$$\frac{\partial n^0}{\partial \tau} - (\Phi_0)_\tau \frac{\partial n^0}{\partial \xi} = \frac{\partial^2 n^0}{\partial \xi^2} + Q_0 \delta(\xi), \quad (33)$$

where the source strength Q_0 is as yet unknown. The boundary conditions for (32)–(33) are

$$\theta^0(-\infty, \tau) = n^0(-\infty, \tau) = 0, \quad (34)$$

$$\theta^0(\infty, \tau) < \infty, \quad n^0(\infty, \tau) = 1.$$

Subtracting Eq. (33) from (32), we see that $\theta^0 - n^0$ satisfies a linear parabolic equation with no source term. Therefore, the boundary conditions imply that

$$\theta_-^0 \equiv n_-^0, \quad (35)$$

$$\theta_+^0 \equiv n_+^0 + c. \quad (36)$$

We note that Eqs. (32)–(33) imply that θ^0 and n^0 are continuous at $\xi = 0$, but their derivatives have jump discontinuities of magnitudes $-Q_0$. The continuity conditions imply that $c = 0$ so that

$$\theta_+^0 \equiv n_+^0 \quad (37)$$

By computing the jump in the derivatives of θ^0 and n^0 across $\xi = 0$, i.e. across the boundary layer, we will have computed the source strength Q_0 . Thus we now turn to the boundary layer analysis.

In the boundary layer we introduce the variable η by the stretching transformation

$$\eta = N\xi, \quad (38)$$

and seek expansions of the form

$$\theta \sim \sum_{j=0} \tilde{\theta}^j(\eta, \tau) \left(\frac{1}{N}\right)^j, \quad n \sim \sum_{j=0} \tilde{n}^j(\eta, \tau) \left(\frac{1}{N}\right)^j$$

for $-\infty < \eta < \infty$. The boundary conditions as $|\eta| \rightarrow \infty$ are obtained by matching to the outer solutions. The leading terms of these expansions are then given by

$$\tilde{\theta}^0 \equiv \tilde{n}^0 \equiv 1. \quad (39)$$

Then $\tilde{\theta}^1$ and \tilde{n}^1 satisfy

$$\frac{\partial^2 \tilde{\theta}^1}{\partial \eta^2} - A_0 \tilde{n}^1 \exp \tilde{\theta}^1 = 0, \quad (40)$$

$$\frac{\partial^2 \tilde{n}^1}{\partial \eta^2} - A_0 \tilde{n}^1 \exp \tilde{\theta}^1 = 0. \quad (41)$$

Therefore

$$\frac{\partial^2(\tilde{\theta}^1 - \tilde{n}^1)}{\partial \eta^2} = 0, \quad (42)$$

so that

$$\tilde{\theta}^1 - \tilde{n}^1 = \alpha \eta + \beta, \quad (43)$$

where the $\alpha = \alpha(\tau)$ and $\beta = \beta(\tau)$ are determined from the matching conditions as $\eta \rightarrow \infty$ to be

$$\alpha = 0, \quad (44)$$

$$\beta = \tilde{\theta}^1(\infty, \tau) = \theta^1(0^+, \tau). \quad (45)$$

Therefore

$$\tilde{n}^1(\eta, \tau) = \tilde{\theta}^1(\eta, \tau) - \theta^1(0^+, \tau), \quad (46)$$

and Eq. (40) becomes

$$\frac{\partial^2 \tilde{\theta}^1}{\partial \eta^2} + A_0 \{\theta^1(0^+, \tau) - \tilde{\theta}^1\} \exp \tilde{\theta}^1 = 0. \quad (47)$$

Multiplying (47) by $2 \tilde{\theta}_\eta^1$, and integrating from ∞ to η , and then letting $\eta \rightarrow -\infty$, we obtain

$$(\tilde{\theta}_\eta^1(-\infty, \tau))^2 + 2A_0 \int_{\tilde{\theta}^1(\infty, \tau)}^{-\infty} \{\tilde{\theta}^1(\infty, \tau) - \tilde{\theta}^1(\eta, \tau)\} \exp \tilde{\theta}^1 d\tilde{\theta}^1 = 0. \quad (48)$$

In the derivation of (48) we have used the results that

$$\lim_{\eta \rightarrow -\infty} \tilde{\theta}_\eta^1(\eta, \tau) = 0, \quad (49)$$

$$\lim_{\eta \rightarrow -\infty} \tilde{\theta}^1(\eta, \tau) = -\infty, \quad (50)$$

which are obtained from the matching conditions. Solving

Eq. (48) for $\tilde{\theta}_\eta^1(-\infty, \tau)$ we obtain

$$\tilde{\theta}_\eta^1(-\infty, \tau) = (2A_0)^{1/2} \exp \frac{\tilde{\theta}^1(\infty, \tau)}{2}. \quad (51)$$

Thus (49) and (51) imply that

$$[\tilde{\theta}_\eta^1] = -(2A_0)^{1/2} \exp \frac{\tilde{\theta}^1(\infty, \tau)}{2} \quad (52)$$

or equivalently that

$$[\theta_\xi^0] = -(2A_0)^{1/2} \exp \frac{\theta^1(0^+, \tau)}{2} \quad (53)$$

where $[f] \equiv f(0^+) - f(0^-)$ denotes the jump of f across the front $\xi = 0$. To calculate A_0 , we note from the definition of A in (25) that it is a function of the normal velocity V_n only. Therefore A_0 depends only on the solution of the problem of plane propagation which is given by

$$\begin{aligned} \theta^p &= e^\xi & \text{for } \xi < 0 \\ &= 1 & \text{for } \xi > 0. \end{aligned} \quad (54)$$

Therefore, in this problem

$$[\theta_\xi^0] = -1 = -(2A_0)^{1/2} \quad (55)$$

so that

$$(2A_0)^{1/2} = 1. \quad (56)$$

Then Eq. (53) becomes

$$[\theta_\xi^0] + \exp \frac{\theta^1(0^+, \tau)}{2} = 0. \quad (57)$$

By comparing with (32), which implies that

$$[\theta_\xi^0] + Q_0 = 0, \quad (58)$$

we see that

$$Q_0 = \exp \frac{\theta^1(0^+, \tau)}{2}. \quad (59)$$

The equations (32)–(33) for θ^0 and n^0 are not a closed system since Q_0 depends on θ^1 . Therefore we consider the next terms in the outer expansion, θ_\pm^1 and n_\pm^1 , which satisfy

$$\frac{\partial \theta^1}{\partial \tau} - (\Phi_0)_\tau \frac{\partial \theta^1}{\partial \xi} - (\Phi_1)_\tau \frac{\partial \theta^0}{\partial \xi} = \frac{\partial^2 \theta^1}{\partial \xi^2} + Q_1 \delta(\xi), \quad (60)$$

$$\frac{\partial n^1}{\partial \tau} - (\Phi_0)_\tau \frac{\partial n^1}{\partial \xi} - (\Phi_1)_\tau \frac{\partial n^0}{\partial \xi} = \frac{\partial^2 n^1}{\partial \xi^2} + \nu \frac{\partial^2 n^0}{\partial \xi^2} + Q_1 \delta(\xi). \quad (61)$$

In a similar manner, we can show that Q_1 depends on θ^2 so that the system of equations does not seem to close. However if we subtract Eq. (61) from (60), we see that the quantity

$$s \equiv \theta^1 - n^1 \quad (62)$$

satisfies

$$\frac{\partial s}{\partial \tau} - (\Phi_0)_\tau \frac{\partial s}{\partial \xi} = \frac{\partial^2 s}{\partial \xi^2} - \nu \frac{\partial^2 n^0}{\partial \xi^2}. \quad (63)$$

We also note that

$$n^1(0^+, \tau) = 0 \quad (64)$$

which implies that

$$s(0^+, \tau) = \theta^1(0^+, \tau). \quad (65)$$

We have thus succeeded in reducing the problem to the solution of the two equations for n^0 and s , given by

$$\frac{\partial n^0}{\partial \tau} - (\Phi_0)_\tau \frac{\partial n^0}{\partial \xi} = \frac{\partial^2 n^0}{\partial \xi^2} + \exp \frac{s(0^+, \tau)}{2} \delta(\xi), \quad (66)$$

$$\frac{\partial s}{\partial \tau} - (\Phi_0)_\tau \frac{\partial s}{\partial \xi} = \frac{\partial^2 s}{\partial \xi^2} - \nu \frac{\partial^2 n^0}{\partial \xi^2} \quad (67)$$

subject to the boundary conditions

$$\begin{aligned} n^0(-\infty, \tau) &= 0, & n^0(\infty, \tau) &= 1, \\ s(-\infty, \tau) &= 0, & s(\infty, \tau) &< \infty. \end{aligned} \quad (68)$$

4. The basic solution and its stability. The basic solution of problem (66)–(68), describing normal, time-independent necking, is given by

$$\begin{aligned} n^0 &= n^b \equiv \exp \xi, \text{ for } \xi < 0 \\ &\equiv 1, \text{ for } \xi > 0, \\ s &= s^b \equiv \xi \exp \xi, \text{ for } \xi < 0 \\ &\equiv 0, \text{ for } \xi > 0, \\ \phi_\tau^0 &= -1. \end{aligned} \quad (69)$$

For a linear analysis of the stability of this solution, we introduce perturbations $u = s - s^b$, $v = n^0 - n^b$, $\phi_\tau = \Phi_\tau - \Phi_\tau^0$, and linearize Eqs. (66), (68) to obtain

$$\frac{\partial v}{\partial \tau} - \phi_\tau \frac{dn^b}{d\xi} + \frac{\partial v}{\partial \xi} = \frac{\partial^2 v}{\partial \xi^2} + \frac{1}{2} u \delta(\xi), \quad (70)$$

$$\frac{\partial u}{\partial \tau} - \phi_\tau \frac{ds^b}{d\xi} + \frac{\partial u}{\partial \xi} = \frac{\partial^2 u}{\partial \xi^2} - \nu \frac{\partial^2 v}{\partial \xi^2}, \quad (71)$$

$$v(\xi, \tau) = 0 \text{ for } \xi \leq 0, \quad (72)$$

$$u(-\infty, \tau) = v(-\infty, \tau) = 0, \quad u(+\infty, \tau) < \infty. \quad (73)$$

We now show that there is a solution of the linear problem (70)–(73) of the form

$$\begin{aligned} \phi(\tau) &= D \exp(i\omega\tau), & u(\tau, \xi) &= D \exp(i\omega\tau) U(\xi), \\ v(\tau, \xi) &= D \exp(i\omega\tau) V(\xi). \end{aligned} \quad (74)$$

Then we show that there are parameter values ν for which the imaginary part of at least one such ω is negative, which corresponds to unstable normal necking.

The problem is thus reduced to the solution of

$$i\omega V - i\omega \frac{dn^b}{d\xi} + \frac{dV}{d\xi} = \frac{d^2V}{d\xi^2} + \frac{1}{2} U\delta(\xi), \quad (75)$$

$$i\omega U - i\omega \frac{ds^b}{d\xi} + \frac{dU}{d\xi} = \frac{d^2U}{d\xi^2} - \nu \frac{d^2V}{d\xi^2} \quad (76)$$

subject to the boundary conditions

$$U(\pm\infty) = V(\pm\infty) = 0. \quad (77)$$

Eqs. (75)–(77) imply that

$$\beta((1 + 4i\omega)^{1/2} - 1) = 1 + 4i\omega, \quad (78)$$

or

$$(i\omega)^2 + (2 + 2\beta - \beta^2)(i\omega) + 1 + 2\beta = 0, \quad (79)$$

where $\beta \equiv \nu/4$. Hence $\text{Re}(i\omega) > 0$ ($\text{Im}\omega < 0$) when $\beta > 1 + \sqrt{3} \equiv \beta_c$ or $\beta < -\frac{1}{2}$. The root $\beta = -\frac{1}{2}$ of Eq. (79) is not a root of the original equation (78). Thus, normal necking is unstable to one-dimensional disturbances if

$$N(L - 1) > 4(1 + \sqrt{3}) \simeq 10.9. \quad (80)$$

Since $N = \gamma(\sigma_\infty(1 - \alpha))/kT$ it follows that by lowering the temperature while leaving all other parameters unchanged, we can move into an instability region.

We note that the critical case ($\beta = \beta_c$) may be achieved at quite realistic values of the physical parameters. For example, at $N = 30$ Eq. (80) gives $L = 1.36$. At $\beta = \beta_c$, the real frequency $\omega = \omega_c$ is given by

$$\omega_c = +\frac{1}{4}(3 + 2\sqrt{3})^{1/2} \simeq 0.64. \quad (81)$$

For $\beta > \beta_c$, the system undergoes a Hopf-type bifurcation, in which the normal necking process becomes unstable and *oscillatory* necking sets in. To determine the bifurcation solution quantitatively, one may employ an analysis similar to that in Matkowsky and Sivashinsky [13].

REFERENCES

- [1] W. H. Carothers and J. W. Hill, *Studies of polymerization and ring formation. XV. Artificial fiber from synthetic linear condensation superpolymers*, J. Amer. Chem. Soc. **54**, N4 (1932)
- [2] A. Nadai, *Theory of flow and fractures of solids*, McGraw-Hill N. Y., Toronto, London (1952)
- [3] G. I. Barenblatt, *On the extension of neck of polymer specimens under tension*, Appl. Math. and Mech. (PMM) **28**, 1264-1276 (1964)
- [4] G. I. Barenblatt, *Isothermal propagation of a neck in polymers*, Mechanics of Solids (MMT) **N6**, 89-96 (1972)
- [5] Y. B. Zeldovich, *Theory of flame propagation*, Zh. Fiz. Khim. **22**, no. 1 (1948) (in Russian)
- [6] Yu. S. Lazurkin, *Cold-drawing of glass-like and crystalline polymers*, J. Polymer Sci. **30**, no. 121 (1958)
- [7] P. J. Vincent, *The necking and cold drawing of rigid plastics*, Polymer **1**, no. 1 (1960)
- [8] G. P. Andrianova, V. A. Kargin and A. S. Kechekyan, *Periodic oscillations in extension of polyethylene terephthalate*, J. Macromolecular Compounds (Vysokomolekulyarnye Soedineniya) **12**, no. 8, 1970 (in Russian)
- [9] Ya. B. Zel'dovich and G. I. Barenblatt, *Theory of flame propagation*, Combustion and Flame **3**, 61-74 (1959)
- [10] G. I. Barenblatt, *Self-oscillatory propagation of a neck in polymers*, Mechanics of Solids (MMT) **N5**, 110-118 (1970)
- [11] G. I. Sivashinsky, *The structure of Bunsen flames*, J. Chem. Physics, **62**, 638-643 (1975)
- [12] G. I. Sivashinsky, *The diffusion stratification effect in Bunsen flames*, J. Heat Transfer, ASME Transactions **N11**, 530-535 (1974)
- [13] B. J. Matkowsky and G. I. Sivashinsky, *Propagation of a pulsating reaction front in solid fuel combustion*, SIAM J. Applied Math. **35**, 465-478 (1978)
- [14] B. J. Matkowsky and G. I. Sivashinsky, *A nonlinear theory of cellular flames*, to appear