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Damping behaviour of polymeric materials subjected to longitudinal loads

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The present paper reports on measurements of shear modulus and damping in a torsion pendulum on polymeric materials and paper. The experiments were carried out in a torsion device which permitted an axial load to be applied to the sample. At low loads, apparent changes in modulus and damping were recorded. At higher loads, under which marked creep occurred, the damping increased at the moment of loading and then decreased slowly, eventually approaching an equilibrium value. After release of the load, a transient decrease in damping was observed, the damping value of the origin sample being recovered after some time. During the creep and recovery processes, the modulus remained constant.

1. Introduction

Dynamic measurements of elasticity and damping have been used extensively in polymer science for studying transitions and thus providing information regarding the structure. The results of this technique have been reviewed by Ward [1] and Nielsen [2]. However, the damping behaviour during a creep process has not been given any more attention, although this probably can provide new information about the deformation process. Some results are given by Nägerl [3] who reports that the loss factor of plasticized poly(vinyl chloride) decreases with time. Hoffmann [4] examined both theoretically and experimentally the influence of an axial load on the damping behaviour of metallic strings in the linear region and found, for lower loads, an almost linear increase in the shear modulus with time. Within metal science the internal friction concept, which is equivalent to the damping measure, is now regarded to be a useful tool for studying structural changes (e.g. Snoek damping). For example, it has been found that the internal friction depends on the amount of plastic strain and in some cases also that it recovers after removal of the applied load [5, 6]. The internal friction concept and its relation to movement of point defects, dislocations etc. have been reviewed by De Batist [7]. Recently Crissman and Zapas [8] reported on the damping behaviour of some polyethylenes during creep to failure. They found that tan δ went through a minimum at strain levels approximately corresponding to the onset of local instability. The rise in damping at larger strains was suggested to be due to an increasing defect concentration within the crystallites.

The present work reports on the behaviour of the complex modulus, determined in a torsion pendulum, in the elastic region and the region where marked flow is taking place. In the first region an apparent variation in real modulus and damping due to the influence of the applied load on the restoring torque is noticed. This effect is momentary and reversible, its extent being determined by the geometry of the samples. This apparent variation of the modulus in the elastic region can be predicted from theory, while during creep marked deviations are observed. The damping always increased when the creep load was applied or removed. The damping value then decreased gradually approaching the value characteristic of the undeformed state. In contrast to this, there was no significant change in real modulus during the flow process.

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2. Theory

In the elastic region, the influence of the axially applied load on the frequency and damping is determined by the geometry of the samples. For samples with cylindrical cross-sections the stress has no influence on either the frequency of damping as long as there are no structural changes in the sample [9]. However, for samples with a rectangular cross-section an increase in frequency and a decrease in damping due to the applied load is observed [10, 11].

The following analysis relates to an inverted pendulum, where the oscillating system consists of the sample supported by a thin wire and a mass attached to the upper end of the sample [9]. The axial load is applied via a balance beam.

For stress-free samples, the following equation relates the torque per unit angle, M', with the frequency of the whole oscillating system, ω_c.

$$M' = I\omega_c^2/F \quad (1)$$

where I is the moment of inertia and F is given by

$$F = \omega_c^2/(\omega_c^2 - \omega_1^2) \quad (2)$$

with ω_1 being the frequency without the sample.

The real part of the total torque per unit angle can be divided in two parts, one depending on the shear modulus, and the other on the axial stress acting on the sample. The real part of the shear modulus, G', is related to M' by

$$G' = aM' \quad (3)$$

where a is dependent on the geometry of the samples. For samples with the length l and with a rectangular cross-section (width b and thickness h) a is given by

$$a = 3l/bh^3. \quad (4)$$

The total real modulus can now be written as [4, 6]

$$G' = G'(\sigma = 0) + G'(\sigma) \quad (5)$$

The first term on the right is

$$G'(\sigma = 0) = aI\omega_c^2/F \quad (6)$$

and the second (i.e. contribution from the axial stress)

$$G'(\sigma) = ab^2h\sigma/12l = (b^2/4h^2)\sigma \quad (7)$$

where σ is the applied stress. If $h/b \ll 1$ the contribution G' thus may become very large. From a physical point of view, however, only $G'(\sigma = 0)$ reflects the properties of the material.

For stress-free samples, the imaginary part of the modulus, G'', is related to the logarithmic decrement, Λ, in the following way

$$G'' = aI\omega_c^2\Lambda/\pi. \quad (8)$$

According to [12] G'' can be expected to remain constant, i.e. independent of the applied axial stress, as the stress is supposed to influence the real modulus, G', only. Evidently, the frequency ω_c^2 and the logarithmic decrement Λ must be inversely related. The above equations imply further a proportionality between ω_c^2 and σ. The decrement thus decreases with increasing σ to render the product $\omega_c^2\Lambda$ constant. The true value of Λ is obtained by extrapolating the measured values to $\sigma = 0$.

As with the decrement, the loss tangent will apparently change with σ. This can be seen directly from the relation

$$\tan\delta = G''/G'.$$

The above analysis only refers to the elastic region. During creep, when structural changes can be produced, the dynamic behaviour becomes more complex as described below. Furthermore no absolute value of the damping can be obtained since the above elastic analysis is not valid in this region.

3. Experimental

For the measurements reported in this work two different inverted-type torsion pendulum devices, which have been described elsewhere [10], were used. They were designed for an axial load of 0 to 300 g and up to 5 kg, respectively. The experiments were carried out at atmospheric pressure, 65% relative humidity and 20 ± 0.1° C. The maximum deformation of the samples due to the torsional strain was of the order 10^{-4}.

4. Materials

The experiments were performed using polymer and paper samples having a length of 150 mm and a width of 15 mm. The following materials were used:

(a) Low density polyethylene (LDPE) density 0.920 g cm^{-3} melt index 2.0 g/10 min (MFI 190/2), $\bar{M}_v = 9.0 \times 10^4$, thickness 0.046 mm or 0.45 mm.

(b) Rubber hydrochloride, density 1.12 g cm^{-3}, $\bar{M}_v = 2.4 \times 10^5$, thickness 0.038 mm.

(c) Cellulose film, glycerol content 19%, DP ~290, $E_{\parallel}/E_{\perp} = 1.8$, moisture content at 65% r.h. 16.4%, thickness 0.043 mm.

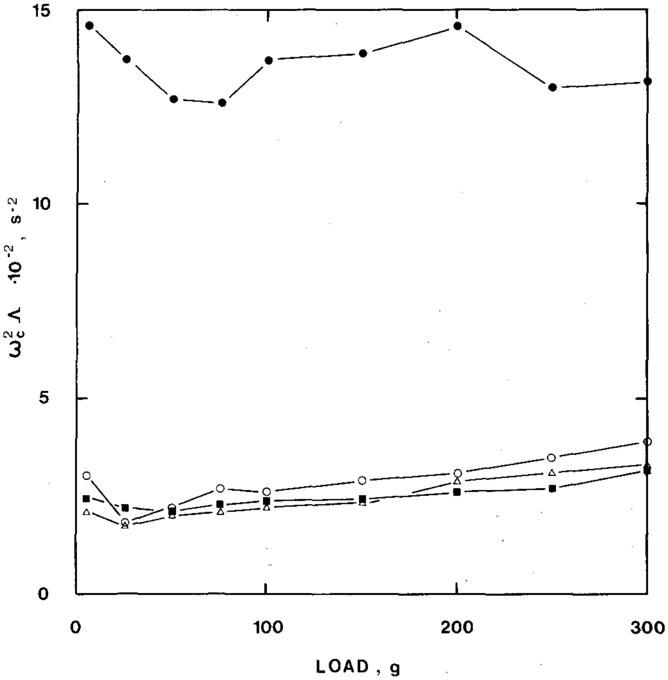

Figure 1 The product $\omega_c^2\Lambda$ versus the applied load (elastic region) for (■) LDPE (stress range 0.01 to 0.44 MPa), (▿) rubber hydrochloride (0.09 to 5.26 MPa), (○) cellulose film (0.08 to 4.65 MPa), and (●) kraft paper (0.02 to 1.35 MPa).

(d) MG kraft paper, density 0.70 g cm^{-3}, $E_{\parallel}/E_{\perp} = 1.6$, weight average fibre length 2.22 mm, moisture content at 65% r.h. 13.0%, thickness 0.148 mm.

$E_{\parallel}$ and $E_{\perp}$ refers to the elastic modulus in the machine (MD) and cross direction (CD), respectively.

5. Results

5.1. Damping behaviour under small axial stresses (elastic region)

In Fig. 1 the product $\omega_c^2\Lambda$ is plotted versus the axial load for different materials. As predicted in the preceding section this quantity is fairly constant for small loads. Only for the highest stresses can a deviation from this constancy be observed. These deviations are probably due to the onset of flow processes in the material.

5.2. Damping behaviour under high stresses

The experiments reported on in the preceding section relate to the elastic region. When the stress level was raised substantially, creep effects became very pronounced. In this case the axial stress was chosen to be about 90% of the rupture stress, giving a creep rupture time of 1 to 2 days. The first measurements of the logarithmic decrement was made a few minutes after application of the load.

For all investigated materials it was found that the frequency of oscillation and hence the real modulus G' was constant and independent of the creep deformation.

During the creep process the damping decreased with the elapsed time. In Fig. 2 this behaviour is illustrated for LDPE together with the corresponding creep curve. The logarithmic decrement Λ

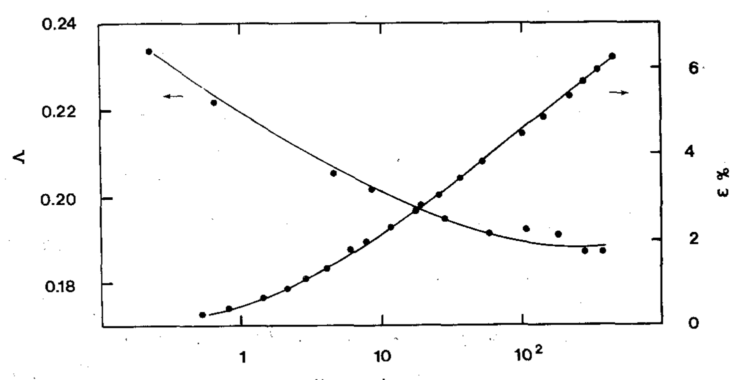

Figure 2 The logarithmic decrement and the creep deformation for LDPE versus time under load. Load 1300 g (13 N), thickness 0.45 mm.

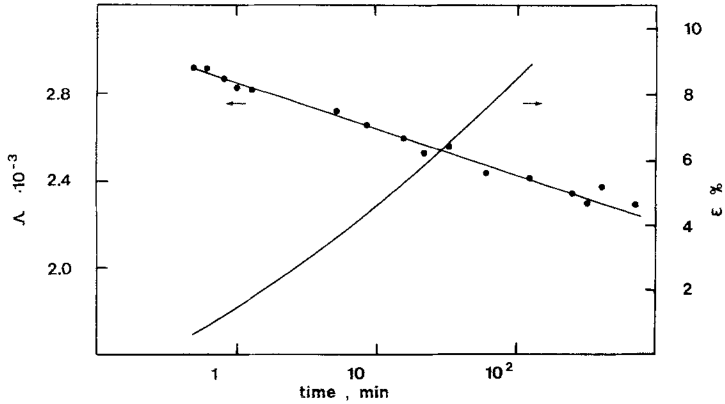

Figure 3 The logarithmic decrement and the creep deformation for rubber hydrochloride against time under load. Load 100 g (10 N).

decreases by 20 to 25% in a few hours and then approaches, asymptotically, a constant value. A similar behaviour was registered for cellulose, rubber hydrochloride (Fig. 3), and paper (Fig. 4). No significant differences were observed in the Λ(time)-variation between paper samples cut in the MD- and CD-directions, although the magnitude of Λ at a given time was not the same.

It may be remarked that the stress was not corrected for the sample elongation, but considering the rather small deformations occurring during creep the error introduced is negligible [13]. No correlation between the magnitude of the decrease in damping and total creep deformation was observed.

5.3. Damping recovery in paper

The time-dependent decrease in damping of paper was reversible. The paper samples were subjected to a load corresponding to about 80% of their rupture stress for 1 minute. After removal of the load the damping was registered as a function of the recovery time. Since the measurements were performed without loading, the recovery process could be expressed in terms of tan δ.

Fig. 5 shows the relation between tan δ and time for kraft paper in the CD-direction. The damping reaches its maximum value immediately after unloading and then returns to its original state within 1 day.

With LDPE and rubber hydrochloride similar behaviour of the damping during recovery was observed, although the effects were not sufficiently pronounced to permit accurate measurements with the present method.

6. Final remarks

The remarks obtained above show that there are two types of change in the dynamic characteristics of strip-shaped samples subjected to an axial load in a torsion pendulum. Firstly, the modulus apparently increases with the load, the amount of increase depending on the geometry of the sample. The damping on the other hand decreases. This applies to small loads, i.e. in the elastic region. At higher loads, where creep effects are pronounced, i.e. where structural changes are taking place, an additional decrease in damping, for constant stress load, with time is observed.

As evident from Figs. 2, 3, and 4 the decay in damping during creep is not exponential but rather more logarithmic extending over a few

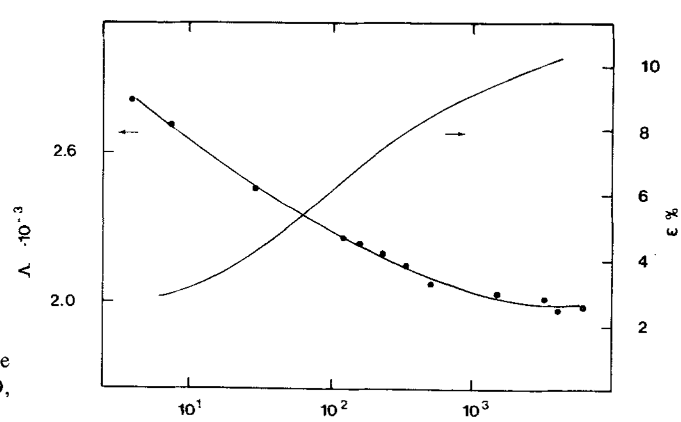

Figure 4 The logarithmic decrement and the creep deformation for MG kraft paper, CD, against time under load. Load 4100 g (41 N).

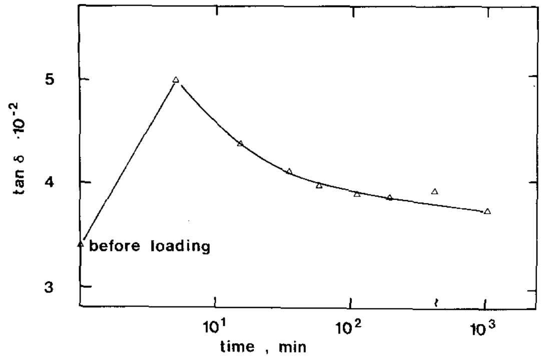

Figure 5 Tan δ plotted against time after loading (3500 g, 35 N) for 1 min to 80% of the rupture load. MG kraft paper, CD.

decades of time. The same behaviour applies to recovery; when the load is removed an increase in damping is noted, then the damping decreases logarithmically with time to its virgin value. A straight correlation of the decrease in damping with the creep deformation was not noted. For metals, similar effects have been studied under plastic flow by Feltham [14] and a linear increase in internal friction with the tensile strain rate was found. A relation of this kind does not seem to apply for polymeric materials.

The time dependence of the logarithmic decrement exhibits strong similarities with the kinetics of other flow processes in solids; e.g. stress relaxation [15], although the results do not permit an exact evaluation of the kinetics involved. Like other flow processes it is tempting to interpret the decrease in damping with time in terms of cooperative effects between the flow units (e.g. structural defects) responsible for the creep process [16]. The movement of coupled flow units, as reflected in the damping value, is a consequence of the creep or recovery process and not of the deformation due to the oscillations of the pendulum. The flow processes, i.e. movements of flow units, taking place in the sample thus give rise to additional damping simply because every irreversible movement in the stress field of the torsional oscillations must lead to energy dissipation. The number of defects or other structural parts may remain unchanged, the important thing being their movement. This is also in agreement with the ideas concerning similar processes in metals [14].

It is at this point difficult to explain the detailed mechanisms of the change in damping with strain. This difficulty is also recognized by Crissman and Zapas [18] who correlated the observed minimum in tan δ during creep with the onset of local instability, but in their work no explanation for the initial decrease in damping was provided. For crystalline solids similar effects have been associated with work hardening [14].

Some creep experiments for paper were continued to rupture but no anomalous damping changes were found in the time period close to the creep rupture. Such changes could be expected if, during that period, there were excessive rearrangements of the paper structure, breaking of bonds and similar loss-causing effects. Thus bond breaking does not seem to be the main flow mechanism in paper.

In a future paper, measurements of the damping during creep at different temperatures for a number of thermoplastics, giving more insight into the mechanisms of the strain dependence of tan δ, will be presented.

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