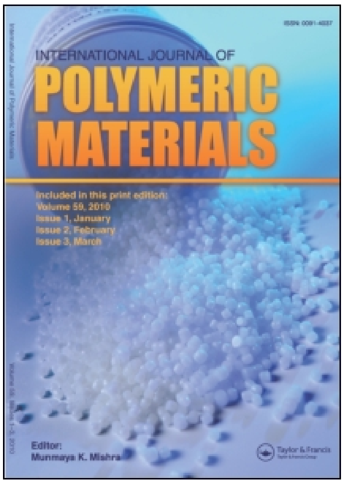

International Journal of Polymeric Materials
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

Dynamic Mechanical Testing of Viscoelastic Solids in Free and Forced Oscillation: Experiments with a Modified Weissenberg Rheogoniometer

Yoon S. Ko[a]; W. C. Forsman[a]
[a] Department of Chemical Engineering, University of Penna, Philadelphia, PA

To cite this Article Ko, Yoon S. and Forsman, W. C.(1980) 'Dynamic Mechanical Testing of Viscoelastic Solids in Free and Forced Oscillation: Experiments with a Modified Weissenberg Rheogoniometer', International Journal of Polymeric Materials, 8: 1, 53 — 63

To link to this Article: DOI: 10.1080/00914038008077934

URL: http://dx.doi.org/10.1080/00914038008077934

Intern. J. Polymeric Mater., 1980, Vol. 8, pp. 53–63
0091–4037/80/0801–0053$04.50/0

Dynamic Mechanical Testing of Viscoelastic Solids in Free and Forced Oscillation: Experiments with a Modified Weissenberg Rheogoniometer

YOON S. KO and W. C. FORSMAN

Department of Chemical Engineering, University of Penna, Philadelphia, PA 19104

(*Received November 1, 1978*)

A Weissenberg rheogoniometer, a device most commonly associated with the measurement of viscoelastic properties of fluids, was modified to measure the storage and loss moduli of viscoelastic solids by both free and forced oscillation. Rigorous equations were derived by Fourier transform techniques for data analysis in both types of experiments. Data obtained for an epoxy resin system by both modes of testing were in excellent agreement.

INTRODUCTION

Dynamic mechanical measurements provide an important means of characterizing the structure of polymers and polymer systems. Indeed viscoelastic properties are useful indicators of processing behavior and utility of finished materials. Among a number of instruments used to measure the viscoelastic properties of polymeric solids, the torsion pendulum is one of the most well known and best suited, especially for the study of low-loss materials. In recent years, a variety of instruments[1,2] of this class have been developed using advanced techniques and instrumentation. In this paper, we describe a particularly versatile instrument for characterizing viscoelastic solids which capitalizes on the precision instrumentation and machinework of the Weissenberg rheogoniometer, an instrument more commonly associated with the testing of viscoelastic fluids. It is important to note that the method requires only auxiliary equipment; no modification of the basic instrument is required.

Analyzing experimental data taken in forced oscillation is straightforward, as long as they are taken at steady-state and *all* calibration constants are properly determined. Analysis of data taken in free oscillation is, however, often treated improperly. Most often, a torsion pendulum system is described mathematically as a simple model such as a Voigt or Maxwell element. This approach leads to simple equations that can be solved for the dynamic modulus of the specimen. If the material does not behave according to the assumed model, the analysis will not give the correct value of the dynamic modulus. The correct analysis has, however, already been worked out.[3] In a similar problem, analysis of the vibrating reed, it has been shown that improper analysis leads to substantial error as the temperature approaches the glass transition.[4] To avoid any such difficulties, all analyses in this paper were done using complete Fourier transform analysis.

APPARATUS

The instrument used in this research was a Model R-18 rheogoniometer, manufactured by Sangamo Controls Ltd. This device is most commonly used as a cone and plate viscometer, and is equipped to operate in dynamic or steady shearing, or combinations of the two. The details of its construction and operation are given elsewhere.[5,6] In order to use this equipment for testing solid specimens, the two platens were replaced with two chucks which clamp both ends of a rod specimen (Figure 1). An electrical heating chamber surrounding the specimen allows a choice of chamber temperature from ambient to 400°C. Provisions were made to pass nitrogen gas through channels in the heating jacket and then into the chamber. This technique helped maintain temperature control, and at the same time purged the chambers in order to prevent oxidation of the specimen at elevated temperatures. The specimen temperature was measured by an iron/constantan thermocouple with its tip slightly touching the surface of the specimen. For forced oscillation, the specimen was given a sinusoidal torsional displacement by the lower chuck, which was driven by the oscillation drive unit consisting of the motor gear box and the variable sine wave generator. The upper chuck was attached to the air bearing rotor, which makes angular displacement against the torsion bar according to the combined material response of the specimen and the torsion bar. The torsion bar, against which the tangential stress is measured, is supplied by the manufacturer in a range of standard sizes. The top of the torsion bar is held rigidly in a clamp at the top of the air bearing casting and the bottom is clamped to the top of the air bearing rotor. In the case of forced oscillation, signals are taken from a transducer located on the oscillation drive shaft and another at the end of a radius arm which is attached to the air bearing rotor. The signals from the two transducers are passed through separate demodulating–amplifying–filtering networks and are recorded on the oscillograph paper by a multichannel ultraviolet recorder. In the case of free oscillation, the oscillation drive unit was not used, and the lower chuck was locked in position. An inertia arm was attached to the air bearing rotor. The signal from the transducer at the end of the radius arm was recorded after pulsing the inertia arm.

ANALYSIS

Free oscillation

Forsman[3] developed a method of analyzing freely oscillating linear viscoelastic systems (Figure 2) using Fourier Transform methods that require no model or assumptions about a distribution of relaxation times. In this treatment one writes:

$$G' = [I(\omega^2 - \alpha^2) - k']/Q \qquad (1)$$

$$G'' = (2I\omega\alpha - k'')/Q \qquad (2)$$

where

G', G'' = Real and imaginary parts of the complex dynamic shear modulus G^*.

I = moment of inertia of the oscillating inertial element.

ω = oscillation frequency (rad/sec).

α = damping constant, i.e., the negative slope of the line obtained from a log–log plot of amplitude as a function of time.

k', k'' = Real and imaginary parts of k^*, the complex torsional spring constant of the oscillating system in the absence of a specimen.

Q = Geometrical constant. For a purely elastic specimen the product of Q and the shear modulus is the proportionality factor relating torque and the torsional displacement (in radians).

Considering a freely oscillating system without a specimen as a special case, Eqs. (1) and (2) give the following relationships between k', k'', and I:

$$k' = I(\omega^2 - \alpha^2) \qquad (3)$$

$$k'' = 2I\omega_0\alpha_0 \qquad (4)$$

In Eqs. (3) and (4), the subscript zero therefore denotes values of resonance frequency and damping constant determined for the system without a specimen in the chuck.

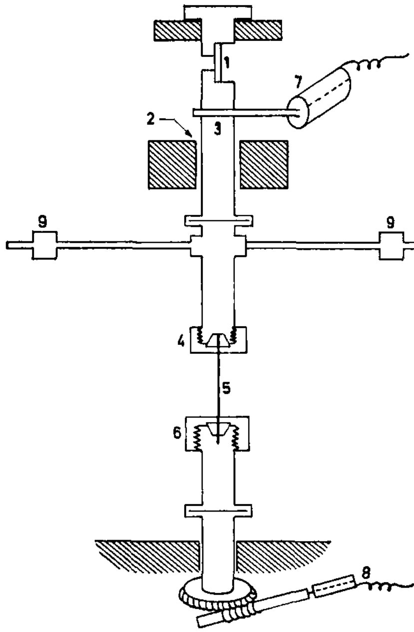

FIGURE 1 Dynamic mechanical measurements on the Weissenberg rheogoniometer. (1) Torsion bar. (2) Air bearing starter. (3) Air bearing rotor. (4) Upper chuck. (5) Solid polymer specimen. (6) Lower chuck. (7) Torsion head transducer. (8) Input transducer for forced oscillation experiments. (9) Inertial arm for free oscillation experiments.

Forced oscillation

Writing a torque balance on the inertial element shown in Figure 2b gives

$$I\left(\frac{d^2\theta_2}{dt^2}\right)+T(t) = T_s(t) \tag{5}$$

where

θ_2 = Angle of deflection of the inertial element.

$T_s(t)$ = Torque transmitted from the specimen which is driven by the lower chuck.

$T(t)$ = Counter-balancing torque due to the torsion bar and friction mechanisms.

The Fourier transform of Eq. (5) gives

$$-I\nu^2\bar{\theta}_2+\bar{T} = \bar{T}_s \tag{6}$$

where ν is the transform variable.

We know that if an element of any linear viscoelastic material is subjected to an arbitrary shear strain as a function of time, $\gamma(t)$, and responds with an associated shear stress, $\tau(t)$, we can write[7]

$$\bar{\tau}(v) = G^*\bar{\gamma}(v)$$

where G^* is the dynamic modulus considered as a function of v.

Consequently, the transform of the viscoelastic torques can be written

$$\bar{T} = k^*\bar{\theta}_2 \tag{7}$$

and

$$\bar{T}_s = QG^*(\bar{\theta}_1 - \bar{\theta}_2) \tag{8}$$

where θ_1 is the angle of deflection of the lower chuck.

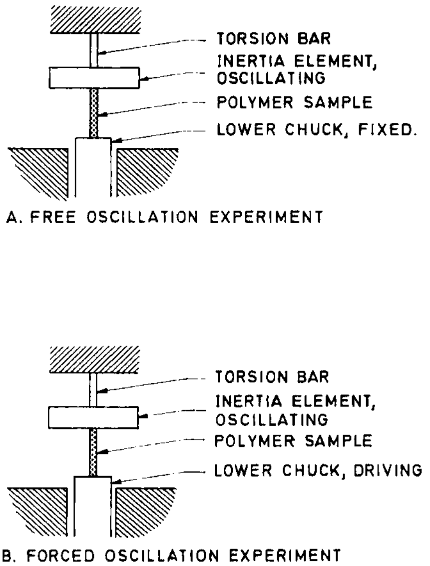

FIGURE 2 Configuration for free and forced oscillation experiments.

Substituting Eq. (7) and (8) in Eq. (6) and rearranging gives

$$QG^*(\bar{\theta}_1 - \bar{\theta}_2) = (-Iv^2 + k^*)\bar{\theta}_2 \tag{9}$$

If the specimen is driven by the lower chuck with sinusoidally varying angular deflection, at steady state, the inertial element would also respond with sinusoidally varying angular deflection if the amplitudes are sufficiently small that both the specimen and the resistant force mechanism are linear viscoelastic. Hence, we can write

$$\theta_1 = A\, e^{i\omega t} \tag{10}$$

$$\theta_2 = B\, e^{i(\omega t + \phi)} \tag{11}$$

where

A and B = amplitudes of angular deflection of the lower chuck and the inertial element respectively.

ω = Frequency of the forced oscillation (rad/sec).

ϕ = Phase lag in the response of the inertial element to the oscillation of the lower chuck.

The transform of Eq. (10) and (11) gives

$$\bar{\theta}_1 = 2\pi A\delta(\nu-\omega) \quad (12)$$

$$\bar{\theta}_2 = 2\pi B\delta(\nu-\omega)\, e^{i\phi} \quad (13)$$

where $\delta(\nu)$ is Dirac delta function.

From Eqs. (9), (12) and (13), we have

$$Q(A-Be^{i\phi})G^* = (k^*-I\nu^2)B\, e^{i\phi} \quad (14)$$

Replacing k^* in Eq. (14) by Eqs. (3) and (4) gives

$$G^*(\nu) = \frac{BI\, e^{i\phi}[(\omega_0^2-\nu^2-\alpha_0^2)+i\, 2\omega_0\alpha_0]}{Q(A-B\, e^{i\phi})}$$

By identifying ν with the oscillation frequency ω, we can write

$$G^*(\omega) = \frac{BI\, e^{i\phi}[\omega_0^2-\omega^2-\alpha_0^2)+i\, 2\omega_0\alpha_0]}{Q(A-B\, e^{i\phi})}$$

Or, by rearranging,

$$G^*(\omega) = \frac{BI[(\omega_0^2-\omega^2-\alpha_0^2)+i\, 2\omega_0\alpha_0]}{Q(A\cos\phi-B-iA\sin\phi)} \quad (15)$$

By separating real and imaginary parts of G^* in the Eq. (15) above, we get

$$G'(\omega) = \left(\frac{I\omega_0^2}{Q}\right)\left(\frac{B}{A}\right)\left(\cos\phi-\frac{B}{A}\right)\left\{\frac{1-\omega^2/\omega_0^2-\alpha_0^2/\omega_0}{1-2(B/A)\cos\phi+B^2/A^2}\right\} \quad (16)$$

$$G''(\omega) = \left(\frac{I\omega_0^2}{Q}\right)\left(\frac{B}{A}\right)\left\{\frac{\sin\phi(1-\omega^2/\omega_0^2-\alpha_0^2/\omega_0^2)+2(\alpha_0/\omega_0)(\cos\phi-B/A)}{1-2(B/A)\cos\phi+B^2/A^2}\right\} \quad (17)$$

$$\tan\delta = G'(\omega)/G''(\omega) \quad (18)$$

where δ is the true phase angle of the specimen.

Since the torsion bar and other parts of the torsion head assembly are made of stainless steel and are thus close to ideal elastic materials and the air damping is negligible, we can assume $\alpha_0/\omega_0 \cong 0$ and, hence, $k' \cong k = I\omega_0^2$. Also $k'' \cong 0$. Then we can write

$$G' \cong \left(\frac{k}{Q}\right)\left(\frac{B}{A}\right)\left(\cos\phi-\frac{B}{A}\right)\left\{\frac{1-\omega^2\omega_0^2}{1-2(B/A)\cos\phi+B^2/A^2}\right\} \quad (19)$$

$$G'' \cong \left(\frac{k}{Q}\right)\left(\frac{B}{A}\right)\left\{\frac{\sin\phi(1-\omega^2/\omega_0^2)}{1-2(B/A)\cos\phi+B^2/A^2}\right\} \tag{20}$$

where the calibration constants can be obtained as follows:

1) ω_0 and α_0 are obtained from a free oscillation experiment without a specimen in the chuck.

2) k', k'' and I are calculated from Eqs. (3) and (4) after any one of the three constants is measured independently. Practically, k' is the one that can be measured most easily and accurately because it is essentially just equal to the static elastic torsional rigidity (k), or spring constant of the torsion bar, and it is given by the manufacturer or can be easily determined by a simple torque-displacement experiment.

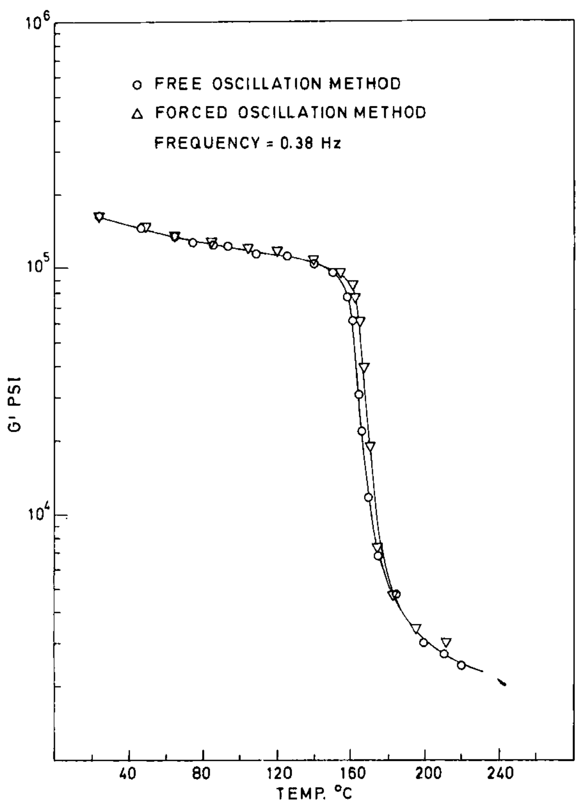

FIGURE 3 G' as a function of temperature for epoxy resin determined in free and forced oscillation at 0.38 Hz.

3) Q, the geometrical constant of the specimen: The elastic solution for torsional boundary value problems can be directly used for the above equations. For a circular rod,[7]

$$Q = \frac{\pi d^4}{32l}.$$

where l and d are the length and diameter of the rod.

TESTING THE EQUIPMENT

The system was tested with an epoxy cast resin prepared from Epon 828 and meta-phenylene diamine (MPDA) supplied by Shell Chemical Company. The resin and the curing agent were well mixed at 70°C according to the stoichiometric ratio supplied by the manufacturer (14.5 parts of MPDA per 100 parts of resin). After mixing, the mixture was degassed under vacuum

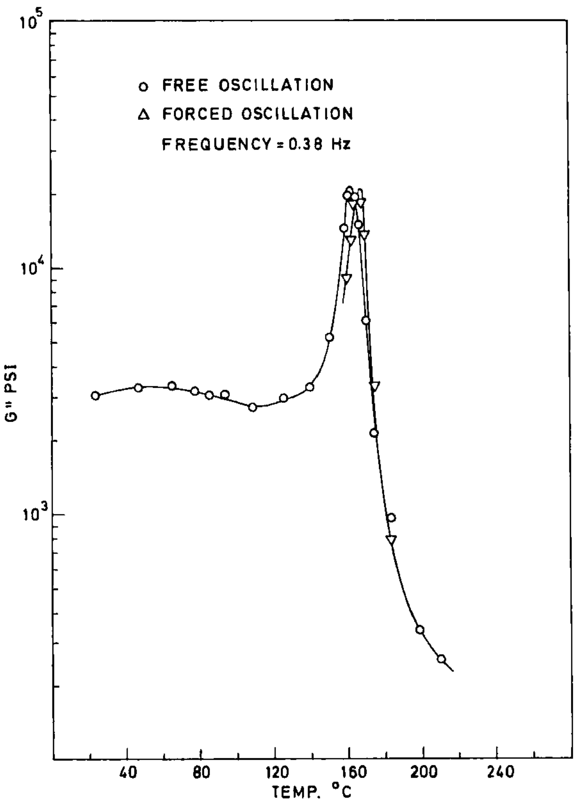

FIGURE 4 G'' as function of temperature for epoxy resin determined in free and forced oscillation at 0.38 Hz.

and cast in glass tubing to give specimens 5.5 cm long and 0.325 cm in diameter. The resin mixture was cured at 200°F for 1 hour. Two specimens made at the same time were used for the free and forced oscillation experiments A gauge length of about 4 cm was used in both experiments.

The specimen chamber was continuously purged with nitrogen during both experiments. In each case, the specimen temperature was raised at an approximate rate of 2°C/min between measurements, and the system was maintained at constant temperature for 15 minutes before each measurement to assure that the testing temperature was uniform throughout the specimen. Every effort was made to keep the testing conditions the same for both methods. The G' and G'' vs. temperature curves obtained by the two methods are compared in Figures 3 and 4 for a fixed frequency of 0.38 Hz, and G' and tan δ vs. temperature curves obtained by forced oscillation for several frequencies are shown in Figure 5. For the free oscillation experiment, the

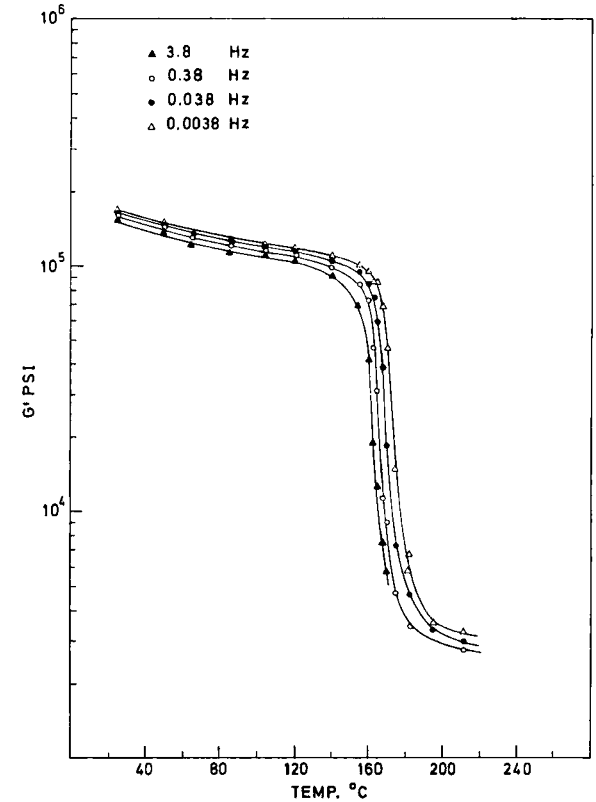

FIGURE 5 G' as a function of temperature for epoxy determined in forced oscillation at frequencies from 3.8×10^{-3} to 3.8 Hz.

frequency was kept constant by adjusting the moment of inertial and the stiffness of the torsion bar.

DISCUSSION

Our experience has been that the term (B/A) in Eqs. (16)–(20) is not negligible when the Rheogoniometer is used in forced oscillation with solid specimens, as is often the case for experiments with fluids.[5,6] Equations considered applicable for determining G' and G'' for fluids[5,6] can therefore not be modified simply by changing geometrical factors. A major limitation in using the Rheogoniometer in viscoelasticity measurements stems from the limitation on the accuracy in determining ϕ, the phase difference between driving and response oscillations. This especially effects the minimum values of G'' or $\tan \delta$ that can be determined by forced oscillation.

Indeed, any reasonable accuracy is lost with decreasing damping well before air damping (principally in the air bearing) contributes significantly to the damping of the entire system. Thus the term (α_0/ω_0) makes a negligible contribution to Eqs. (16) and (17), and the approximations given in Eqs. (19) and (20) are perfectly adequate.

On the other hand, experiments in free oscillation are particularly applicable to low-damping materials. Air damping must then be considered, so both terms α and α_0 must be retained in Eqs. (1)–(4). As for forced oscillation, then, standard Rheogoniometer equations for free oscillation[5,6] are inapplicable to experiments on viscoelastic solids.

If $\tan \delta$ is less than about 0.2, the forced oscillation method will not give usable values of $\tan \delta$ and G''. Figures 3 and 4 thus only display values of G' and G'' that could be determined to $\pm 5\%$ or better. Over the temperature regime for which G' and G'' can be determined by both methods, the agreement is excellent. The difference in the values of the moduli determined by free and forced oscillation in the temperature interval 160–164°C undoubtedly represents temperature error and not error in determining G' and G''.

In conclusion, we find that fitting the Rheogoniometer with modest auxiliary equipment, and applying proper analysis, give a precision instrument for determining the viscoelastic properties of polymeric solids by both forced and free oscillation. Which mode of operation would be selected would, of course, depend upon the problem. Since the advantages and disadvantages of the two methods are well known, we will not consider them here.

Acknowledgment

This work was supported by the National Science Foundation MRL Program under grant number DMR 76-0078.

References

1. J. K. Gillham and M. B. Roller, *Poly. Eng. Sci.* **11,** 295 (1971).
2. B. E. Read and G. D. Dean, AGARD Lecture Series No. 55 on Composition Materials, 4–1, published May, 1972.
3. W. C. Forsman, *Macromolecules* **4,** 262 (1971).
4. P. G. Wapner and W. C. Forsman, *Transactions of the Society of Rheology* **15,** 262 (1971).
5. A. Jobling and J. E. Roberts, Weissenberg Theogoniometer Instruction manual, Sangamo Controls Ltd., Bognor Regis, England.
6. K. Weissenberg, Testing of Materials by Means of the Rheogoniometer, Sangamo Controls Ltd., Bognor Regis, England.
7. Timoshenko, *Theory of Elasticity*, McGraw-Hill, New York, 1934.