

Strain-Coupling Effects in Extensional Flows

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SYNOPSIS

A recently developed strain-coupling constitutive equation for nonlinear viscoelasticity is used to analyze extensional flows. Comparisons are presented between the predictions of the theory and experiment for equibiaxial- and uniaxial-step strain-stress relaxation experiments. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

A strain-coupling constitutive equation has recently been proposed¹⁻⁵ as a possible improvement to the K-BKZ constitutive equation. Furthermore, it has been shown that the strain-coupling constitutive model provides satisfactory descriptions of strain and time variations for double-step shear strain-stress relaxation experiments for both linear⁶ and branched⁷ polymers. The objective of this study is to extend the application of the strain-coupling constitutive equation to the analysis of nonlinear viscoelasticity in extensional flows. The generalization of the strain-coupling model to the analysis of extensional flows is described in the second section of this article, and predictions for uniaxial and equibiaxial extensional flows are presented in the third section. Comparison of the predictions of the theory with available extensional flow data is considered in the fourth section of this article.

GENERALIZATION OF STRAIN-COUPLING MODEL

The strain-coupling model is described by the following equation for the extra stress \mathbf{S} :

$$\begin{aligned} \mathbf{S} = & \int_0^\infty [\phi_1(s, I, II) \\ & + \int_0^\infty \phi_3\{s_1, s, I(s_1)\} ds_1][\mathbf{N}(s) - \mathbf{I}] ds \\ & + \int_0^\infty [\phi_2(s, I, II)][\mathbf{N}^{-1}(s) - \mathbf{I}] ds \quad (1) \end{aligned}$$

where

$$\phi_3(s_1, s, 0) = 0 \quad (2)$$

$$I = \text{tr}[\mathbf{N} - \mathbf{I}] \quad (3)$$

$$\begin{aligned} II = & \frac{1}{2}[I^2 - \text{tr}(\mathbf{N} - \mathbf{I})^2] \\ = & \text{tr}[\mathbf{N}^{-1} - \mathbf{I}] - 2 \text{tr}[\mathbf{N} - \mathbf{I}] \quad (4) \end{aligned}$$

$$\mathbf{N}(s) = \mathbf{C}_t^{-1}(t - s) \quad (5)$$

$$\mathbf{N}^{-1}(s) = \mathbf{C}_t(t - s) \quad (6)$$

In these equations, t is the present time; s , the backward running time; $\mathbf{C}_t(t - s)$, the right Cauchy-Green tensor relative to time t ; and \mathbf{I} , the identity or unit tensor. The strain-coupling constitutive equation, which has three scalar-valued material functions, ϕ_1 , ϕ_2 , and ϕ_3 , reduces to the K-BKZ constitutive equation when $\phi_3 = 0$. For the strain-coupling model, the influence of each strain increment on the stress is dependent on other strain increments. For the K-BKZ model, such coupling of strains is assumed to be negligible.

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For single-step shear strain-stress relaxation experiments, it is often possible to write the shear stress for viscoelastic fluids in the following factored form:

$$\sigma(\gamma_1, t) = \gamma_1 G(t) h(\gamma_1^2) \quad (7)$$

Here, γ_1 is the instantaneous shear strain applied at $t = 0$; $\sigma(\gamma_1, t)$, the resulting shear stress for $t > 0$; $G(t)$, the shear stress relaxation modulus of linear viscoelasticity; and $h(\gamma_1^2)$, a monotonically decreasing function of strain with $h(0) = 1$. For many materials, time-strain factorability is valid at least for low values of γ_1 , whereas for some materials, the factored form given by eq. (7) is valid for a wide range of γ_1 . Time-strain separability will occur for the strain-coupling model if

$$\phi_1(s, I, II) - \phi_2(s, I, II) = m(s)H(I, II) \quad (8)$$

$$\begin{aligned} \phi_1(s, I, -I) - \phi_2(s, I, -I) \\ = m(s)H(I, -I) = m(s)H^*(I) \end{aligned} \quad (9)$$

$$m(s) = -\frac{dG(s)}{ds} \quad (10)$$

$$\phi_3\{s_1, s, I(s_1)\} = \beta(s_1, s)K[I(s_1)] \quad (11)$$

For simple shear deformations, $II = -I$. In addition, analysis of a single-step shear strain experiment leads to the result

$$h(I) = H^*(I) + K(I) \quad (12)$$

Also, it has been shown elsewhere⁷ that $\beta(s_1, s)$ and $K(I)$ can be evaluated using the following expressions:

$$\beta(s, s_1) = \frac{9}{1-k} \sum_{i=1}^N \frac{a_i}{\lambda_i} e^{8s/\lambda_i} e^{-9s_1/\lambda_i} s_1 > s \quad (13)$$

$$\beta(s, s_1) = -\frac{9k}{1-k} \sum_{i=1}^N \frac{a_i}{\lambda_i} e^{-9s/\lambda_i} e^{8s_1/\lambda_i} s > s_1 \quad (14)$$

$$\frac{K(I)}{8(1-k)} = \frac{h(4I) - h(I)}{2} \quad (15)$$

Here, k is a constant, and a_i and λ_i are constants in a commonly used expression for $m(t)$:

$$m(t) = \sum_{i=1}^N a_i e^{-t/\lambda_i} \quad (16)$$

The following equation can be used to evaluate the constant k ⁷:

$$k = 1 + \frac{2\Delta\sigma(\gamma, 0, t, t_1)}{\gamma[G(t) - G(t + 8t_1)][h(\gamma^2) - h(4\gamma^2)]} \quad (17)$$

This equation is based on the utilization of a double-step shear strain-stress relaxation experiment with an instantaneous strain γ_1 introduced at $t = 0$ and a second total strain γ_2 introduced at $t = t_1$. The quantity $\Delta\sigma(\gamma_1, \gamma_2, t, t_1)$ represents the difference between the shear stress $\sigma(\gamma_1, \gamma_2, t, t_1)$ measured for $t > t_1$ and the prediction of the K-BKZ theory for the double-step experiment. From eq. (17), it is clear that a *single* data point (a particular value of t) from a *single* double-step experiment with $\gamma_1 = \gamma$, $\gamma_2 = 0$, and fixed t_1 is needed for the evaluation of k . Furthermore, it has been shown previously⁷ that k can be estimated without using double-step data. The following equations provide a reasonable estimate for k :

$$k = \frac{2[-1-\xi]}{3[1-\xi]} \quad (18)$$

$$\xi = \frac{\frac{K(9\gamma^2)}{8(1-k)}}{\frac{K(\gamma^2)}{8(1-k)}} \quad (19)$$

These equations appear to be valid at moderate strain levels. For example, it has been shown that a choice of $\gamma^2 = \frac{1}{9}$ yields good estimates for k for two polymeric systems.⁷

It was shown previously⁷ that data taken from an appropriate series of single-step shear strain-stress relaxation experiments and from, at most, a single double-step shear strain experiment can be used to evaluate $G(t)$, $H^*(I)$, $\beta(s_1, s)$, and $K(I)$. Consequently, ϕ_3 and $\phi_1 - \phi_2$ (for cases for which $II = -I$) can be determined, and the more important aspects of simple shear deformations can be analyzed. However, more experimental data and/or additional constitutive hypotheses are needed if more general deformations, like extensional flows, are to be analyzed. In this paper, we introduce two additional constitutive assumptions and show that only one additional experimental fact is needed to achieve a complete description of the nonlinear viscoelastic behavior of the material. We introduce the following two constitutive hypotheses:

1. The functions $\phi_1(s, I, II)$ and $\phi_2(s, I, II)$ are related as follows:

$$\phi_2 = -\epsilon\phi_1 \quad (20)$$

where ϵ is a constant that can be determined using an appropriate experiment.

2. The two functions ϕ_1 and ϕ_2 can be derived from a potential W .

The first constitutive hypothesis is suggested by the relatively modest shear rate dependence of $-N_2/N_1$,^{1,8} where N_1 and N_2 are the first and second normal stress differences for steady shear flows. For steady shear flows with shear rate $\dot{\gamma}$, the deformation field is described by the following equations:

$$[N(s) - I] = \begin{bmatrix} \dot{\gamma}^2 s^2 & \dot{\gamma} s & 0 \\ \dot{\gamma} s & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (21)$$

$$[N^{-1}(s) - I] = \begin{bmatrix} 0 & -\dot{\gamma} s & 0 \\ -\dot{\gamma} s & \dot{\gamma}^2 s^2 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (22)$$

$$I = -II = \dot{\gamma}^2 s^2 \quad (23)$$

Consequently, for the strain-coupling model, it can be shown using eqs. (1) and (20) that the ratio of normal stress differences is given by the following expression:

$$-\frac{N_2(\dot{\gamma})}{N_1(\dot{\gamma})} = \frac{\epsilon}{1 + \epsilon + Q} \quad (24)$$

$$Q = \frac{\int_0^\infty \int_0^\infty \beta(s_1, s) K(\dot{\gamma}^2 s_1^2) s^2 ds_1 ds}{\int_0^\infty \phi_1(s, \dot{\gamma}^2 s^2, -\dot{\gamma}^2 s^2) s^2 ds} \quad (25)$$

In the limit as $\dot{\gamma} \rightarrow 0$, $Q = 0$, and eq. (24) reduces to the following result:

$$\left[-\frac{N_2(\dot{\gamma})}{N_1(\dot{\gamma})} \right]_{\dot{\gamma}=0} = \frac{\epsilon}{1 + \epsilon} \quad (26)$$

Thus, normal stress data taken using steady shear flow experiments,^{1,8} can be used to determine the constant ϵ . Furthermore, a shear rate dependence for $-N_2(\dot{\gamma})/N_1(\dot{\gamma})$ is possible because of the presence of Q , the strain-coupling contribution to the normal stress ratio. The utilization of a constant value for ϵ is thus reasonable since it does not pre-

clude the possibility of a shear rate dependence for the normal stress ratio.

The second constitutive hypothesis is suggested by the fact that the expression containing the contribution to the stress of the uncoupled strains in a viscoelastic fluid can be regarded as a generalization of the stress expression for an incompressible elastic body. Indeed, Larson⁹ has shown how the K-BKZ equation can be derived by generalizing the theory of rubber elasticity by using a history integral for the potential. It seems reasonable to suppose that only the part of the strain-coupling constitutive model that involves uncoupled strains should be derived from a potential W since the strains for elastic bodies are, of course, not coupled. Hence, if we follow the development of Larson, the following expressions are valid for ϕ_1 and ϕ_2 in terms of the potential

$W(s, \hat{I}, \hat{II})$:

$$\phi_1 = 2 \frac{\partial W}{\partial \hat{I}} \quad (27)$$

$$\phi_2 = -2 \frac{\partial W}{\partial \hat{II}} \quad (28)$$

$$\hat{I} = \text{tr } N \quad (29)$$

$$\hat{II} = \frac{1}{2} [\hat{I}^2 - \text{tr } N^2] \quad (30)$$

Combination of eqs. (20), (27), and (28) produces the following first-order partial differential equation for W :

$$\frac{\partial W}{\partial \hat{II}} = \epsilon \frac{\partial W}{\partial \hat{I}} \quad (31)$$

The solution to this equation takes the form

$$W = W(I^*) \quad (32)$$

$$I^* = \frac{\hat{I} + \epsilon \hat{II} - 3(1 + \epsilon)}{1 + \epsilon} \quad (33)$$

Since

$$\hat{I} = I + 3$$

$$\hat{II} = 2I + II + 3 \quad (34)$$

the generalized invariant I^* can be written as follows:

$$I^* = \frac{I(1 + 2\epsilon)}{1 + \epsilon} + \frac{\epsilon II}{1 + \epsilon} \quad (35)$$

Clearly, $I^* = 0$ when $I = II = 0$. Also, $I^* = I$ when $II = -I$ (as in simple shear flows). Time-strain factorability will occur if the potential function $W(s, \hat{I}, \hat{II})$ is factorable:

$$W(s, \hat{I}, \hat{II}) = m(s) \hat{W}(\hat{I}, \hat{II}) \quad (36)$$

It is evident, then, from eqs. (8), (9), (27), and (28) that

$$\phi_1(s, I, II) = \frac{m(s)H^*(I^*)}{1 + \epsilon} \quad (37)$$

$$\phi_2(s, I, II) = -\frac{m(s)\epsilon H^*(I^*)}{1 + \epsilon} \quad (38)$$

The above development and previous work^{6,7} suggest the following procedure for the complete determination of the material functions of the strain-coupling theory, ϕ_1 , ϕ_2 , and ϕ_3 , for materials for which time-strain factorability is applicable:

1. A series of single-step shear strain experiments is conducted over an appropriate range of γ_1 . These data can be used to determine $G(t)$ and $h(I)$ using eq. (7). The parameters a_i and λ_i can be determined from standard procedures using the $G(t)$ data and eqs. (10) and (16).
2. The function $K(I)/(1 - k)$ can be determined over an appropriate range of I using $h(I)$ data in eq. (15).
3. A *single* data point from a *single* double-step shear strain experiment with $\gamma_2 = 0$ and an appropriate value of $\gamma_1 = \gamma$ can be used to determine k from eq. (17). In the absence of double-step data, eq. (18) can be used to estimate k . The function $K(I)$ can then be determined over the complete range of I for which $K(I)/(1 - k)$ values are available.
4. It is then possible to determine $\beta(s, s_1)$ using eqs. (13) and (14), and, thus, ϕ_3 can be computed using the known $K(I)$ and $\beta(s, s_1)$ from eq. (11).
5. The quantity $H^*(I)$ and, hence, $H^*(I^*)$ can be calculated using eq. (12).
6. Steady shear experiments over an appropriate range of $\dot{\gamma}$ can be used to determine N_2/N_1 near $\dot{\gamma} = 0$. This ratio can be calculated using data taken on cone-and-plate and parallel-plate viscometers.^{1,8} The constant ϵ can

be calculated from eq. (26) using the value of N_2/N_1 at the zero shear rate limit.

7. The quantities $\phi_1(s, I, II)$ and $\phi_2(s, I, II)$ can be calculated using eqs. (37) and (38) with I^* defined by eq. (35).

The above procedure requires only data taken in simple shear deformations, using both step strain and steady shear histories. Only one double-step experiment, at most, is needed, and the steady shear data are required only near $\dot{\gamma} = 0$.

ANALYSIS OF EXTENSIONAL FLOWS

Extensional flows can be conveniently characterized using extension ratios. In a uniaxial single-step strain-stress relaxation experiment, the material is stretched in a ratio λ_1 in the x direction at $t = 0$ and contracts in a ratio $(\lambda_1)^{-1/2}$ in the y and z directions. For this uniaxial-step strain experiment, the deformation tensors for $t > 0$, $s > t$ are simply

$$[\mathbf{N}(s) - \mathbf{I}] = \begin{bmatrix} \lambda_1^2 - 1 & 0 & 0 \\ 0 & \frac{1}{\lambda_1} - 1 & 0 \\ 0 & 0 & \frac{1}{\lambda_1} - 1 \end{bmatrix} \quad (39)$$

$$[\mathbf{N}^{-1}(s) - \mathbf{I}] = \begin{bmatrix} \frac{1}{\lambda_1^2} - 1 & 0 & 0 \\ 0 & \lambda_1 - 1 & 0 \\ 0 & 0 & \lambda_1 - 1 \end{bmatrix} \quad (40)$$

with the invariants given by the following expressions:

$$I = \lambda_1^2 + \frac{2}{\lambda_1} - 3 \quad (41)$$

$$II = \frac{1}{\lambda_1^2} + 2\lambda_1 - 2\lambda_1^2 - \frac{4}{\lambda_1} + 3 \quad (42)$$

For $t > 0$ and $s < t$, both deformation tensors and both invariants are identically zero. Hence, the strain-coupling model [eq. (1)] yields the following expression for the normal stress difference:

$$\frac{S_{xx} - S_{yy}}{\lambda_1^2 - \lambda_1^{-1}} = \int_t^\infty \left[\phi_1(s, I, II) - \frac{\phi_2(s, I, II)}{\lambda_1} \right] ds + \int_t^\infty \int_t^\infty \phi_3(s_1, s, I) ds_1 ds \quad (43)$$

This equation can be simplified by substituting eqns. (11), (37), and (38), by using the results^{6,7}

$$\int_t^\infty m(s) ds = G(t) \quad (44)$$

$$\int_t^\infty \int_t^\infty \beta(s_1, s) ds_1 ds = G(t) \quad (45)$$

and by defining a damping function h_U for uniaxial extension:

$$\frac{S_{xx} - S_{yy}}{\lambda_1^2 - \lambda_1^{-1}} = h_U(I^*)G(t) \quad (46)$$

When all these equations are used in eq. (43), the following expression can be derived:

$$h_U(I^*) = H^*(I^*) \left[\frac{\lambda_1 + \varepsilon}{\lambda_1(1 + \varepsilon)} \right] + K(I) \quad (47)$$

Finally, utilization of eq. (12) produces the following result:

$$h_U(I^*) = q_1 [h(I^*) - K(I^*)] + K(I) \quad (48)$$

$$q_1 = \frac{\lambda_1 + \varepsilon}{\lambda_1(1 + \varepsilon)} \quad (49)$$

Equation (48) provides a simple relationship between the uniaxial damping function, h_U , the shear damping function, h , and the strain-coupling function $K(I)$. Clearly, an extensional flow prediction can be made based on material functions derived using only data from simple shear flows.

For an equibiaxial single-step strain-stress relation experiment, the material is stretched in a ratio λ_1 in the x and y directions at $t = 0$ and contracts in a ratio λ_1^{-2} in the z direction. For this equibiaxial-step strain experiment, the deformation tensors for $t > 0$, $s > t$ are as follows:

$$[\mathbf{N}(s) - \mathbf{I}] = \begin{bmatrix} \lambda_1^2 - 1 & 0 & 0 \\ 0 & \lambda_1^2 - 1 & 0 \\ 0 & 0 & \frac{1}{\lambda_1^4} - 1 \end{bmatrix} \quad (50)$$

$$[\mathbf{N}^{-1}(s) - \mathbf{I}] = \begin{bmatrix} \frac{1}{\lambda_1^2} - 1 & 0 & 0 \\ 0 & \frac{1}{\lambda_1^2} - 1 & 0 \\ 0 & 0 & \lambda_1^4 - 1 \end{bmatrix} \quad (51)$$

and the invariants can be expressed in the following forms:

$$I = 2\lambda_1^2 + \frac{1}{\lambda_1^4} - 3 \quad (52)$$

$$II = \frac{2}{\lambda_1^2} - 4\lambda_1^2 + \lambda_1^4 - \frac{2}{\lambda_1^4} + 3 \quad (53)$$

Again, for $t > 0$, $s < t$, both deformation tensors and both invariants are identically zero. The strain-coupling constitutive equation [eq. (1)] produces the following result for the normal stress difference:

$$\frac{S_{xx} - S_{zz}}{\lambda_1^2 - \lambda_1^{-4}} = \int_t^\infty [\phi_1(s, I, II) - \lambda_1^2 \phi_2(s, I, II)] ds + \int_t^\infty \int_t^\infty \phi_3(s_1, s, I) ds_1 ds \quad (54)$$

If we now define a biaxial damping function using the equation

$$\frac{S_{xx} - S_{zz}}{\lambda_1^2 - \lambda_1^{-4}} = h_B(I^*)G(t) \quad (55)$$

and proceed in a manner similar to the procedure used for the derivation of the result for a uniaxial extension, we deduce the following expression:

$$h_B(I^*) = q_2 [h(I^*) - K(I^*)] + K(I) \quad (56)$$

$$q_2 = \frac{1 + \varepsilon \lambda_1^2}{1 + \varepsilon} \quad (57)$$

The form of eq. (56) is, of course, very similar to the form of eq. (48) since the equibiaxial extension can be regarded as a special type of uniaxial experiment with a compression in the stretching direction.

RESULTS AND DISCUSSION

Extensional flow predictions are presented here for two polymers: The IUPAC branched low-density polyethylene sample and a commercial low-density polyethylene sample similar to the IUPAC sample.¹⁰ Values of $K(I)/8(1 - k)$ for the IUPAC sample are presented in Figure 1.⁷ The curve in this figure was computed using an average of the shear damping function, $h(I)$, for the IUPAC A (Ref. 11) and IUPAC \times (Ref. 12) samples. A value of $k = 1.5$ was reported elsewhere⁷ for the IUPAC sample. The strain dependence of $K(I)/8(1 - k)$ for the low-

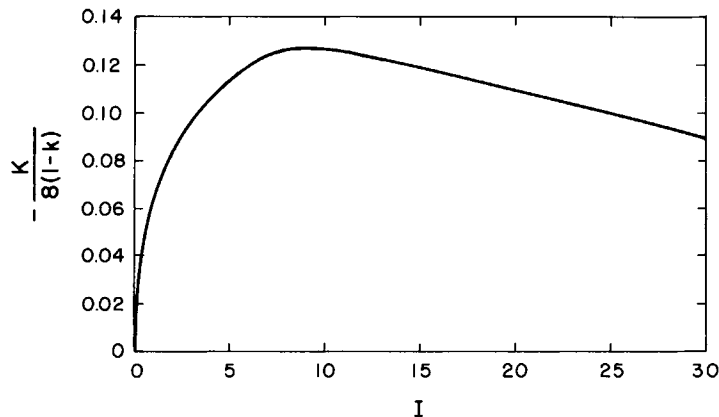


Figure 1 Strain dependence of $K(I)/8(1 - k)$ for IUPAC sample for temperature range 125–150°C.

density polyethylene sample used by Khan and Tanner¹⁰ is presented in Figure 2.⁷ Since there are some differences between the calculated shear damping function and the experimental damping function data, all calculations in this study were based on $h(I)$ values between the experimental and calculated values. No value of k could be calculated for this sample using eq. (17) because double-step data with $\gamma_2 = 0$ were not available. Consequently, a value of $k = 1.14$ was estimated for this commercial polyethylene sample using eqs. (18) and (19) with $\gamma^2 = \frac{1}{9}$. Finally, a value of $N_2/N_1 = -0.22$ has been reported¹³ for low-density polyethylene, and this leads to a value of $\epsilon = 0.28$, which was used for both of the above polyethylene samples.

A comparison of the predictions of the strain-coupling theory for equibiaxial step strain-stress relaxation experiments with data¹¹ for the low-

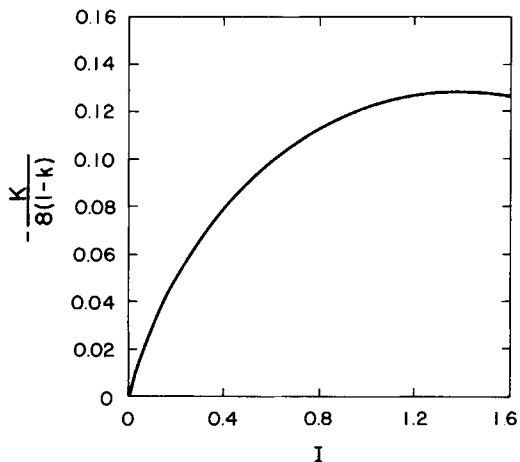


Figure 2 Strain dependence of $K(I)/8(1 - k)$ for the commercial polyethylene sample¹⁰ at 130°C.

density polyethylene IUPAC sample is presented in Figure 3. From this figure, it is evident that there is good agreement between the predicted and experimental values of h_B , the biaxial damping function. In addition, a comparison of the predictions of the strain-coupling theory for uniaxial-step strain-stress relaxation experiments with data¹⁰ for the commercial low-density polyethylene sample is presented in Figure 4. In this case, there is reasonably good agreement between the predicted and experimental values of h_U , the uniaxial damping function,

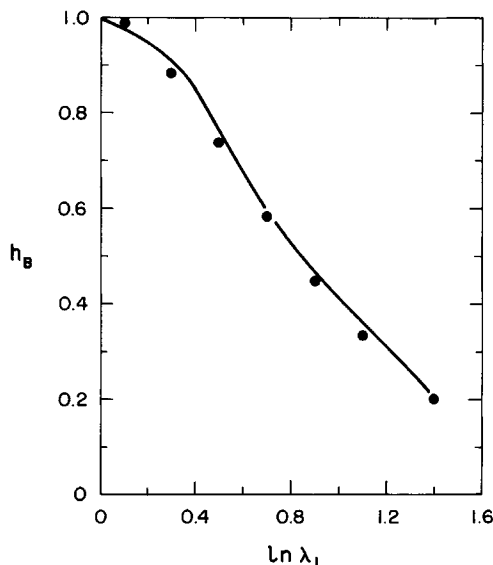


Figure 3 Comparison of theory and experiment for equibiaxial-step strain-stress relaxation. The curve is the prediction of the strain-coupling theory for the damping function, and the solid circles represent experimental data.¹¹

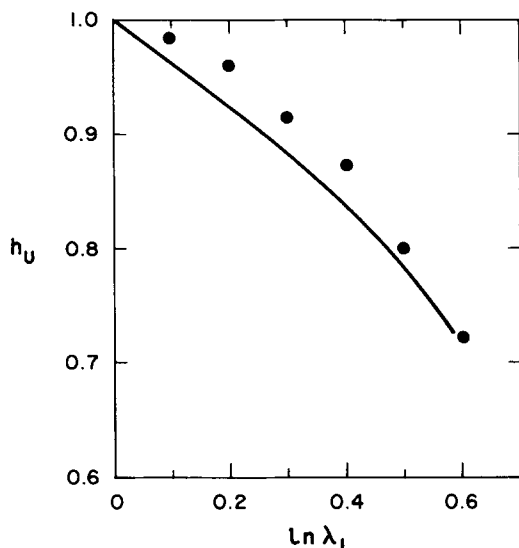


Figure 4 Comparison of theory and experiment for uniaxial-step strain-stress relaxation. The curve is the prediction of the strain-coupling theory for the damping function, and the solid circles represent experimental data.¹⁰

particularly when consideration is taken off the fact that there is some uncertainty in the shear damping function for this polymer melt. Finally, the predicted dependence of h_B and h_U on I for the IUPAC sample is presented in Figure 5 along with experimental data for the shear damping function h . At low values of

I , $h_B > h > h_U$, and this prediction appears to be in agreement with experimental data presented by Larson⁹ for the IUPAC sample. At higher values of I , $h > h_B$, and again this agrees with the data presented by Larson. Also, at higher values of I , h and h_U are very close together, whereas Larson shows that h_U is significantly larger than h for sufficiently high strains. However, it must be remembered that the value of h_U presented by Larson is calculated using data from a stress growth experiment, and there is no reason to expect that such an h_U is the same as the true h_U predicted for an actual uniaxial-step strain-stress relaxation experiment.

It appears that the strain-coupling theory is capable of predicting extensional flow data for low-density polyethylene samples using only data collected in shear deformations. It is important, of course, to keep the apparent success of this model in perspective since data for only one material have been analyzed. However, it is fair to conclude that the present form of the strain-coupling model might well provide a reasonably good method of describing both shear and extensional deformations in polymeric materials.

It is important to emphasize that the comparisons between data and results from the strain-coupling theory presented here for extensional flows are based on *predictions* of the extensional flow results since no extensional flow data are utilized. On the other hand, the usual comparisons of data and results from

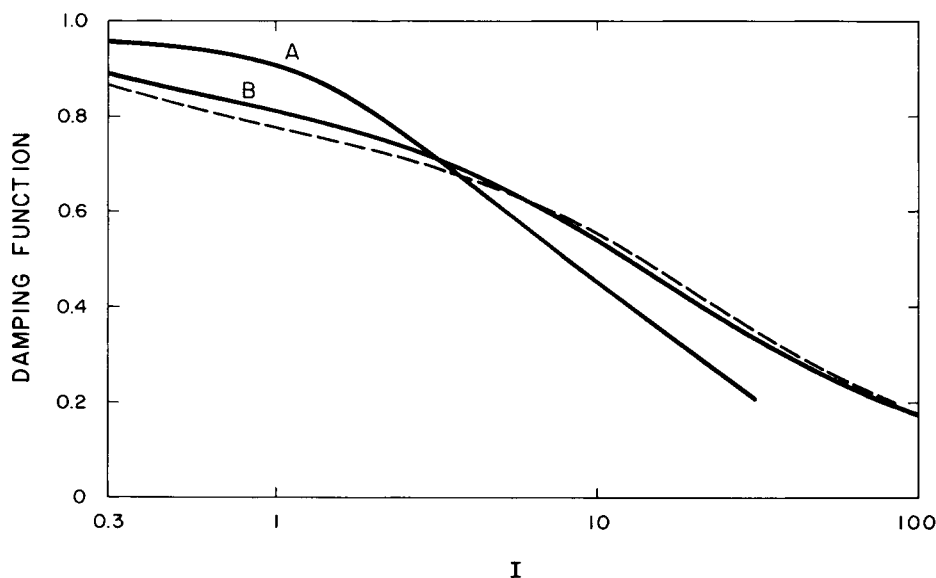


Figure 5 Dependence of damping functions on I . Curve A is the strain-coupling prediction for the biaxial damping function, h_B , and the dashed curve is the strain-coupling prediction for the uniaxial damping function, h_U . Curve B represents experimental data for the shear damping function, h .

the K-BKZ theory for extensional flows are based on *correlations* of the extensional flow results. A generalized invariant is usually proposed and constants in the expression for the invariant are determined using extensional flow data.^{9,10} Such an approach does not provide a meaningful evaluation of the predictive capabilities of the K-BKZ theory. However, planar extensional flows can be used to check directly the applicability of the K-BKZ theory to extensional flows. It has been shown that the K-BKZ theory provides an inadequate prediction for planar extension.¹² Furthermore, if strain-coupling effects are excluded in our theory, the predictions for equibiaxial extension will be poor.

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