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Two-Dimensional Linear Viscoelasticity of Paper

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Summary. Linear viscoelastic properties of laboratory handsheets have been investigated from the two dimensional aspect. According to the linear theory of viscoelasticity, the behavior of transverse isotropic materials such as handsheets subjected to plane stresses is fully described by the two in-plane relaxation functions $G_{11}(t)$ and $G_{12}(t)$. In the present paper, some viscoelastic characteristic functions describing responses to in-plane deformation histories are derived from $G_{11}(t)$ and $G_{12}(t)$ determined by strip biaxial stress relaxation testing. The predicted uniaxial relaxation function curve was in good agreement with the experimental one, and the viscoelastic Poisson's ratios in uniaxial stress relaxation and in uniaxial constant strain rate extension were decrease functions of time. Effects of beating on the areal dilatation and shear relaxation functions are discussed by introducing the classical concept of relaxation spectrum.

Introduction

Linear viscoelastic properties of engineering materials such as polymer sheets, fibers, woods and paper have been extensively studied by many authors (Ferry 1960). For isotropic viscoelastic materials their properties are, as well known, fully characterized by two material functions, that is, relaxation moduli (or creep compliances) in shear and in dilatation (Christensen 1971); for incompressible isotropic materials such as rubber, the number of the independent material functions are reduced to one. For anisotropic materials, which are often found in many engineering materials, complete representation of their properties, however, requires a large number of material functions (Leitman 1973); the experimental determination of all functions is highly difficult and troublesome compared with the isotropic case; therefore results obtained for anisotropic materials have been so far discussed on the basis of the one-dimensional theory of linear viscoelasticity (Gross 1953).

A knowledge of two or three-dimensional viscoelastic properties will provide foundations for viscoelastic stress analyses of many practically important problems as well as for general evaluation of material properties.

In the previous paper (Uesaka 1979a), the constitutive relation of an orthotropic linear viscoelastic material for plane stresses was considered, and the in-plane relaxation functions of some machine-made papers were determined from the strip biaxial stress

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relaxation tests using a biaxial tensile tester. In the present paper the in-plane relaxation functions are determined on laboratory handsheets (transverse isotropic materials), and some characteristic functions for in-plane deformation histories are predicted from the functions using the Laplace transform technique. The characteristic functions chosen are: the uniaxial relaxation function E(t) (relaxation modulus), the Poisson's ratio in uniaxial stress relaxation ν^r (t), the Poisson's ratio in uniaxial constant strain-rate extension ν^u(t), the relaxation function in areal dilatation K(t) and the relaxation function in shear G(t). The experimental value for E(t) is compared with the prediction and the time dependences of ν^r(t) and ν^u(t) are considered. Effects of beating on K(t) and G(t) are also studied by introducing the concept of relaxation spectrum.

Viscoelastic Characteristic Functions for In-Plane Deformation Histories

The infinitesimal viscoelastic constitutive equation of an anisotropic body restricted to isothermal conditions is given in cartesian coordinates by (Rogers 1963)

$$\sigma_{ij}(t) = \int_{-\infty}^{t} G_{ijkl}(t-\tau)\,\frac{de_{kl}(\tau)}{d\tau}\,d\tau \tag{1}$$

where σ_{ij} and e_{kl} are components of stress tensor and infinitesimal strain tensor, respectively, and G_{ijkl} are relaxation functions. For the transverse isotropic material which is at rest prior to t = 0 and is subjected to plane stresses ($\sigma_{33}(t) = \sigma_{23}(t) = = \sigma_{13}(t) = 0$), Eq. (1) becomes (Uesaka 1979a)[1]

$$\sigma_{11}(t) = G_{11} * de_{11} + G_{12} * de_{22} \tag{2.a}$$

$$\sigma_{22}(t) = G_{12} * de_{11} + G_{11} * de_{22} \tag{2.b}$$

$$\sigma_{12}(t) = \frac{1}{2}(G_{11} - G_{12}) * d\gamma_{12} \tag{2.c}$$

where $f * dg$ stands for the function defined by the Stieltjes integral

$$f * dg = f(t)g(0+) + \int_{0}^{t} f(t-\tau)\,\frac{dg(\tau)}{d\tau}\,d\tau \tag{3}$$

and γ_{12} is the engineering shear strain (= $2e_{12}$). From the material symmetry, the in-plane relaxation functions $G_{ij}(t)$ are associated with $G_{ijkl}(t)$ by

$$\begin{aligned} \overline{G}_{11}(s) &= \overline{G}_{1111}(s) - \frac{\overline{G}_{3311}(s)\,\overline{G}_{1133}(s)}{\overline{G}_{3333}(s)} \\ \overline{G}_{12}(s) &= \overline{G}_{1122}(s) - \frac{\overline{G}_{3311}(s)\,\overline{G}_{1133}(s)}{\overline{G}_{3333}(s)} \end{aligned} \tag{4}$$

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[1] For an orthotropic body, the independent in-plane relaxation functions are $G_{11}(t)$, $G_{12}(t)$, $G_{22}(t)$, $G_{21}(t)$ and $G_{33}(t)$, of which two (G_{12}, G_{21}) are equal if $G_{ijkl} = G_{klij}$ is assumed (Uesaka 1979a)

where $\bar{G}_{ij}(s)$ denotes the Laplace transform of $G_{ij}(t)$. The functions $G_{11}(t)$ and $G_{12}(t)$ in Eq. (2) are the independent in-plane relaxation functions for a transverse isotropic body, from which viscoelastic characteristic functions describing the behavior of materials for in-plane deformation histories can be derived as follows.

In the case of stress relaxation in isotropic areal dilatation, such conditions are given by

$$e_{11}(t) = e_{22}(t) = \frac{1}{2}\, e(t) = \frac{1}{2}\mathring{e}U(t)\,,\quad \gamma_{12}(t) = 0 \tag{5}$$

where $e(t)$ is the areal dilatation (= $e_{11}(t) + e_{22}(t)$), $\mathring{e}$ is the applied initial strain, and $U(t)$ is the unit step function. The corresponding stresses are given, from Eq. (2), by

$$\sigma_{11}(t) = \sigma_{22}(t) = \frac{1}{2}\,\{G_{11}(t) + G_{12}(t)\}\,\mathring{e} = K(t)\mathring{e}\,,\ \sigma_{12}(t) = 0 \tag{6}$$

and we shall call the function $K(t)$ (= $\{G_{11}(t) + G_{12}(t)\}/2$) in Eq. (6) "relaxation function in areal dilatation". In a similar way as above, the relaxation function in shear $G(t)$ is also defined as follows.

$$G(t) = \frac{1}{2}\,\{G_{11}(t) - G_{12}(t)\} \tag{7}$$

For uniaxial stress histories ($\sigma_{22}(t) = \sigma_{12}(t) = 0$), the Laplace transform of Eq. (2) gives

$$\bar{\sigma}_{11} = s\bar{G}_{11}\bar{e}_{11} + s\bar{G}_{12}\bar{e}_{22} \tag{8.a}$$

$$\bar{\sigma}_{22} = s\bar{G}_{12}\bar{e}_{11} + s\bar{G}_{11}\bar{e}_{22} = 0 \tag{8.b}$$

In the case of uniaxial stress relaxation ($e_{11}(t) = \mathring{e}_{11}U(t)$), substituting the conditions to Eq. (8) and evaluating the inverse Laplace transforms yield

$$\sigma_{11} = E(t)\,\mathring{e}_{11} \tag{9}$$

where $E(t)$ is the uniaxial relaxation function which is related to $G_{11}(t)$ and $G_{12}(t)$ by

$$\bar{E}(s) = \bar{G}_{11}(s) - \frac{\{\bar{G}_{12}(s)\}^2}{\bar{G}_{11}(s)} \tag{10}$$

In this case, Eq. (8) gives the following expression for the Poisson's ratio $\nu^r(t)$ (= $-e_{22}(t)/\mathring{e}_{11}$).

$$\bar{\nu}^r(s) = \frac{\bar{G}_{12}(s)}{s\bar{G}_{11}(s)} \tag{11}$$

In the case of uniaxial extension at a constant strain rate ($e_{11}(t) = \kappa\, tU(t)$, κ : the rate of strain), the Poisson's ratio $\nu^u(t)$ (= $-e_{22}(t)/\kappa\, t$) can be expressed from Eq. (8) as

$$\nu^u(t) = \frac{1}{t}L^{-1}\left\{\frac{\bar{G}_{12}(s)}{s^2\bar{G}_{11}(s)}\right\} \tag{12}$$

where $L^{-1}\{\ \}$ denotes the inverse Laplace transform.

In order to calculate E(t), $\nu^r(t)$ and $\nu^u(t)$ from the functions $G_{11}(t)$ and $G_{12}(t)$, it is required to evaluate the Laplace transforms and the inversions in Eqs. (10), (11) and (12). To this end, Christensen et al. (1971) assumed $G_{ij}(t)$ can be expressed in the following form.

$$G_{ij}(t) = \mathring{G}_{ij} + \sum_{k=1}^{n} G_{ij}^{k} \exp\left\{-\frac{t}{\tau_k}\right\} \tag{13}$$

where $\mathring{G}_{ij}$, G_{ij}^k and τ_k are the constants which are to be determined experimentally. Equation (13) corresponds to the discrete spectral representation of the relaxation functions. The Laplace transform of Eq. (13) is given by

$$\overline{G}_{ij}(s) = \frac{\mathring{G}_{ij}}{s} + \sum_{k=1}^{n} \frac{G_{ij}^{k}}{s + \tau_k^{-1}} \tag{14}$$

Substituting Eq. (14) into Eqs. (10), (11) and (12), we finally obtain the expression for E(t), $\nu^r(t)$ and $\nu^u(t)$ in the form $L^{-1}\{f(s)/g(s)\}$, where f(s) and g(s) are polynomials in s. The inverse transform of f(s)/g(s) is easily accomplished through the use of residue theory (Derrick 1972).

Experimental

Laboratory handsheets were prepared according to JIS P-8209 from spruce bleached kraft pulps beaten in a PFI mill. Basic properties of handsheets are shown in Table 1.

The in-plane relaxation functions $G_{11}(t)$ and $G_{12}(t)$ were determined from a strip biaxial stress relaxation test by use of a biaxial tensile tester (Uesaka 1979b). The conditions of this test are given by

$$e_{11}(t) = \mathring{e}_{11}\, U(t)\,,\; \mathring{e}_{22}(t) = \gamma_{12}(t) = 0 \tag{15}$$

Table 1. Basic properties of samples

Sample	C. S. F.[a] ml	Basis weight g/m^2	Apparent density g/m^3
R-0	687	67.2	0.443
R-3	574	63.8	0.580
R-9	334	63.5	0.639
R-20	87	58.9	0.672

[a] Canadian Standard Freeness

Substituting Eq. (15) into Eq. (2), we can determine $G_{11}(t)$ and $G_{12}(t)$ by the following equations:

$$G_{11}(t)=\frac{\sigma_{11}(t)}{\overset{\circ}{e}_{11}}\,,\quad G_{12}(t)=\frac{\sigma_{22}(t)}{\overset{\circ}{e}_{11}} \tag{16}$$

where $\overset{\circ}{e}_{11}$ is the applied initial strain which was taken as about 1.7×10^{-3} in this experiment. Measurements related to the prediction of E(t), $\nu^r(t)$ and $\nu^u(t)$ were made at 21 °C and 43% R.H. and those of K(t) and G(t) at 23 °C and 57 % R.H.. Plots are the mean values for three to five specimens. For a description of the testing apparatus and the procedures, the reader is referred to the earlier papers (Uesaka 1979a, b).

Results and Discussion

Uniaxial Relaxation Function E(t) and Poisson's Ratios $\nu^r(t)$ and $\nu^u(t)$

Figure 1 shows the in-plane relaxation fuctions $G_{11}(t)$ and $G_{12}(t)$ obtained from the strip biaxial stress relaxation data for the sample R-3 in Table 1. The solid lines re-

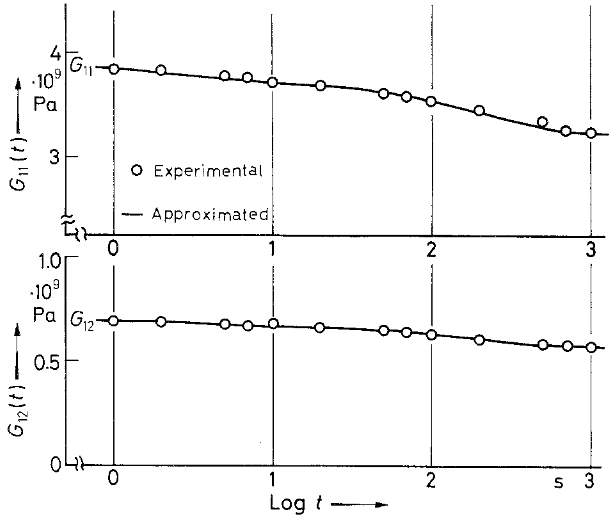

Fig. 1. In-plane relaxation functions $G_{11}(t)$ and $G_{12}(t)$ for sample (R-3)

Table 2. Constants of relaxation functions $G_{ij}(t)$

k	τ_k (s)	G_{11}^k (x 10^9 Pa)	G_{12}^k (x 10^9 Pa)
0	–	3.194	0.578
1	2	0.174	0.031
2	20	0.009	0.007
3	200	0.490	0.094

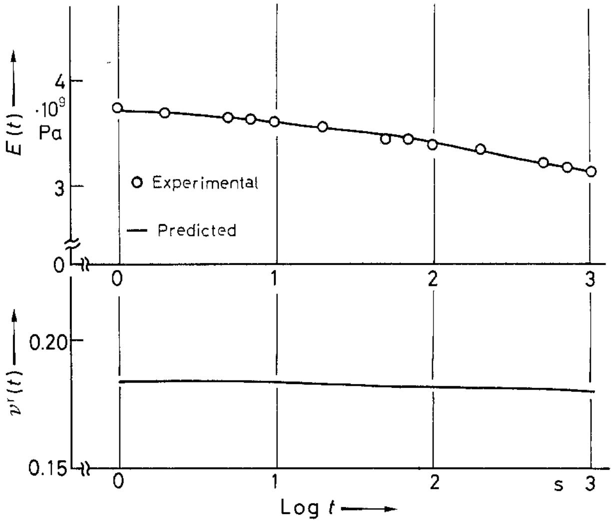

Fig. 2. Uniaxial relaxation function E(t) and Poisson's ratio $\nu^r(t)$

present the curves for the functions approximated by Eq. (13) with G_{ij}^k and τ_k given in Table 2. The curves fitting procedure employed is the systematic one proposed by Gradowczyk and Moavenzadeh (1969), which assures the positiveness of the constants G_{ij}^k. As can be seen in Fig. 1, the approximated curves are in fair agreement with the experimental values over the range of times investigated.

Figure 2 shows the results for the relaxation function E(t) and the Poisson's ratio $\nu^r(t)$ in uniaxial stress relaxation. The curve for E(t) predicted from $G_{11}(t)$ and $G_{12}(t)$ (solid line) agrees very closely with the experimental values (circles) obtained from the ordinary uniaxial stress relaxation test. The predicted Poisson's ratio $\nu^r(t)$ shows a slight decrease with increasing time, but in practice it may be regarded as constant.

Figure 3 shows the result for the Poisson's ratio $\nu^u(t)$ in uniaxial constant strain-rate extension. The ratio $\nu^u(t)$ slightly decreases with straining time. This indicates

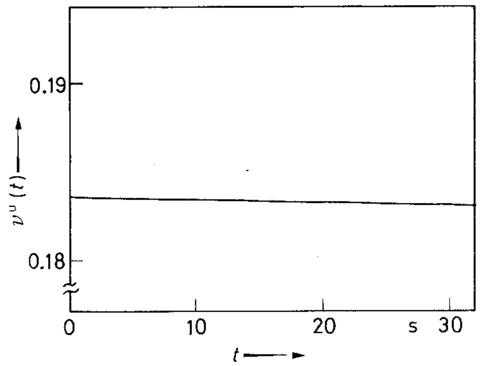

Fig. 3. Poisson's ratio $\nu^u(t)$ in constant strain-rate extension

that with increasing strain rate the ratio $\nu^u(t)$ slightly increases within a linear viscoelastic range in the uniaxial tensile test.

In elasticity problems the Poisson's ratio is a constant depending only on the nature of the material, while in viscoelasticity problems it depends not only on the material but also on the stress or strain histories. Takemura (1976) reported that the Poisson's ratio for wood under constant load condition was a complex function of time, depending on the magnitude and applied direction of load. In order to characterize the Poisson's ratio from a viscoelastic point of view, we shall now consider the material, as a reference system, having the in-plane relaxation functions in the following form (Fig. 4):

$$\begin{aligned} G_{11}(t) &= \mathring{G}_a + G_a \exp\left\{-\frac{t}{\tau_a}\right\} \\ G_{12}(t) &= \mathring{G}_b + G_b \exp\left\{-\frac{t}{\tau_b}\right\} \end{aligned} \tag{17}$$

where G_a and G_b are the relaxation parts of $G_{11}(t)$ and $G_{12}(t)$, respectively, and $\mathring{G}_a$ and $\mathring{G}_b$ are the equilibrium parts (Fig. 4). To simplify the problem we take $\tau_a = \tau_b = \tau_0$. In this system the Poisson's ratio $\nu^r(t)$ can be calculated by substituting Eq. (17) into (11) and evaluating the inverse Laplace transform, as follows.

$$\nu^r(t) = p + q \exp\{-\beta t\} \tag{18}$$

where

$$p = \frac{\mathring{G}_b}{\mathring{G}_a}, \quad q = \frac{\mathring{G}_a G_b - G_a \mathring{G}_b}{\mathring{G}_a(\mathring{G}_a + G_a)} \quad \text{and} \quad \beta = \frac{\mathring{G}_a}{\mathring{G}_a + G_a}\,\frac{1}{\tau_0} \tag{19}$$

Since $\beta > 0$, $\nu^r(t)$ decreases or increases depending upon whether $q > 0$ or $q < 0$. When $q = 0$, then is,

$$\frac{G_a}{\mathring{G}_a} = \frac{G_b}{\mathring{G}_b} \tag{20}$$

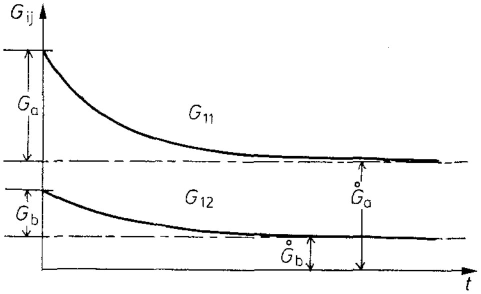

Fig. 4. In-plane relaxation functions $G_{11}(t)$ and $G_{12}(t)$ given in Eq. (17)

the ratio $\nu^r(t)$ becomes independent of time. The similar relation can be found for the ratio $\nu^u(t)$. Thus the time dependence of the Poisson's ratios in this system is largely affected by the ratios $G_a/\mathring{G}_a$ and $G_b/\mathring{G}_b$. For a more general system such as real paper sheets, Poisson's ratios will depend also on the relaxation-time distribution in a complex manner.

Relaxation Function in Areal Dilatation K(t) and Relaxation Function in Shear G(t)

Figures 5 and 6 show the relaxation functions in areal dilatation K(t) and in shear G(t) calculated from $G_{11}(t)$ and $G_{12}(t)$ using Eqs. (6) and (7), where these are expressed as the specific form (that is, the relaxation function devided by density) for comparison

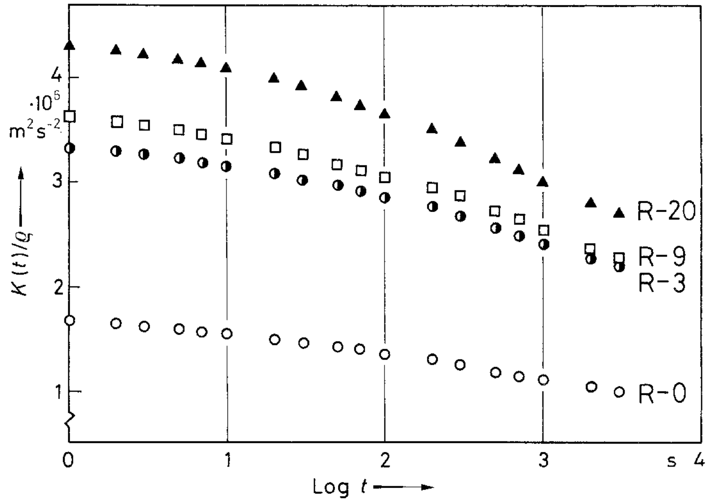

Fig. 5. Relaxation function K(t) in areal dilatation

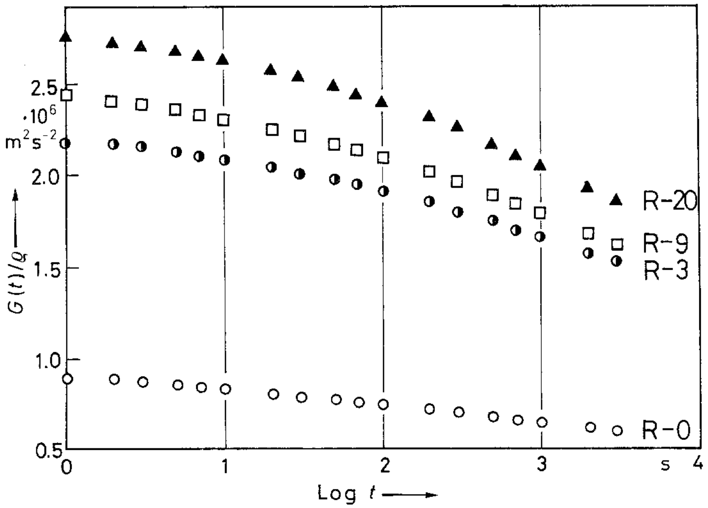

Fig. 6. Relaxation function G(t) in shear

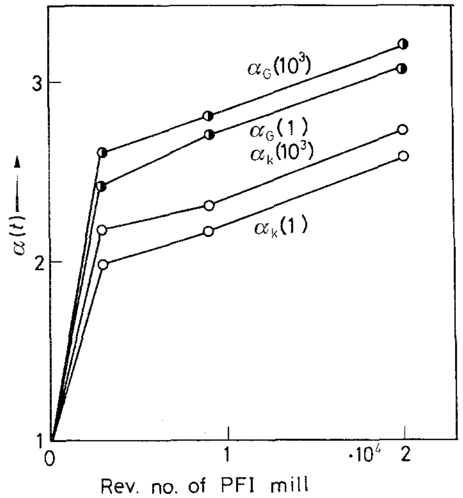

Fig. 7. Effect of beating on isochronous relaxation functions

of samples with different degree of beating (Uesaka 1979c). Both relaxation functions increase with the beating as expected. In Figure 7, the isochronous relaxation function values at t = 1 and 10^3 sec. are plotted against a PFI mill revolution number as an indication of beating degree. The symbol $\alpha_K(t)$ means the ratio of K(t) for the beaten sample to that for the unbeaten one, and $\alpha_G(t)$ is defined in a similar way. Over this beating range the values of $\alpha_G(t)$ are higher than those of $\alpha_K(t)$; this result is consistent with the elastic property data (Uesaka 1979c). In this figure it is interesting to note that both ratios at 10^3 show the higher values than those at 1 s. To clarify further such time dependence of these ratios, we shall introduce the relaxation spectra for K(t) and G(t) as in the one-dimensional theory (Gross 1953).

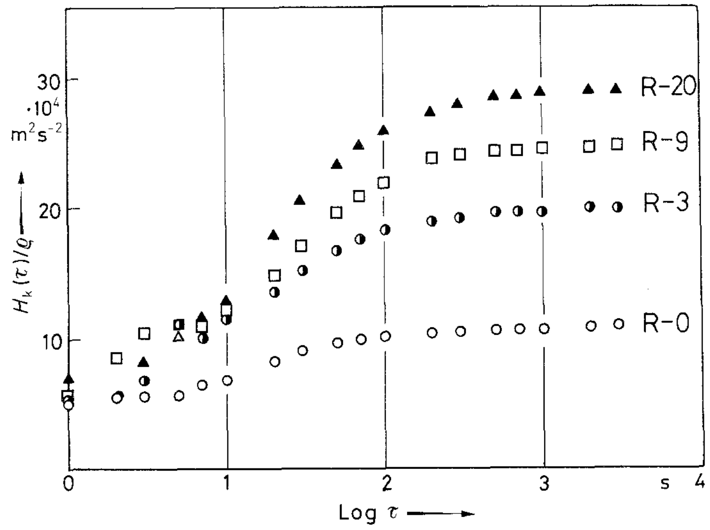

Fig. 8. Effect of beating on relaxation spectrum $H_K(\tau)$

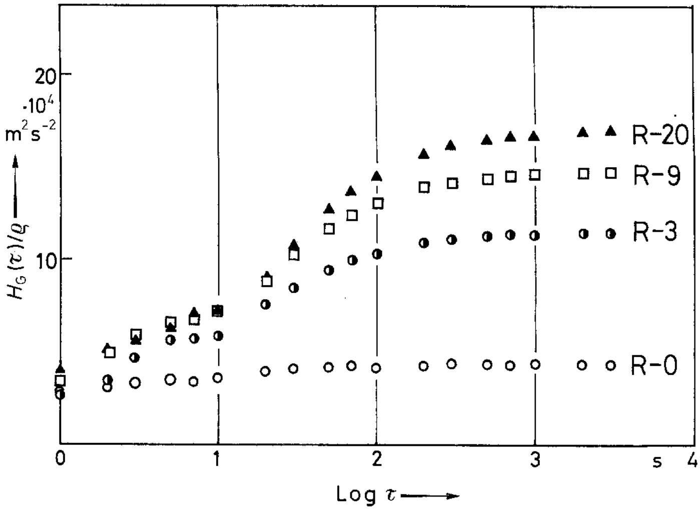

Fig. 9. Effect of beating on relaxation spectrum $H_g(\tau)$

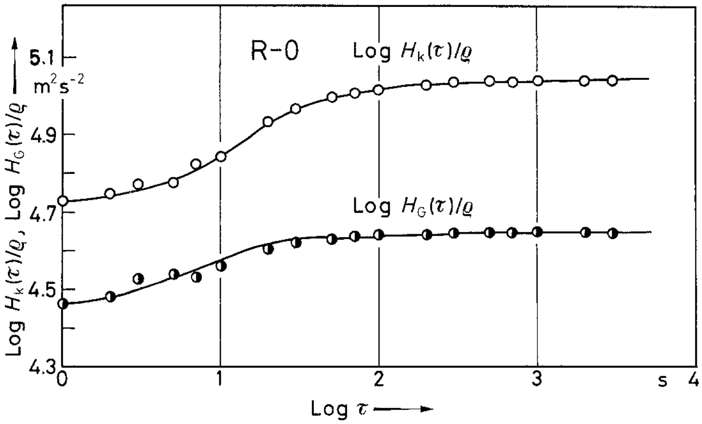

Fig. 10. Comparison between $H_k(\tau)$ and $H_g(\tau)$ for sample (R-0)

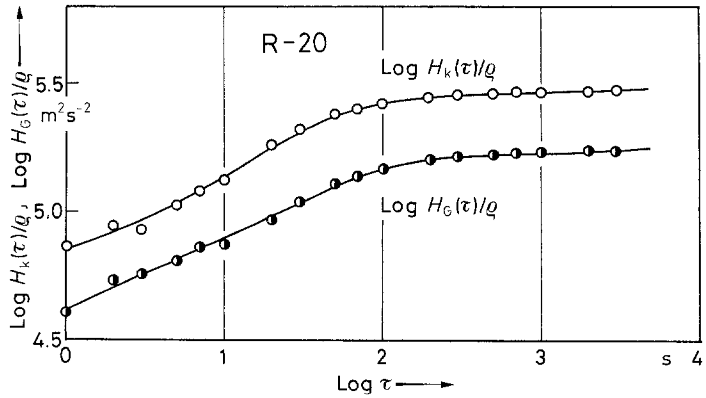

Fig. 11. Comparison between $H_k(\tau)$ and $H_g(\tau)$ for sample (R-20)

Figures 8 and 9 show the relaxation spectra $H_k(\tau)$ and $H_g(\tau)$ calculated from the Alfrey's first order approximation formula (Ferry 1960). As the beating proceeds, the spectral intensities markedly increase, especially at longer relaxation times. This implies that beating primarily affects the relaxation process with longer relaxation time. Comparisons between $H_k(\tau)$ and $H_g(\tau)$ for the unbeaten sample (R-0) and the beaten sample (R-20) are presented in Figures 10 and 11. For the unbeaten specimen the relaxation function G(t) has lower spectral intensity than K(t), particularly at longer relaxation times, while for the beaten one both curves show a very similar shape over the time range investigated. This similarity means that the following approximate relation between $H_k(\tau)$ and $H_g(\tau)$ holds for the beaten sample:

$$H_k(\tau) \doteqdot \eta\, H_g(\tau) \tag{21}$$

where log η is the longitudinal shifting factor of the curves in Figure 11.

The results described above will characterize beating effects from a linear viscoelastic aspect and also provide the basis for further structual considerations.

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