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Analysis of the Maxwell Orthogonal Rheometer

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Synopsis

An analysis of the Maxwell orthogonal rheometer for polymer melts is made by using the constitutive equation of White and Metzner. The results indicate that the shear deformation involved is oscillatory and that the storage and loss moduli of the melt may be derived from the measured stress response.

The rheology of polymer melts is an area of considerable importance both from a practical standpoint and for understanding material behavior. Recently Maxwell and Chartoff[1] reported on a new device, termed an orthogonal rheometer, for characterizing polymer melt properties. To date, however, its acceptance has been hindered by the need for an analysis of the manner in which the three forces measured by the device are related to the response of the material to the imposed deformation.

Figure 1 depicts the rheometer schematically. The molten sample of thickness l is contained between parallel circular plates, the axes of which are not colinear but separated by a distance a along the y axis. The lower plate is driven at a rotational velocity ω. The upper plate is allowed to rotate freely at the same angular velocity. Three orthogonal forces are measured along the x, y, and z axes.

We have analyzed the rheometer by formulating the kinematics in a nonorthogonal curvilinear coordinate system in which the motion of the fluid can be easily written. This system, depicted in Figure 2, has coordinates (θ', r', z') related to the Cartesian coordinates (x, y, z) by

$$\theta' = \tan^{-1}\{[y-(a/l)z]/x\} \quad (1a)$$

$$(r')^2 = x^2 + [y-(a/l)z]^2 \quad (1b)$$

$$z' = [(a^2 + l^2)^{1/2}/l]z \quad (1c)$$

The observed motion in the rheometer is one for which each layer of melt parallel to the rheometer plates rotates about its intersection with the z' axis at angular velocity ω. Thus in the (θ', r', z') system the velocity field has the simple form

$$\dot{\theta}' = \omega \quad (2a)$$

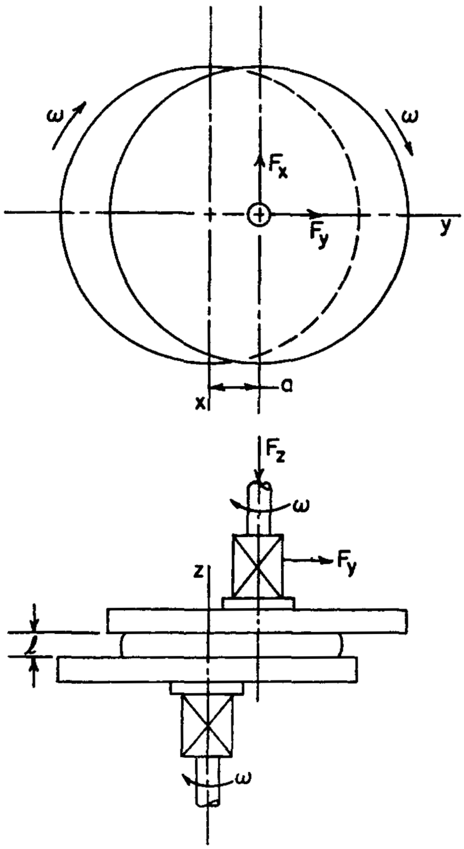

Fig. 1. Schematic drawing of orthogonal rheometer.

$$\dot{r}' = 0 \tag{2b}$$

$$\dot{z}' = 0 \tag{2c}$$

By using eqs. (1) the velocity field in the (x, y, z) system may be obtained:

$$\dot{x} = -\omega[y-(a/l)z] \tag{3a}$$

$$\dot{y} = \omega x \tag{3b}$$

$$\dot{z} = 0 \tag{3c}$$

Because the deformation experienced by the material is uniform in the rheometer, the stress response is also uniform, neglecting edge effects, inertia, and gravitational forces; hence the equations of motion are identically satisfied and need not be considered.

To obtain the stress response, a constitutive law for the material must be assumed. We have chosen the equation for a Maxwell fluid as formu-

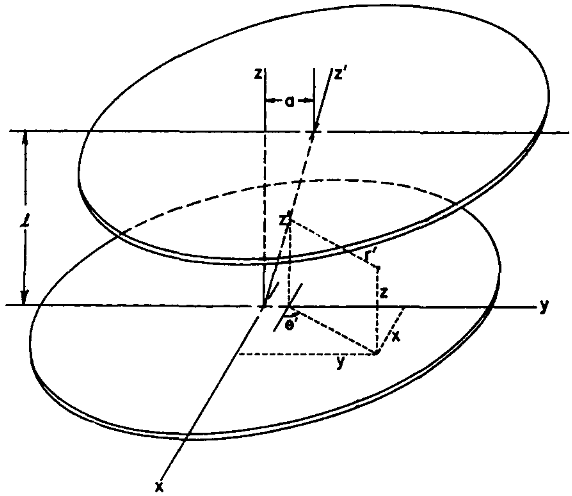

Fig. 2. Nonorthogonal coordinate system used for analysis of orthogonal rheometer.

lated by White and Metzner[2] for large deformations of polymer systems Their equation in component form is

$$P^{ij} = 2G\lambda D^{ij} - \lambda(\delta P^{ij}/\delta t) \tag{4}$$

where

$$D^{ij} = {}^1/_2(\dot{x}^i{}_{,m}g^{mj} + \dot{x}^j{}_{,m}g^{im}) \tag{5a}$$

$$T^{ij} = -\alpha g^{ij} + P^{ij} \tag{5b}$$

$$\delta P^{ij}/\delta t = (\partial P^{ij}/\partial t) + \dot{x}^k(\partial P^{ij}/\partial x^k) - \dot{x}^i{}_{,m}P^{mj} - \dot{x}^j{}_{,m}P^{im} \tag{5c}$$

Here T^{ij} and P^{ij} are the contravariant components of the total and deviatoric stress tensors, respectively, D^{ij} is the rate of deformation tensor, g^{kl} is the metric tensor, and α is the hydrostatic pressure. The material parameters for the Maxwell fluid are the modulus G and the relaxation time λ. The rate of deformation tensor D^{ij} may be formulated by using eqs. (3) and the system of equations above solved for the three stresses measured by the rheometer.

An alternate approach to analyzing the motion of material in the orthogonal rheometer makes use of a nonorthogonal rotating coordinate system, depicted in Figure 3. In particular, the material motion may be matched exactly with the following relation between the laboratory coordinate system (x, y, z) and the rotating system (r, θ, z'):

$$x = r \cos(\theta + \omega t) \tag{6a}$$

$$y = r \sin(\theta + \omega t) + [a/(a^2 + l^2)^{1/2}]z \tag{6b}$$

$$z = [l/(a^2 + l^2)^{1/2}]z' \tag{6c}$$

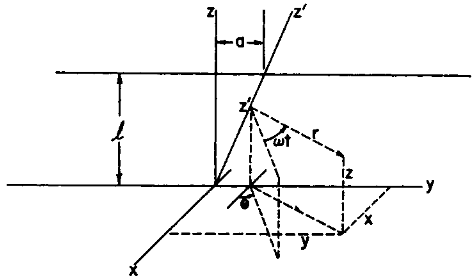

Fig. 3. Nonorthogonal rotating coordinate system used for analysis of orthogonal rheometer.

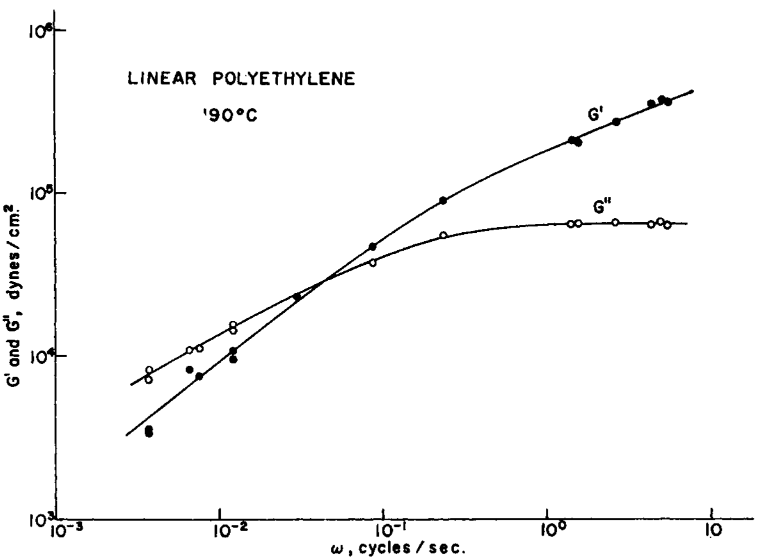

Fig. 4. Storage and loss moduli of linear polyethylene at 190°C.

With this formulation the covariant and contravariant base vectors ($\mathbf{e}_a$,$\mathbf{e}^a$) and metric tensors (g_{ab},g^{ab}) for the rotating coordinate frame may be calculated. The utility of this rotating system is that in it the velocity of each material point becomes identically zero. It is then straightforward to find the deviatoric stress with a constitutive equation of the form:

$$p^{ab} = f^{ab}(g^{cd}) \tag{7}$$

This matrix in turn can be used to give the components of the stress measured in the laboratory system:

$$P_{kl} = p^{ab}\, \mathbf{e}_a(\mathbf{e}_b \cdot \mathbf{i}_l) \cdot \mathbf{i}_k \tag{8}$$

The results of the analysis from both approaches with the use of the White and Metzner constitutive equation are:

$$T_{xz} = \frac{F_x}{A} = \frac{G\lambda\omega}{1 + (\lambda\omega)^2}\frac{a}{l} \tag{9a}$$

$$T_{yz} = \frac{F_y}{A} = \frac{G(\lambda\omega)^2}{1 + (\lambda\omega)^2}\frac{a}{l} \tag{9b}$$

$$T_{zz} = \frac{F_z}{A} = -p - \frac{2}{3}\frac{a}{l}T_{yz} \tag{9c}$$

where $p = -\,^1/_3$ trace T^{ij} is the isotropic hydrostatic pressure and A is the area of material contacting the upper plate of the rheometer. Equations (9a) and (9b) are seen to yield respectively the familiar expressions for the loss and storage moduli, $G''(\omega)$ and $G'(\omega)$, of a Maxwell body undergoing oscillatory shear deformation of strain amplitude a/l. The normal force F_z is related to the storage modulus and hence the elastic response of the material.

These results indicate that the orthogonal rheometer is a dynamic tester for polymer melts in which $G'(\omega)$ and $G''(\omega)$ may be measured. Figure 4 shows these viscoelastic functions as measured in the rheometer for linear polyethylene of melt index 0.9 at 190°C. The curves are characteristic of these functions in the time range corresponding to the entanglement and terminal zones of the relaxation spectrum.

We believe that the orthogonal rheometer should prove to be a valuable research tool for obtaining the linear viscoelastic properties of polymer melts.

We wish to express our appreciation to Professor Bryce Maxwell, Professor W. R. Schowalter, and Mr. R. P. Chartoff for many helpful discussions with regard to the analysis presented here. We further thank Professor Maxwell and Mr. Chartoff for allowing us to use the data appearing in Fig. 4 prior to its publication.

References

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2. J. L. White and A. B. Metzner, *J. Appl. Polymer Sci.*, **7**, 1867 (1963).

Résumé

On a effectué l'analyse du rhéomètre orthogonal de Maxwell pour des polymères fondus en utilisant l'équation de constitution de White et de Metzner. Les résultats indiquent que la déformation au cisaillement est oscillatoire et que le module d'accumulation de perte de la masse fondue peut être dérivé au départ de la réponse à la tension.

Zusammenfassung

Eine Analyse des Maxwell-Orthogonalrheometers für Polymerschmelzen wird mit Verwendung der grundlegenden Beziehung von White und Metzner durchgeführt. Die Ergebnisse zeigen, dass die auftretende Scherungsverformung Oszillationscharakter besitzt und dass aus dem gemessenen Verhalten unter Spannungsbeanspruchung Real- und Imaginärteil des komplexen Modul abgeleitet werden können.

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