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Analysis of Steady State Shearing and Stress Relaxation in the Maxwell Orthogonal Rheometer

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A nonlinear, integral viscoelastic model is used to predict the stresses in the Maxwell orthogonal rheometer. The resulting expressions indicate that the instrument yields data on material functions not heretofore measured, and also show how the data may be analyzed to get the complex viscosity. Expressions are given for stress relaxation after cessation of shearing. Some experimental data are analyzed to give model parameters.

The Maxwell orthogonal rheometer (*1*) is an interesting rheological instrument, because the flow is nonviscometric. Recently an analysis was given (*2*) for the steady state operation of this device. Because it is based on a rheological model which is inadequate, the analysis does not account for possible deviations from linear viscoelasticity. The analysis is repeated here using a model which is felt to be more appropriate; and, in addition, expressions describing stress relaxation (*3*) are given. It is hoped that these analyses will help to call attention to the capabilities of the Maxwell orthogonal rheometer as a rheological tool.

THE RHEOLOGICAL MODEL

We use here an integral model of the form (*4*):

$$\tau = -\int_{-\infty}^{t} m(t-t', II(t')) \left[\left(1+\frac{\epsilon}{2}\right)\overline{\mathbf{\Gamma}} - \left(\frac{\epsilon}{2}\right)\mathbf{\Gamma} \right] dt' \quad (1)$$

Here $\overline{\mathbf{\Gamma}}$ and $\mathbf{\Gamma}$ are strain tensors defined, in cartesian coordinates, by

$$\overline{\Gamma_{ij}} = \Sigma_m (\partial x_i/\partial x'_m)(\partial x_j/\partial x'_m) - \delta_{ij} \quad (2)$$

$$\Gamma_{ij} = \delta_{ij} - \Sigma_m (\partial x'_m/\partial x_i)(\partial x'_m/\partial x_j) \quad (3)$$

in which x_i and x_i' are the position coordinates at times t (current time) and t' (past time) respectively. The memory function $m(t-t', II(t'))$ is taken to be

$$m(t-t', II(t')) = \sum_{p=1}^{\infty} \frac{\eta_p}{\lambda_{2p}^2} \frac{e^{-(t-t')/\lambda_{2p}}}{1+\frac{1}{2}\lambda_{1p}^2 II(t')} \quad (4)$$

The quantity $II(t')$ is $(\dot{\boldsymbol{\gamma}}:\dot{\boldsymbol{\gamma}}) = \Sigma_i\Sigma_j\ \gamma_{ij}^2$ where $\gamma_{ij} = (\partial v_j/\partial x_i) + (\partial v_i/\partial x_j)$ is the symmetrized velocity gradient tensor. The quantities λ_{1p} and λ_{2p} are two sets of time constants, and the η_p are constants with dimensions of viscosity; convenient forms for these parameters have been found (5, 6) to be

$$\eta_p = \eta_0 \left(\frac{\lambda_{1p}}{\Sigma\, \lambda_{1p}}\right) \tag{5}$$

$$\lambda_{jp} = \lambda_j \left(\frac{1 + n_j}{p + n_j}\right)^{\alpha_j} \quad (j = 1, 2) \tag{6}$$

Two specific choices have been studied extensively:

Spriggs (5):

$$n_j = 0, \quad \alpha_1 = \alpha_2 = \alpha, \quad \lambda_1 = c\lambda_2 = c\lambda \tag{7}$$

Carreau (6):

$$n_j = 1, \quad \alpha_1 \text{ may or may not equal } \alpha_2 \tag{8}$$

Equation (7) results in a five constant model (η_0, λ, α, ϵ, c), and Equation (8) yields a six constant model (η_0, λ_1, λ_2, α_1, α_2, ϵ).

For this model the non-Newtonian viscosity $\eta(\dot{\gamma})$, the normal stress function (4) $\theta(\dot{\gamma})$, and the components of the complex viscosity $\eta'(\omega)$ and $\eta''(\omega)$ have the form:

$$\eta = \sum_{p=1}^{\infty} \frac{\eta_p}{1 + (\lambda_{1p}\,\dot{\gamma})^2} \tag{9}$$

$$\theta = \sum_{p=1}^{\infty} \frac{2\eta_p\lambda_{2p}}{1 + (\lambda_{1p}\,\dot{\gamma})^2} \tag{10}$$

$$\eta' = \sum_{p=1}^{\infty} \frac{\eta_p}{1 + (\lambda_{2p}\,\omega)^2} \tag{11}$$

$$\eta'' = \sum_{p=1}^{\infty} \frac{\eta_p\,\lambda_{2p}\,\omega}{1 + (\lambda_{2p}\,\omega)^2} \tag{12}$$

Insertion of the Equations (5) and (6) into these results yields realistic functions capable of fitting experimental data for polymer solutions and polymer melts. The expressions in Equation (8) have been found (6) to be somewhat better than those in Equation (7). Equations (1) through (7) have been found to be adequate to describe the experimental data obtained by superposing steady and oscillatory shear flow (7).

STEADY STATE FLOW IN THE MAXWELL ORTHOGONAL RHEOMETER

The details of the apparatus sketched in Figure 1 have been described elsewhere (1, 2). The constant angular velocity of the disks is Ω, the horizontal separation of their centers is a, and the vertical plate spacing is b. We use the abbreviation $\psi = a/b$.

The displacement functions are given by

$$\left.\begin{aligned} x &= x' \cos[\Omega(t-t')] - (y' - \psi z') \sin[\Omega(t-t')] \\ y &= x' \sin[\Omega(t-t')] + (y' - \psi z') \cos[\Omega(t-t')] + \psi z' \\ z &= z' \end{aligned}\right\} \tag{13}$$

From these expressions, all the components of $\overline{\boldsymbol{\Gamma}}$ and $\boldsymbol{\Gamma}$ may be calculated as

$$\begin{aligned}
\overline{\Gamma}_{xx} &= \psi^2 \sin^2 \Omega(t-t') & \Gamma_{xx} &= 0 \\
\overline{\Gamma}_{yy} &= \psi^2[1 - \cos \Omega(t-t')]^2 & \Gamma_{yy} &= 0 \\
\overline{\Gamma}_{zz} &= 0 & \Gamma_{zz} &= -2\psi^2\,[1 - \cos \Omega(t-t')] \\
\overline{\Gamma}_{xz} &= \psi \sin \Omega(t-t') & \Gamma_{xz} &= \psi \sin \Omega(t-t') \\
\overline{\Gamma}_{yz} &= \psi\,[1 - \cos \Omega(t-t')] & \Gamma_{yz} &= \psi\,[1 - \cos \Omega(t-t')] \\
\overline{\Gamma}_{xy} &= \psi^2 \sin \Omega(t-t') \cdot [1 - \cos \Omega(t-t')] & \Gamma_{xy} &= 0
\end{aligned} \tag{14}$$

It is further easy to show that the shear rate tensor $\dot{\boldsymbol{\gamma}}$, the vorticity tensor $\boldsymbol{\omega}$, and $II(t')$ are:

$$\dot{\boldsymbol{\gamma}} = \begin{Bmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{Bmatrix} \Omega\psi \tag{15}$$

$$\boldsymbol{\omega} = \begin{Bmatrix} 0 & 2 & -\psi \\ -2 & 0 & 0 \\ \psi & 0 & 0 \end{Bmatrix} \Omega \tag{16}$$

$$II(t') = 2(\Omega\psi)^2 \tag{17}$$

The rheological model in Equations (1) to (4) then yields:

$$\tau_{xz} = -\,\Omega\psi \sum_{p=1}^{\infty} \frac{\eta_p}{[1 + (\lambda_{1p}\,\Omega\psi)^2]\,[1 + (\lambda_{2p}\,\Omega)^2]} \tag{18}$$

$$\tau_{yz} = -\,\Omega\psi \sum_{p=1}^{\infty} \frac{\eta_p(\lambda_{2p}\,\Omega)}{[1 + (\lambda_{1p}\,\Omega\psi)^2]\,[1 + (\lambda_{2p}\,\Omega)^2]} \tag{19}$$

Note that these results are structured differently from the material functions in Equations (9) to (12), which have been the subject of considerable experimental study. In the orthogonal rheometer one measures the values of τ_{xz} and τ_{yz}; but Equations (18) and (19) demonstrate that such measurements do not necessarily yield η' and η'' as indicated earlier (2). It is clear that Equations (18) and (19) suggest that

$$\lim_{\psi\to 0} \left(\frac{\tau_{xz}}{-\Omega\psi}\right) = \eta' \tag{20}$$

$$\lim_{\psi\to 0} \left(\frac{\tau_{yz}}{-\Omega\psi}\right) = \eta'' \tag{21}$$

in which η' and η'' would be given as functions of the angular velocity Ω.

The model used in the previous analysis (2) (a nonlinear Maxwell model with G and λ, in their notation,

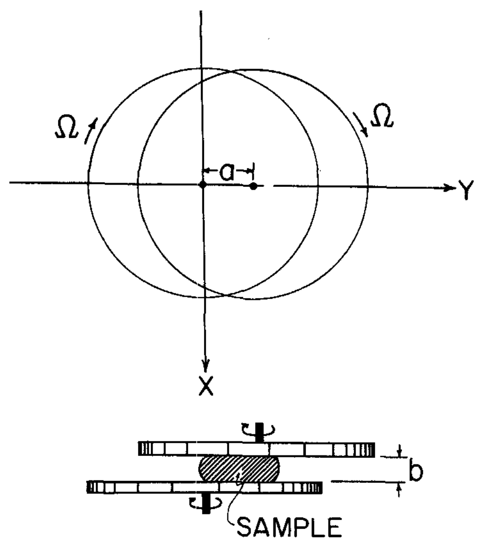

Fig. 1. Schematic Diagram of the Orthogonal Rheometer.

taken to be constants) gives the Maxwell-type expressions for η' and η'' [far less realistic than Equations (11) and (12)] a constant viscosity η_0 (that is, no non-Newtonian effects) and a constant normal stress coefficient θ. In view of the obvious shortcomings of the model, the previous conclusions (2) that measurements of τ_{xz} and τ_{yz} give η' and η'' directly [that is, without a limiting process described in Equations (20) and (21)] have to be seriously questioned.

The experimental data presented by Maxwell and Chartoff (1) were apparently taken at such low values of ψ that the data given in their Figure 5 are actually $\eta'(\omega)$ and those in Figure 7 are $G'(\omega) = \omega\eta''$. Therefore it is not necessary for them to apply a limiting procedure to their experimental data. We have come to this conclusion after a careful examination of their data and extensive correspondence with the authors.

STRESS RELAXATION IN THE MAXWELL ORTHOGONAL RHEOMETER

Consider next an unsteady state experiment in which the steady state flow of §2 prevails for $t < 0$. Then at $t = 0$, the motion is instantaneously stopped. For $t > 0$, the decay of the stresses τ_{xz} and τ_{yz} can be observed (3).

For this experiment, the rheological model in Equations (1) to (4) can easily be used to give:

$$\frac{\tau_{xz}(t>0)}{\tau_{xz}(t<0)} = \frac{\sum_{p=1}^{\infty} \dfrac{\eta_p \exp(-t/\lambda_{2p})}{[1+(\lambda_{1p}\,\Omega\psi)^2]\,[1+(\lambda_{2p}\,\Omega)^2]}}{\sum_{p=1}^{\infty} \dfrac{\eta_p}{[1+(\lambda_{1p}\,\Omega\psi)^2]\,[1+(\lambda_{2p}\,\Omega)^2]}} \tag{22}$$

$$\frac{\tau_{yz}(t>0)}{\tau_{yz}(t<0)} = \frac{\sum_{p=1}^{\infty} \dfrac{\eta_p\,\lambda_{2p} \exp(-t/\lambda_{2p})}{[1+(\lambda_{1p}\,\Omega\psi)^2]\,[1+(\lambda_{2p}\,\Omega)^2]}}{\sum_{p=1}^{\infty} \dfrac{\eta_p\,\lambda_{2p}}{[1+(\lambda_{1p}\,\Omega\psi)^2]\,[1+(\lambda_{2p}\,\Omega)^2]}} \tag{23}$$

The experiments described in this and the preceding section, combined with measurements of the four material functions in Equations (9) to (12) should provide an excellent means of testing rheological models.

ASYMPTOTIC EXPRESSIONS AND NUMERICAL COMPARISONS

By using the model given in Equations (1) through (4) and the expressions given in Equations (5), (6), and (8), one finds the following for the two functions $(\tau_{xz}/-\eta_0\Omega\psi)$ and $(\tau_{yz}/-\eta_0\Omega\psi)$:

$$\left(\frac{\tau_{xz}}{-\eta_0\Omega\psi}\right) = \frac{1}{Z(\alpha_1)-1}\cdot \sum_{p=2}^{\infty} \frac{p^{\alpha_1+2\alpha_2}}{[p^{2\alpha_1}+(2^{\alpha_1}\lambda_1\,\Omega\psi)^2]\,[p^{2\alpha_2}+(2^{\alpha_2}\lambda_2\,\Omega)^2]} \tag{24}$$

$$\left(\frac{\tau_{yz}}{-\eta_0\Omega\psi}\right) = \frac{2^{\alpha_2}\lambda_2\,\Omega}{Z(\alpha_1)-1}\cdot \sum_{p=2}^{\infty} \frac{p^{\alpha_1+2\alpha_2}}{[p^{2\alpha_1}+(2^{\alpha_1}\lambda_1\,\Omega\psi)^2]\,[p^{2\alpha_2}+(2^{\alpha_2}\lambda_2\,\Omega)^2]} \tag{25}$$

where $Z(\alpha_1)$ is the Riemann zeta function.

In order to examine these two functions more closely, let us examine the asymptotic expressions for small ψ:

$$\left(\frac{\tau_{xz}}{-\eta_0\Omega\psi}\right) = \frac{\eta'(\Omega)}{\eta_0} - \frac{(2^{\alpha_1}\lambda_1\,\Omega\psi)^2}{Z(\alpha_1)-1}\cdot \sum_{p=2}^{\infty} \frac{p^{2\alpha_2-3\alpha_1}}{[p^{2\alpha_2}+(2^{\alpha_2}\lambda_2\,\Omega)^2]} + O(\psi^4) \tag{26}$$

$$\left(\frac{\tau_{yz}}{-\eta_0\Omega\psi}\right) = \frac{\eta''(\Omega)}{\eta_0} - \frac{2^{\alpha_2}\lambda_2\,\Omega(2^{\alpha_1}\lambda_1\,\Omega\psi)^2}{Z(\alpha_1)-1}\cdot \sum_{p=2}^{\infty} \frac{p^{\alpha_2-3\alpha_1}}{[p^{2\alpha_2}+(2^{\alpha_2}\lambda_2\,\Omega)^2]} + O(\psi^4) \tag{27}$$

in which $\eta'(\Omega)$ and $\eta''(\Omega)$ are the series given in Equations (11) and (12). Equations (26) and (27) enable one to estimate the discrepancy between the experimental data and the linear response functions η' and η'', in terms of the fluid parameters λ_1, λ_2, α_1, and α_2.

Maxwell and Chartoff (2) presented experimental data for $\psi = 0.094$ and $\psi = 0.44$. When these data of $\tau_{xz}/-\eta_0\Omega\psi$ are plotted as a function of Ω, the data points for both ψ values fall on essentially the same curve (see their Figure 5 and our Figure 2). This means that the terms of $O(\psi^2)$ and higher are not contributing and hence that they are measuring η'.

The data of Maxwell and Chartoff in our Figure 2 can be used to determine some of the parameters in Equations (1) through (7). Maxwell and Chartoff give the value $\eta_0 = 3.8 \times 10^5$ posie. The slope of the curve in the power law region is given by $(1-\alpha_1)/\alpha_2$; from Figure 2 it can be seen that the slope is -0.72. In the absence of viscosity data we make the not unreasonable assumption that $\alpha_1 = \alpha_2 = \alpha$ and conclude that $\alpha = 3.6$. The time constant λ_2 is found to be 14 sec. by using the analog of Equation (47) given in reference 5.

$$\lambda_2 = 2^{-\alpha}\left([Z(\alpha_1)-1]\,(2\alpha/\pi)\,\sin\left[\frac{(\alpha+1)\pi}{2\alpha}\right]\right)^{\frac{\alpha}{1-\alpha}}\cdot (1/\Omega_{\text{int.}}) \tag{28}$$

where $\Omega_{\text{int.}}$ is the value of Ω at which the two linear asymptotes intersect. The constants λ_1 and ϵ cannot be determined from the Maxwell and Chartoff data; experiments on non-Newtonian viscosity and normal stresses could be used to determine them.

ADDITIONAL RESULTS FOR MORE GENERAL MODELS

The rheological model introduced in Equation (1) has the disadvantage of being an empirical one. On the other hand it has been proven to be moderately successful and it has the advantage that it enables one to interconnect the results of viscometric flows and the orthogonal rheometer flow (a nonviscometric flow). The results in Equa-

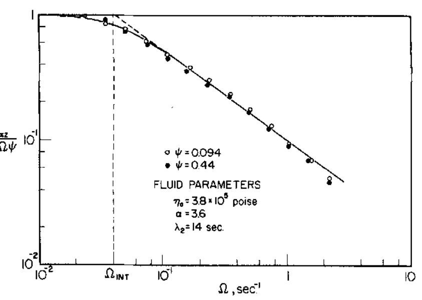

Fig. 2. $(\tau_{xz}/-\eta_0\Omega\psi)$ vs. Ω for a linear polyethylene melt from Figure 4 (1). The solid curve gives η'/η_0 from Equations (5), (6), (8), and (11).

tions (18) and (19) and the corresponding expressions for the other four components of the stress tensor suggest that the stress state in the orthogonal rheometer is given by six material functions, none of which are coincident with any of the three material functions for viscometric flows. It would be helpful to establish this (rather negative) result definitely by using a rheological model less specific than Equation (1).

For flows in which each particle of the fluid executes the same path, rather general functional constitutive equations simplify (*8*) to the Rivlin-Ericksen model (*9*). The latter, in turn, has been further simplified by Giesekus (*10, 11*) for flows in which the substantial derivatives ($D/Dt \equiv \partial/\partial t + \mathbf{v} \cdot \nabla$) of the rate-of-strain tensor and all higher order rate-of-strain tensors [that is, the $\mathbf{e}^{(N)}$ of Oldroyd] are zero. When the latter is further simplified for the special case of the orthogonal rheometer one finds that:

$$
\begin{aligned}
(p + \tau_{xx})\varphi &= \varphi_2\Omega^2\psi^2 + \varphi_{10} \\
(p + \tau_{yy})\varphi &= \varphi_4\Omega^4\psi^2 + \varphi_{10} \\
(p + \tau_{zz})\varphi &= (\varphi_2 + 2\varphi_3)\Omega^2\psi^2 + \varphi_4\Omega^4\psi^2 \\
&\quad + 4(\varphi_4 + \varphi_6)\Omega^4\psi^4 + \varphi_{10} \\
\tau_{xy}\ \varphi &= -\varphi_5\Omega^3\psi^2 + \varphi_9\Omega^5\psi^2 - 2\varphi_7\Omega^5\psi^4 \\
\tau_{xz}\ \varphi &= \varphi_1\Omega\psi - \varphi_8\Omega^3\psi + 2\varphi_5\Omega^3\psi^3 \\
&\quad + (\varphi_7 - 2\varphi_9)\Omega^5\psi^3 + 4\varphi_7\Omega^5\psi^5 \\
\tau_{yz}\ \varphi &= -\varphi_3\Omega^2\psi - (2\varphi_4 + \varphi_6)\Omega^4\psi^3
\end{aligned}
\qquad (29)
$$

in which φ and φ_i are the coefficients used by Rivlin and Ericksen (*9*). They are functions of the joint invariants of all of the Oldroyd *n*th rate-of-strain tensors. For the flow pattern being considered here it can be shown that the φ_i depend only on the invariants tr $\mathbf{e}^{(1)2}$ and tr $\mathbf{e}^{(1)}\mathbf{e}^{(3)}$, where $\mathbf{e}^{(1)}$ and $\mathbf{e}^{(3)}$ are the first and third Oldroyd rate-of-strain tensors; these invariants are just functions of Ω and ψ. None of the functions φ_i can be determined from a steady simple shearing flow. Therefore this rather general approach does not seem to lead to any useful results.

CONCLUSIONS

1. The orthogonal rheometer is a useful device for measuring η' and η'', provided that ψ is sufficiently small so that the material being studied exhibits linear viscoelastic behavior. Equations (26) and (27) provide a means for estimating errors or for making small corrections to data taken in the region where nonlinear effects are evident.

2. The Maxwell-Chartoff data on η' and η'' for polyethylene melts were taken with sufficiently small ψ that the $O(\psi^2)$ terms in Equations (26) and (27) are not important.

3. The Maxwell-Chartoff data are adequately described by the model used in Equations (1) to (7).

4. The model of Equations (1) to (7) suggests that for large ψ new material functions can be measured by using the Maxwell orthogonal rheometer, namely the stress components in steady flow [see Equations (18) and (19)] and the stress components in stress relaxation [see Equations (22) and (23)]. Such experiments would be most helpful for testing constitutive equations, inasmuch as the flow involved is nonviscometric.

5. The simple fluid model results show how much more complicated the orthogonal rheometer flow is than the viscometric flows. The analytical expressions have little predictive value, however.

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NOTATION

a = horizontal displacement of disk centers
b = vertical separation of disks
c = shift factor
m = memory function
n_1, n_2 = dimensionless constants
t = time
x_i = coordinates at time t
x_i' = coordinates at time t'
α_1, α_2, α = dimensionless exponents
$\dot{\boldsymbol{\gamma}}$ = rate of strain tensor
$\overline{\boldsymbol{\Gamma}}$, $\boldsymbol{\Gamma}$ = strain tensors
ϵ = parameter indicating deviation from Weissenberg hypothesis
η', η'' = components of complex viscosity, η^*
η = non-Newtonian viscosity
η_p = parameters
η_0 = zero-shear rate viscosity
θ = normal stress function
λ_{1p}, λ_{2p} = time constants
$\boldsymbol{\tau}$ = stress tensor
ψ = a/b ratio
ω = frequency of oscillation
$\boldsymbol{\omega}$ = vorticity tensor
Ω = angular velocity in rad./sec.
II = second invariant of rate of deformation tensor

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