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A VISCOELASTIC FLUID APPROACH TO DEFORMATION AND YIELD OF GLASSY POLYMERS

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In this study a mathematical model is presented that covers the deformation behaviour of a glassy polymer from the linear viscoelastic range up to yield. In this approach the material is regarded as a viscoelastic fluid whose relaxation times are decreased under the influence of stress. The features of the model are demonstrated on a commercial grade of Polycarbonate.

Introduction

With respect to modelling of the deformation behaviour, glassy polymers are usually regarded as viscoelastic solids. A prime example being the approach to yield of these materials, using yield criteria of which the pressure modified Von Mises criterion appears to be the most successful. This "critical" stress approach inherently assumes that no irreversible "flow" deformation will occur below a certain stress limit, and hence a new deformation mechanism is activated above this level.

In the past decade several attempts have been made to come to an alternative approach to the deformation behaviour of polymeric materials [1-2]. In these models the yield behaviour is described using a single Maxwell element with a strongly stress dependent viscosity. In constant strain rate conditions, this results in a sharp transition from solid-like to fluid-like behaviour, almost identical to an elastic-perfectly plastic response employing a rate-dependent Von Mises criterion. At intermediate stress levels, however, the use of a single stress dependent relaxation time can not account for the multi-relaxation time viscoelastic response that is generally observed in glassy polymers. In this study a similar model is presented that can be combined into a "multi-relaxation time" expression that is also able to describe the spectrum of relaxation times required to describe the nonlinear behaviour at intermediate stress.

Basic approach: Time-stress superposition

In the case of a viscoelastic fluid the linear viscoelastic compliance $D(t)$ may be represented by a generalised Kelvin-Voigt model:

$$D(t) = D_0 + \sum_{i=1}^n D_i \exp\left(-\frac{t}{\tau_i}\right) + \frac{t}{\eta_0} \quad (1)$$

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The prime difference with a viscoelastic solid being the presence of a flow-term, represented here by the viscosity η_0 . In the constitutive approach stress is assumed to influence all retardation times in the same way, which, similar to time-temperature superposition, leads to time-stress superposition, in fact a simplification of the well known Schapery model [3]:

$$\varepsilon(t, \sigma) = \int_0^t D(\Psi - \Psi') \dot{\sigma}(t') dt' \quad (2)$$

with the reduced time variables defined as:

$$\Psi = \int_0^t \frac{dt''}{a_\sigma(\sigma)} ; \Psi' = \int_0^{t'} \frac{dt''}{a_\sigma(\sigma)} ; \quad (3)$$

where a_σ is a stress dependent shift function. In the case of stress activation according to Eyring it can be expressed as:

$$a_\sigma(\sigma) = \frac{\sigma v}{kT} \frac{1}{\sinh\left(\frac{\sigma v}{kT}\right)} \quad (4)$$

where v is the activation volume, and k Boltzmann's constant.

For very low stress levels ($\sigma < kT/v$) $a_\sigma = 1$, and the model reduces to the classic expression for linear viscoelasticity. With increasing stress the shift function a_σ decreases, which leads nonlinear viscoelastic behaviour. At very high stress levels the material behaviour reduces to that of a nonlinear fluid, which displays the strain rate dependence of an Eyring dashpot that is typically observed in the yield behaviour of many glassy polymers. To be able to describe the post-yield behaviour a 3-D finite strain formulation was developed [4].

Experimental

All experiments were performed on injection moulded tensile bars (ISO R527) produced from a commercial grade of Polycarbonate (Lexan 161R, GE). Each of the experiments was performed on a new sample. All test samples had the same age, which exceeded by far the longest relaxation time in the experiments.

Results

To visualise the influence of stress the material was studied using creep tests at various stress levels (Fig 1). The curves were superimposed to the master curve shown in Fig.2 employing the shift function a_σ mentioned above (Eq.4), yielding an activation volume v of 2.96 nm³. At this point it must be emphasised that the master curve in Fig. 2 is a "virtual" curve that, due to physical ageing effects, is very likely to deviate strongly from an experimental creep curve on the same time scale. Also the experimental time range covered in Fig. 1 (3 decades) does not seem to justify the enormous extrapolation in time span. On the other hand, however, it seems encouraging that

the apparent shift of the curves correlates very well with Eq. 4.

The master curve was fitted using a generalised Kelvin-Voigt model with 14 retardation times. The prediction of nonlinear creep is also shown in Fig 1. It is shown that application of time-stress superposition to the master curve in Fig.2 yields a good description of the short term creep behaviour of polycarbonate. Also the tensile behaviour at various strain rates (Fig.3), seems to be represented quite well by the model. It can therefore be concluded that, within the range of experiments performed here, the phenomenological constitutive model presented here seems a promising approach.

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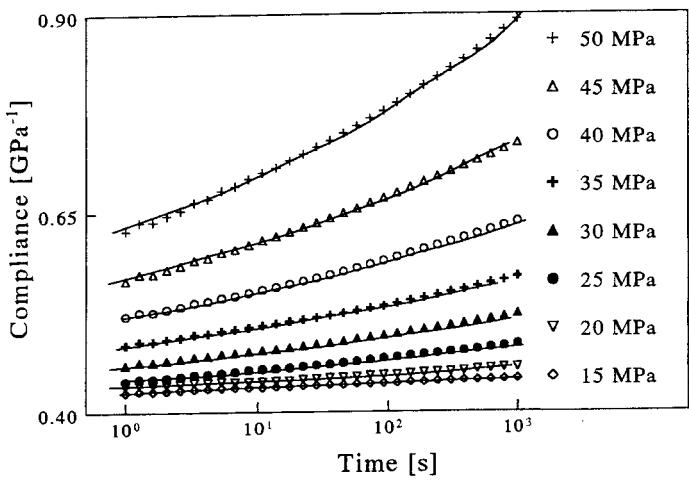


Figure 1 Creep compliance of polycarbonate at 22.5 °C and various stress levels (symbols) compared to model predictions (solid lines).

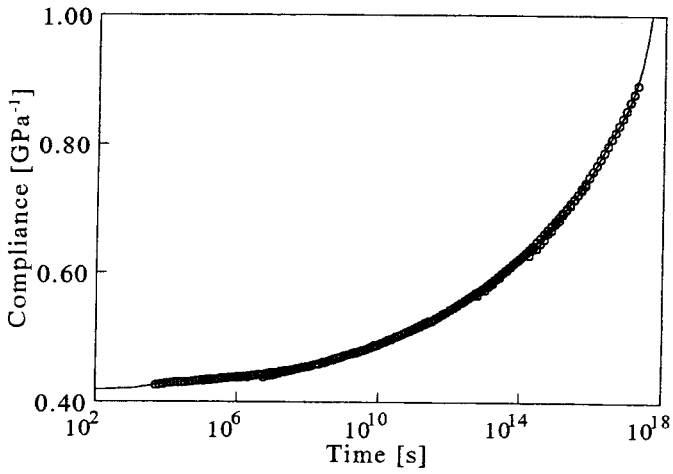


Figure 2. Master curve of the linear compliance obtained from the data in Figure 1

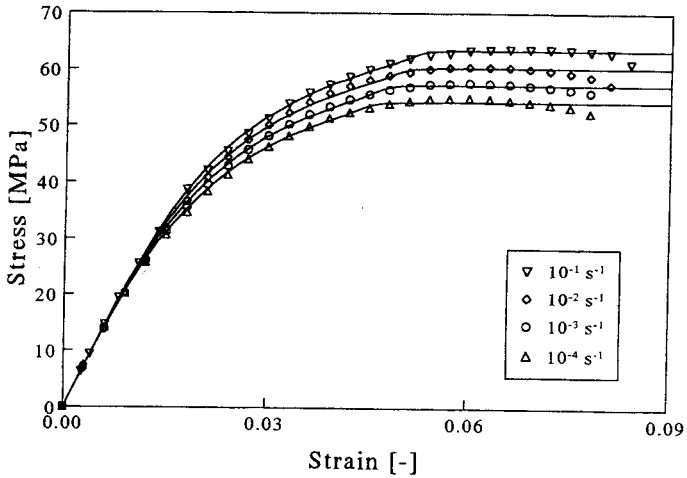


Figure 3 Tensile tests at various strain rates (symbols), compared to model predictions (solid lines)