

# Report

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**Modelling the Long-term  
Deformation Behaviour of  
Polymers for Finite Element  
Analysis**

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## Modelling the Long-term Deformation Behaviour of Polymers for Finite Element Analysis

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Industry and Innovation Division

### SUMMARY

Creep studies have been carried out under uniaxial tension and uniaxial compression on poly(oxymethylene). Measurements under tension have been modelled using a function with four material parameters. One of these parameters, related to a mean retardation time for the relaxation process responsible for creep, decreases with increasing stress, and this gives rise to non-linear creep behaviour. Measurements of creep under compression indicate that the retardation time parameter is determined by the stress state as well as the stress magnitude. A proposed relationship for this parameter has enabled the description of non-linear creep under a general, multi-axial stress.

A theoretical extension of the model has been proposed that relates stresses and strains under situations where the stress is not constant but varies with time. This is necessary for the model to be implemented in a finite element system to carry out design calculations that take proper account of the time-dependent properties of polymers. The validity of the theory is evaluated through the use of the creep model to predict deformation for a variety of simple stress and strain histories under uniaxial tension. These include increasing and decreasing stress in step changes, constant strain (stress relaxation) and constant deformation rate. Predictions are compared with experimental measurements. The close agreement between calculation and measurement supports the development of code, in future work, to obtain solutions for multi-axial stresses and strains using a finite element system.

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Approved on behalf of the Managing Director, NPL,  
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## 1 INTRODUCTION

Polymeric materials are increasingly being used in the manufacture of components where a knowledge of deformation of the component under long-term loading is needed for confident design. Finite element analysis is a powerful design tool and is used to calculate stress and strain distributions within a component under service loads as well as overall changes in dimensions. The accuracy of results produced by a stress analysis depends on a number of factors including the validity of the materials model used to describe deformation behaviour. Polymeric materials are viscoelastic, so properties change with time under load as a result of relaxation processes in the molecular structure. Creep tests are generally used to characterise the time-dependent properties of polymers. Here, a constant stress is applied to a specimen, and the strain is measured with time under load. Creep models are used to relate measurements of creep compliance to time under load and material parameters. The stress magnitude also needs to be included in the model if properties are non-linear. For such a model to be used in a stress analysis under long-term loading, a theoretical framework is needed that relates stresses and strains for situations where the stress is not constant but where some more general stress or strain history occurs, including multi-axial stress and strain states. The theory for linear behaviour is well developed in terms of the Boltzmann superposition integral [1]. Here, changes in strain, resulting from incremental changes in stress that define the stress history, are obtained by adding creep compliance contributions from each increment of stress that have been retarded by the time delay in the application of that increment. This approach cannot be used, without modification, when behaviour is non-linear since the superposition procedure for incremental changes is then invalid.

In finite element systems, creep models for plastics generally represent the material as a series of elastic springs and viscous dashpots [1]. One problem with the application of such models is that a large number of spring and dashpot elements is needed to describe the change of properties with time over a long timescale since the relaxation time spectrum of molecular motions making up a relaxation process is usually very broad. A large number of parameters, therefore, needs to be obtained in order to characterize material behaviour reliably. Furthermore, without major adaption, these models can only be used for low-stress applications where deformation behaviour is linear.

Alternative functions have been used to model the long-term deformation behaviour of polymers which can be traced to the parameters in spring and dashpot models but contain fewer parameters. A versatile function that is commonly used to model creep relaxation in semi-crystalline polymers and secondary relaxations in glassy polymers expresses a creep compliance function in the form [2-5]

$$D(t) = D_0 + \Delta D \left[ 1 - \exp \left\{ - (t / t_0)^n \right\} \right] \quad (1)$$

When creep loading is carried out under uniaxial tension,  $D(t)$  is then the tensile compliance function and is the ratio of the time varying tensile strain to the applied, constant tensile stress. The parameter  $D_0$  is the tensile compliance in the limit of short times,  $\Delta D$  is the magnitude of the creep relaxation process and is the difference between compliances in the limit of long and short times and  $t_0$  and  $n$  are parameters that identify the time range spanned by the process. The parameter  $t_0$  can be regarded as a mean retardation time for the process whilst  $n$  determines the distribution of retardation times such that, the smaller is  $n$ , the broader is the process.

This function is used to model relaxation processes whose relaxation magnitude is relatively small and where the creep time is comparable with, or less than, the mean retardation time parameter  $t_0$  for the process. It is therefore generally suitable for describing  $\beta$  relaxations in amorphous and semicrystalline polymers and  $\alpha$  relaxations in semi-crystalline polymers. Whilst the form of this function is relatively simple and it captures the main features of a relaxation process, it cannot be expected to accurately describe compliance changes over the whole time frame spanned by the process.

Under low levels of applied stress, compliances  $D(t)$  do not vary with applied stress, and deformation behaviour is therefore linear. With increasing stress, creep experiments on many polymers reveal a departure from linear behaviour. The main factor contributing to this non-linear behaviour is a reduction in the retardation time parameter  $t_0$  with increasing stress [6-10]. The effect of elevated stresses is therefore to increase molecular mobility so that the polymer creeps faster. Non-linearity is therefore an inherent feature of viscoelastic behaviour. It is generally more evident under long-term loading since most engineering plastics have relaxation processes at temperatures well

above ambient. Under short-term loading, deformation behaviour is therefore predominantly elastic. However, with increasing time under load, molecular relaxation associated with a high temperature relaxation process will be more extensive, and the effect of a change in  $t_0$  on material properties will be greater (see for example Figure 1).

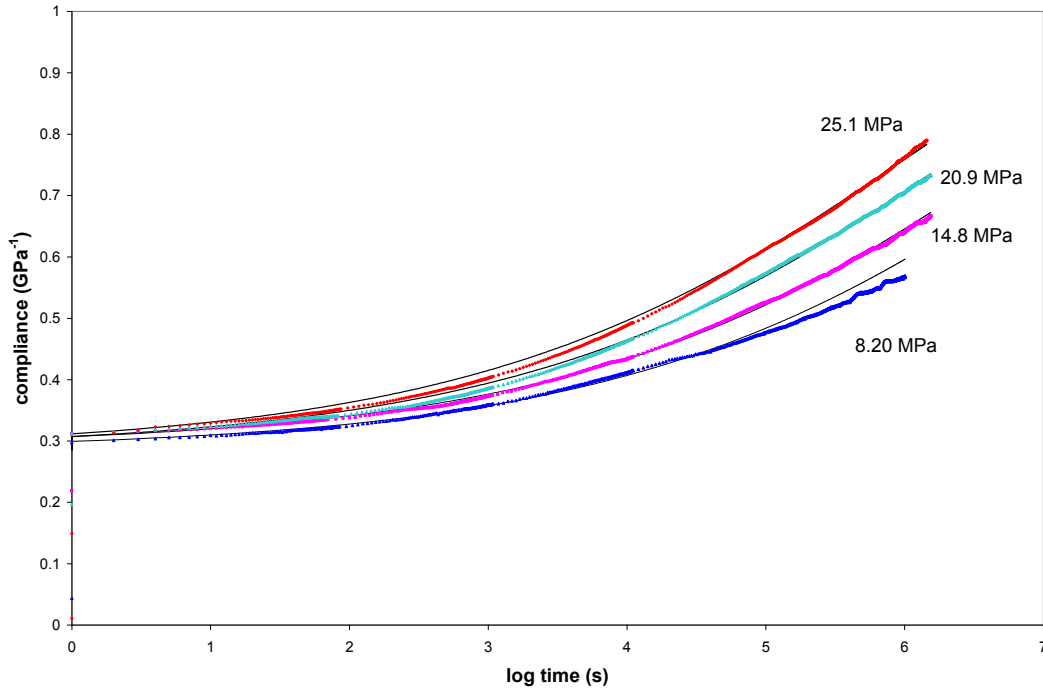


Figure 1. Tensile creep compliance curves for polyoxymethylene at different levels of stress. Curve fits are obtained using Equation (1) and the parameters in Table 1.

Creep tests under uniaxial compression reveal that the dependence of  $t_0$  on stress is different under tension from that under compression. This indicates that  $t_0$  depends on stress state as well as the stress magnitude. The proposal that  $t_0$  should depend on an effective stress has been explored in previous work [9, 10]. This concept has been extended in the model reported here to develop relationships for non-linear creep compliance under a general multi-axial stress.

In this paper, a non-linear creep model is introduced and applied to creep data obtained from tests on a polyoxymethylene measured under uniaxial tension and compression. Theoretical expressions are formulated that relate time-dependent stresses and strains for arbitrary, multi-axial stress or strain histories. Solutions to these expressions have been obtained for the case of uniaxial tension. These solutions are compared with

measurements obtained from a variety of tensile tests. These tests include intermittent loading and unloading, where a series of stresses are applied or removed in discrete steps with periods of constant stress between the steps. Also included is a stress relaxation experiment, where the strain is held constant, and tests carried out under constant displacement rate.

## **2 MATERIAL**

The polyoxymethylene material studied here was supplied by Du Pont under the trade name Delrin 1260 NC010. It was supplied as compression moulded plates 160 mm square and 3 mm thick. The tests reported in this paper were carried out on material where the time elapsed since the plates were moulded was in excess of 2 years. All the tests were carried out within a period of 4 months and the duration of each test did not exceed 2 weeks. All tests were therefore carried out on specimens of the same age, and changes to relaxation behaviour caused by physical ageing can be neglected.

## **3 EXPERIMENTAL**

Tensile creep tests were carried out on specimens consisting of rectangular bars of width 10 mm, thickness 3 mm and length 160 mm. These are clamped in creep machines such that one clamp is fixed and the other is attached to a lever arm, which supports an applied load at the other end. A pair of extensometers is attached to opposite faces of the specimen, and the gauge length of each extensometer is 25 mm. The tensile strain is determined from the average of the extensometer readings to eliminate the influence of specimen bending on the strain measurement. True values of stress and strain are used in the derivation of creep compliances. True stresses were calculated from engineering values and the value of Poisson's ratio in Table 2 that was assumed to be independent of time for the duration of the creep tests.

Creep tests under uniaxial compression were carried out on specimens 10 mm long by 10 mm wide and approximately 3 mm thickness. Specimens were loaded along the length direction between parallel platens in a universal test machine. An alignment jig was attached to one of the platens to enable the faces to be set parallel using slip gauges. The top and bottom faces of the specimens were machined smooth and

accurately parallel. Also the surfaces of the platens were lubricated with light oil to facilitate free lateral expansion of the specimen in the width direction. Extensometers were used to measure changes in the platen separation on loading. A creep load was applied by driving the moveable platen at a constant speed until the required stress was reached. This load was then maintained for the duration of the test. A plot of load against platen displacement during load introduction was curved at low stresses indicating an error in strain measurement arising possibly through the expulsion of oil between the ends of the specimen and the platen. The low-stress tail on this plot was used to derive a correction of 0.07 mm that was applied to all strain values during a creep test. In an attempt to reduce this correction, compressive creep tests were carried out on a specimen that was 10 mm long by 5 mm wide. The end constraint to lateral expansion will be less with this specimen so no lubricant was applied to the platen surface. The displacement correction was then reduced to 0.04 mm. This residual displacement error is presumably associated with establishing good contact between the specimen end faces and the platen surface. After correction, the compliance values from these two compressive creep tests differed by less than 2%

## **4 A MODEL FOR CREEP IN POLYOXYMETHYLENE**

### **4.1 MOLECULAR RELAXATION PROCESSES IN POLYOXYMETHYLENE**

Dynamic mechanical measurements over a wide temperature range can be used to reveal the presence of the various relaxation processes in a polymer that are responsible for viscoelastic behaviour. Dynamic property curves for the materials studied here were measured at a frequency of 3Hz and are shown in Figure 2. Three relaxation regions are evident centred at temperatures around  $-60^{\circ}\text{C}$ ,  $-5^{\circ}\text{C}$  and  $120^{\circ}\text{C}$ . The high temperature process is believed to be associated with the crystalline phase [11] and is responsible for long-term creep at ambient temperatures measured in the work reported here. There is some evidence that the relaxation process centred at  $-5^{\circ}\text{C}$  is not fully relaxed at a temperature of  $23^{\circ}\text{C}$ , and there is possibly some overlap with the low-temperature region of the high temperature process. This overlap would be evident in creep tests at very short times, but will be much less and maybe negligible at times greater than 10 s.

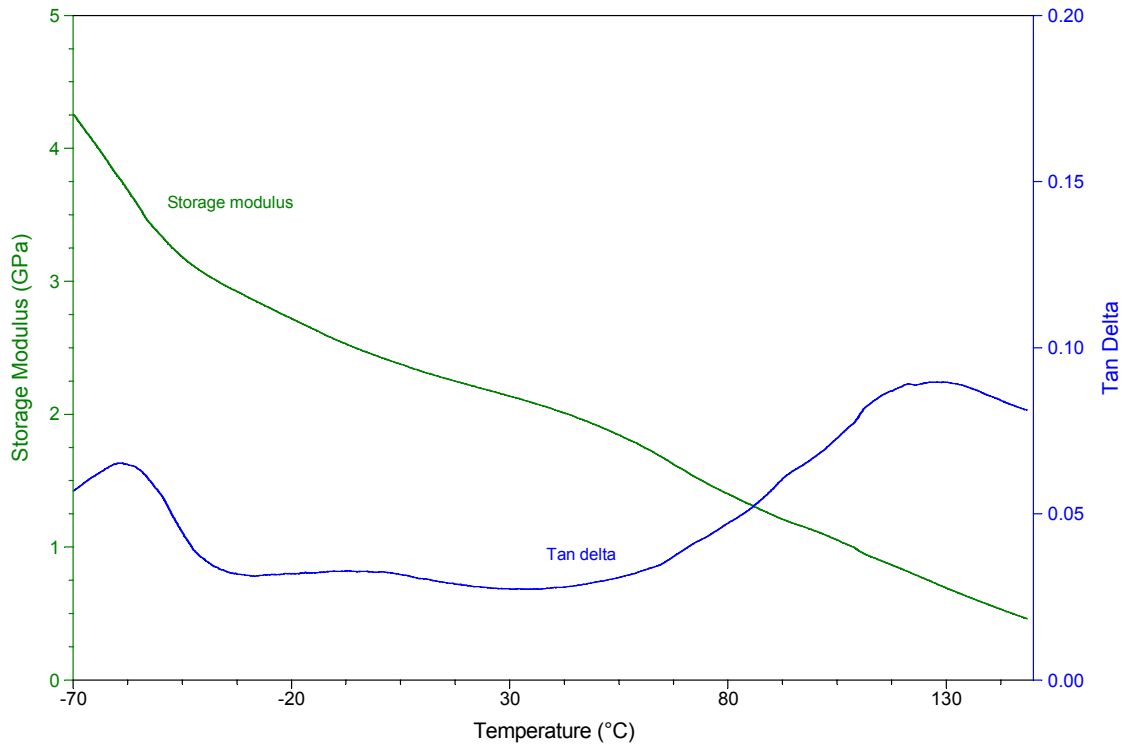


Figure 2. Temperature dependence of the dynamic mechanical properties of poly(oxyethylene) under tension.

#### 4.2 MODELLING NON-LINEAR CREEP UNDER UNIAXIAL TENSION.

The results of creep tests on polyoxymethylene under uniaxial tension are presented in Figure 1, which shows changes in the tensile creep compliance with log time measured under different tensile loads. Here the time-dependent compliance  $D(t)$  is defined by the ratio of the time-dependent strain  $\epsilon(t)$  to the applied constant tensile stress  $\sigma_T$  thus

$$D(t) = \epsilon(t) / \sigma_T \quad (2)$$

For the calculation of creep compliance, true values of stress and strain have been used whilst the stress levels recorded alongside each curve in Figure 1 are engineering stresses. The continuous lines with each set of experimental data have been obtained using Equation (1) with the parameter values listed in Table 1.

**Table 1: Values for the parameters in Equation (1) used to obtain the fits to data in Figure 1.**

| $\sigma_T$<br>(MPa) | $D_0$<br>(GPa <sup>-1</sup> ) | $\Delta D$<br>(GPa <sup>-1</sup> ) | $n$  | $t_0$<br>(s)       |
|---------------------|-------------------------------|------------------------------------|------|--------------------|
| 8.2                 | 0.285                         | 0.75                               | 0.24 | $1.35 \times 10^7$ |
| 14.8                | 0.285                         | 0.75                               | 0.24 | $6.3 \times 10^6$  |
| 20.9                | 0.285                         | 0.75                               | 0.24 | $2.2 \times 10^6$  |
| 25.1                | 0.285                         | 0.75                               | 0.24 | $1.0 \times 10^6$  |

In determining the parameter values shown in Table 1, we have followed other workers [3-5] by assuming that the only parameter that is influenced by the applied stress is the mean retardation time  $t_0$ . The possibility of small changes to some of the other parameters, especially  $n$  and  $\Delta D$ , cannot be discounted. In fact, the small departures between measured data and the curve predictions at 8 MPa and 25 MPa can be reduced by allowing  $n$  to increase with stress from a value of 0.22 at 8 MPa to 0.26 at 25 MPa. However, if changes in  $n$  or  $\Delta D$  are allowed in addition to changes in  $t_0$ , then the application of the proposed model to the prediction of stress and strain changes under a general time-varying stress or strain, considered in Section 5.1, becomes more difficult or impossible. In order for the model to have wide-ranging application, we have therefore attempted to obtain satisfactory fits to creep data based on the assumption that  $D_0$ ,  $n$  and  $\Delta D$  are, to a good approximation, independent of stress.

The dependence of the parameter  $t_0$  upon stress has been found to follow the empirical relationship

$$t_0 = A \exp(-\alpha \sigma_T^2) \quad (3)$$

where  $A$  and  $\alpha$  are additional material parameters. Figure 3 shows a plot of  $\log_e t_0$  against  $(\text{stress})^2$  obtained using the values of stress and  $t_0$  in Table 1. The continuous line is the best fit to the data obtained using Equation 3 with the values for  $A$  and  $\alpha$  recorded in Table 2. The form of this fit is consistent with creep behaviour being linear at low stress levels where  $t_0$  is essentially independent of stress.

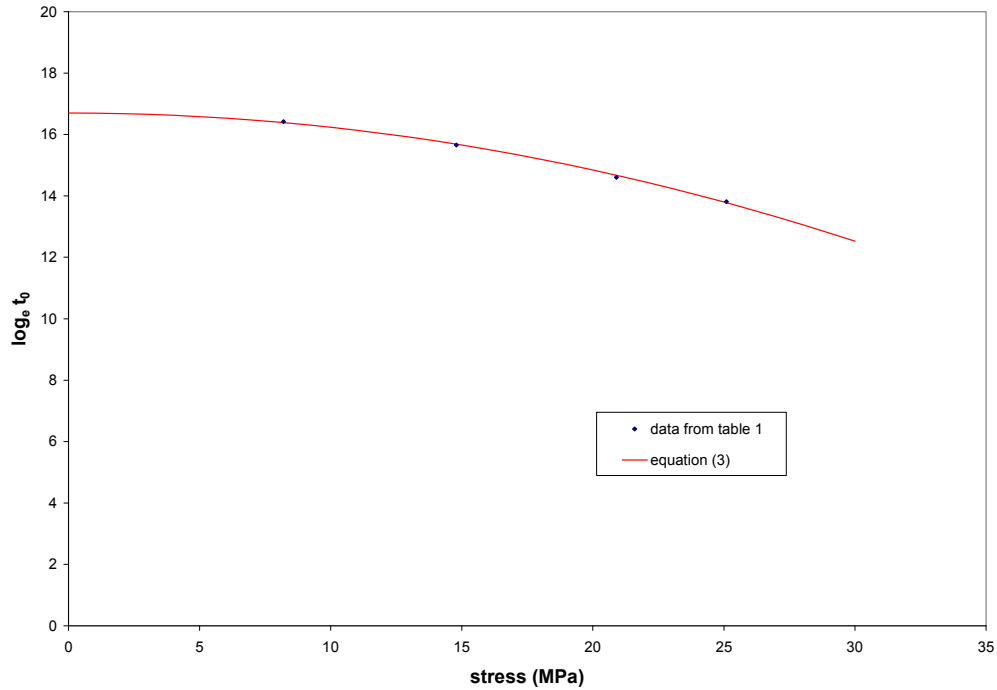


Figure 3. Plot of  $\log_e t_0$  against stress. Values for  $t_0$  were obtained from the curve fits to the data in Figure 1.

**Table 2: Values for additional creep model parameters measured on compression moulded polyoxymethylene.**

| A (s)              | $\alpha$ (MPa <sup>-2</sup> ) | $\mu$ | $\nu$ |
|--------------------|-------------------------------|-------|-------|
| $1.79 \times 10^7$ | 0.00464                       | 0.90  | 0.4   |

#### 4.3 MODELLING CREEP UNDER UNIAXIAL COMPRESSION

Creep tests under uniaxial compression were carried out in a preliminary attempt to investigate whether, in addition to the stress magnitude, there was any influence of the state of stress on creep behaviour. The state of stress is determined by the magnitude of the effective shear and hydrostatic components of the stress field, and the hydrostatic component is negative under a compressive stress and positive under tension. Measurements of creep compliance under a compressive stress of 26 MPa are shown in Figure 4, and compared with the tensile creep curve at a stress of 25.1 MPa from Figure 1. The compressive creep data in Figure 4 are results from two creep tests on specimens of different width as explained in Section 3. The best fit to the compression data has been obtained with the parameters listed in Table 3 and shows that creep under

compression has the same shape as under tension but is shifted less to shorter creep times by an elevated stress.

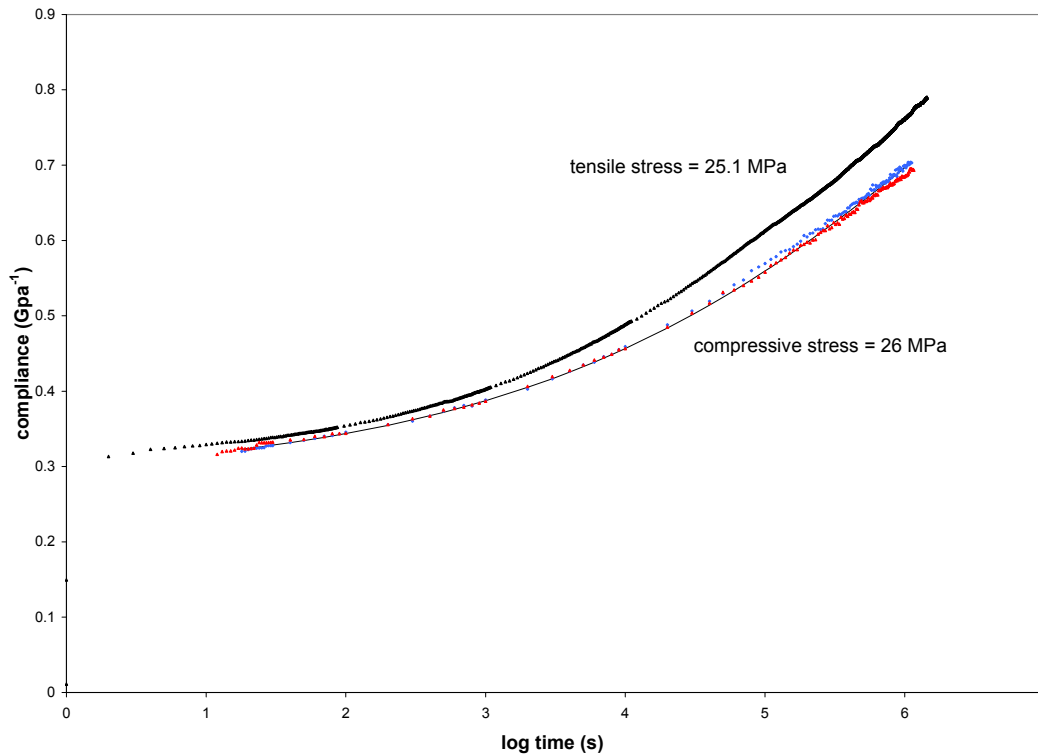


Figure 4. Comparison of creep compliance curves obtained under uniaxial tension and compression at similar levels of stress.

**Table 3. Values for the parameters in Equation (1) giving the best fit to the compressive creep data at 26 MPa in Figure 4.**

| $D_0$ (GPa <sup>-1</sup> ) | $\Delta D$ (GPa <sup>-1</sup> ) | $n$  | $t_0$ (s)         |
|----------------------------|---------------------------------|------|-------------------|
| 0.280                      | 0.75                            | 0.24 | $2.4 \times 10^6$ |

Assuming that the creep behaviour under tension and compression is the same at low stresses where behaviour is linear, this result demonstrates that the reduction in  $t_0$  with stress is less under compression than under tension. This suggests that in the relationship between  $t_0$  and stress given by Equation (3), the stress should be replaced by an effective stress  $\bar{\sigma}$  that is a combination of shear and hydrostatic parts. Thus Equation (3) under a multi-axial stress will become

$$t_0 = A \exp(-\alpha \bar{\sigma}^2) \quad (4)$$

Assuming the effective stress is a simple linear combination of shear and hydrostatic components,  $\bar{\sigma}$  will take the form

$$\bar{\sigma} = \mu\sigma_e + \lambda\sigma_m \quad (5)$$

where  $\mu$  and  $\lambda$  are parameters that determine the sensitivity of the effective stress to the shear and hydrostatic components of stress  $\sigma_e$  and  $\sigma_m$ , respectively. These components of stress are given in terms of principal components of a multi-axial stress  $\sigma_1$ ,  $\sigma_2$  and  $\sigma_3$  by

$$\sigma_e = \frac{1}{\sqrt{2}} [(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2]^{1/2} \quad (6)$$

and

$$\sigma_m = \frac{1}{3}(\sigma_1 + \sigma_2 + \sigma_3) \quad (7)$$

It is possible to define the effective stress  $\bar{\sigma}$  as being numerically equal to the uniaxial tensile stress  $\sigma_T$ . The parameters  $\mu$  and  $\lambda$  are then related, and Equation (5) becomes

$$\bar{\sigma} = \mu\sigma_e + 3(1 - \mu)\sigma_m \quad (8)$$

Therefore, under a uniaxial compressive stress  $-\sigma_C$  so that  $\sigma_C$  is positive

$$\bar{\sigma} = (2\mu - 1)\sigma_C \quad (9)$$

The continuous line alongside the compression data in Figure 4 has been obtained using Equation (1) with the same parameters obtained for tensile data, shown in Table 1, but with a value of  $t_0$  of  $2.4 \times 10^6$  s. Substituting the value for  $t_0$ , shown in Table 3, for the compressive creep test together with Equation (9) into Equation (4) gives a value of  $\mu = 0.90$ .

An extension of the model to the prediction of creep under a multi-axial stress state based on the reasoning behind Equation (8) is formulated in Section 5.1.3. Comparisons

of predictions and experimental measurements under more complex stress states will be reported in a separate paper.

## 5 APPLICATION OF THE CREEP MODEL UNDER TIME-VARYING STRESSES OR STRAINS

### 5.1 BACKGROUND THEORY

In order for creep data and the creep model described here to be used with a finite element system to carry out a stress analysis under long-term loading, a theoretical framework is needed that enables creep data to be used in situations where the local stress is not constant with time. Expressions are developed in this Section that relate time-dependent stresses and strains, for an arbitrary, 3-dimensional stress or strain history, and take account of the physical origin of non-linearity in the creep model.

#### 5.1.1 Linear behaviour under uniaxial tension

Under a time-varying stress that is low enough that behaviour is at all times linear, the resulting strain can be calculated by summing strain increments produced by incremental changes in stress. These strain increments are the product of the stress increment and the creep compliance that is retarded by the time at which the stress changes occur. For continuous changes in stress, the summation becomes the Boltzmann superposition integral [1], which gives the time-dependent strain under a uniaxial stress history  $\sigma(\tau)$  as

$$\varepsilon(t) = \int_0^t \dot{\sigma}(\tau) \left[ D_0 + \Delta D \left( 1 - \exp \left\{ - \left( \frac{t-\tau}{t_0} \right)^n \right\} \right) \right] d\tau \quad (10)$$

#### 5.1.2 Non-linear behaviour under uniaxial tension

The superposition principle embodied in Equation (10) is not valid when deformation is non-linear because the time parameter  $t_0$  changes with stress, and these changes will influence the creep response from the previous stress history. To account for changes in

the time parameter, it is proposed to replace the time term in Equation (10) by a reduced time  $s$  that is defined by

$$s = \int_0^t \frac{du}{t_0(\sigma(u))} \quad (11)$$

This approach follows the work carried out by many authors [12, 14-16] to model creep in polymers undergoing simultaneous physical ageing. Under these circumstances, the parameter  $t_0$  is changing with time, even under a constant stress, as the polymer ages.

The strain history produced by a time-varying stress will be obtained by incorporating Equation (11) into Equation (10) to give

$$\epsilon(t) = \int_0^t \dot{\sigma}(\tau) \left[ D_0 + \Delta D \left( 1 - \exp \left\{ - \left( \int_{\tau}^t \frac{du}{t_0(\sigma(u))} \right)^n \right\} \right) \right] d\tau \quad (12)$$

### 5.1.3 Creep response for multi-axial stress states

Relationships between multi-axial stresses and strains involve the use of tensor stress and strain components  $\sigma_{ij}(t)$  and  $\epsilon_{ij}(t)$  which, in general will be time-dependent. The stress and strain tensors can be decomposed into their deviatoric and hydrostatic parts as follows

$$\begin{aligned} \sigma_{ij} &= \sigma_{ij}^d + \frac{1}{3} \sigma_{kk} \delta_{ij}, \\ \epsilon_{ij} &= \epsilon_{ij}^d + \frac{1}{3} \epsilon_{kk} \delta_{ij}, \end{aligned} \quad (13)$$

where  $\sigma_{ij}^d$  and  $\epsilon_{ij}^d$  are components of the deviatoric stress and strain tensors, respectively,  $\sigma_{kk}$  and  $\epsilon_{kk}$  are the scalar hydrostatic stress and strain, respectively, and all of these components will be time-dependent in general. Also  $\delta_{ij}$  has the value 1 when  $i = j$  and the value 0 when  $i \neq j$ .

For isotropic materials, deviatoric stress and strain components are related through equations similar to Equation (12) where the tensile creep compliance function is replaced by the shear creep compliance giving

$$\epsilon_{ij}^d(t) = \int_0^t \dot{\sigma}_{ij}^d(\tau) \left[ \frac{J_0}{2} + \frac{\Delta J}{2} \left( 1 - \exp \left\{ - \left( \int_{\tau}^t \frac{du}{t_0(\bar{\sigma}(u))} \right)^n \right\} \right) \right] d\tau. \quad (14)$$

where  $J_0$  is the shear compliance in the limit of short times and  $\Delta J$  is the magnitude of the creep relaxation process under a pure shear stress. Also, the value of the retardation time parameter  $t_0$  is determined by the effective stress  $\bar{\sigma}$ , which is given by Equation (8). Note that the effective stress also determines the value for  $t_0$  in Equation (12) but, under uniaxial tension, the effective stress, as defined by Equation (4), is the same as the tensile stress.

Similarly, the hydrostatic stress and strain are related through a similar relationship involving the bulk creep compliance function as follows

$$\epsilon_{kk}(t) = \int_0^t \dot{\sigma}_{kk}(\tau) \left[ \frac{B_0}{3} + \frac{\Delta B}{3} \left( 1 - \exp \left\{ - \left( \int_{\tau}^t \frac{du}{t_0(\bar{\sigma}(u))} \right)^n \right\} \right) \right] d\tau, \quad (15)$$

where  $B_0$  is the bulk compliance in the limit of short times and  $\Delta B$  is the magnitude of the creep relaxation process under a pure hydrostatic stress.

There are assumptions in Equations (14) and (15). The retardation time parameters under shear and hydrostatic stresses are assumed to be determined by the effective stress, given by Equation (8). Also, it is assumed that the distribution of retardation times under shear and hydrostatic stresses is the same as under uniaxial tension and is determined by the parameter  $n$ . Based on these assumptions, a multi-axial stress analysis of a non-linear, viscoelastic material involves the determination of solutions to equations (13), (14) and (15) and the measurement of the materials parameters in these equations. Tensile creep tests can be readily used to determine  $t_0$ ,  $\bar{\sigma}$  and  $n$  as well as  $D_0$  and  $\Delta D$ . Additional tests would need to be carried out to determine the time-dependence

of either the shear compliance, the bulk compliance or Poisson's ratio. These are difficult and non-routine tests. In the absence of data from such tests, it is possible to proceed by assuming either that there is negligible time-dependence in the bulk compliance, in which case  $\Delta B$  is zero, or that there is negligible time-dependence in Poisson's ratio. If the Poisson's ratio can be assumed to be constant with time, the parameters  $J_0$ ,  $\Delta J$ ,  $B_0$  and  $\Delta B$  are related to measurable quantities  $D_0$  and  $\Delta D$  and a constant Poisson's ratio as follows

$$\begin{aligned} J_0 &= 2D_0(1 + \nu) , \\ \Delta J &= 2\Delta D(1 + \nu) , \\ B_0 &= 3D_0(1 - 2\nu) , \\ \Delta B &= 3\Delta D(1 - 2\nu) . \end{aligned} \tag{16}$$

## 5.2 EXPERIMENTAL EVALUATION UNDER UNIAXIAL TENSILE STRESS HISTORIES

In related work, code is being developed to obtain solutions to Equations (13), (14) and (15) using the solver in a finite element system. In the remainder of this paper, the validity of the solution under uniaxial tension, presented in Section 5.1.2, is explored by comparing predictions of stress or strain with experimental measurements under a variety of uniaxial tensile stress and strain histories. The relationship between time-dependent stresses and strains under uniaxial tension is given in Section 5.1.2 by Equation (12). A Fortran routine has been written to obtain solutions to this equation for any prescribed stress or strain history.

### 5.2.1 Step loading and unloading

In these tests, the stress was held constant for a period of time and then increased or decreased in discrete steps. In the step-loading test, a stress of 10 MPa was applied for 167000 s and then increased to 20 MPa for times up to 342000 s when it was increased to 25 MPa for the remainder of the experiment. Calculated strains for this load history were obtained using Equation (12), and are compared with experimental measurements in Figure 5. The agreement is seen to be very good.

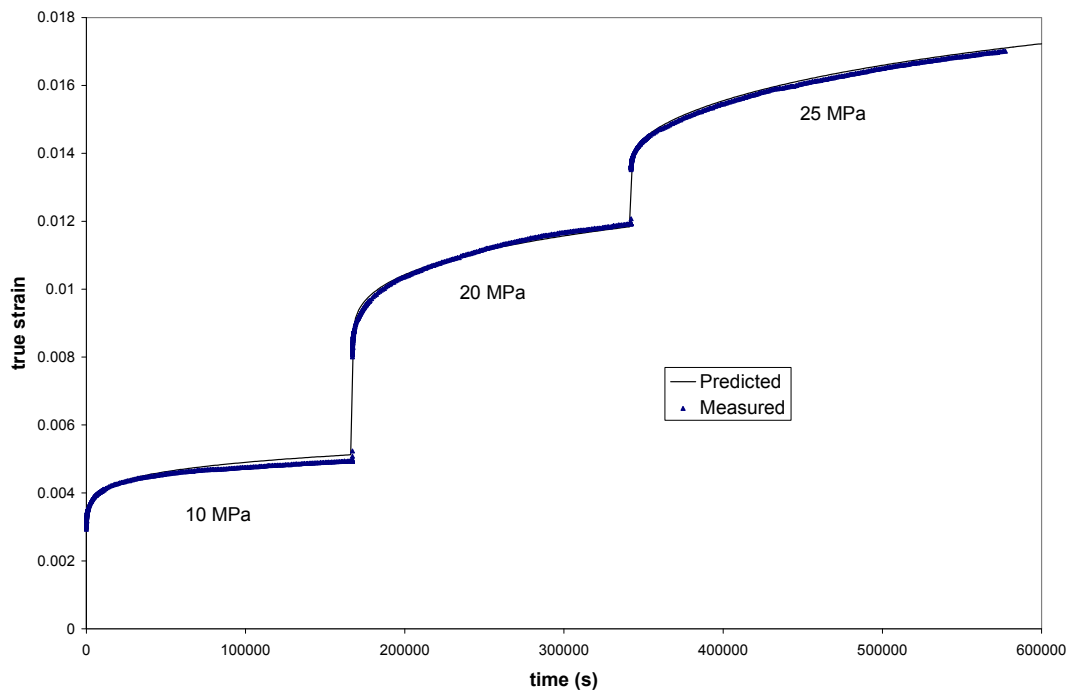


Figure 5. Comparison of measured and predicted values of strain against time for the step-loading test.

In Figure 6, measurements and calculations are compared for the step-unloading test. The stress history in this test was 25 MPa for times up to 167000 s at which point the stress was reduced to 20 MPa for times up to 342000 s when it was reduced to 10 MPa for the remainder of the test. Here there is some departure of the calculated strain from measured values following the reduction of the stress from 20 MPa to 10 MPa. This result was reproduced in repeat tests and indicates that the parameter  $t_0$  has a lower value than expected for a stress of 10 MPa leading to more rapid recovery initially. Separate calculations reveal that if the value for  $t_0$  on reduction of the stress from 20 MPa to 10 MPa is held at the value appropriate to the higher stress rather than increasing instantaneously to the value associated with 10 MPa, then the predicted recovery is very close to the measured values. Predictions fall below measured values after about 200,000 s suggesting that the increase in  $t_0$  associated with a drop in stress occurs slowly leading to more rapid recovery initially. This interpretation is also consistent with the results when the stress is reduced from 25 MPa to 20 MPa, but the predicted change in  $t_0$  is lower in this case, and the associated error in strain prediction is less and barely noticeable. This issue of the predictive accuracy during stress removal is investigated further in Section 5.2.4 using creep recovery experiments.

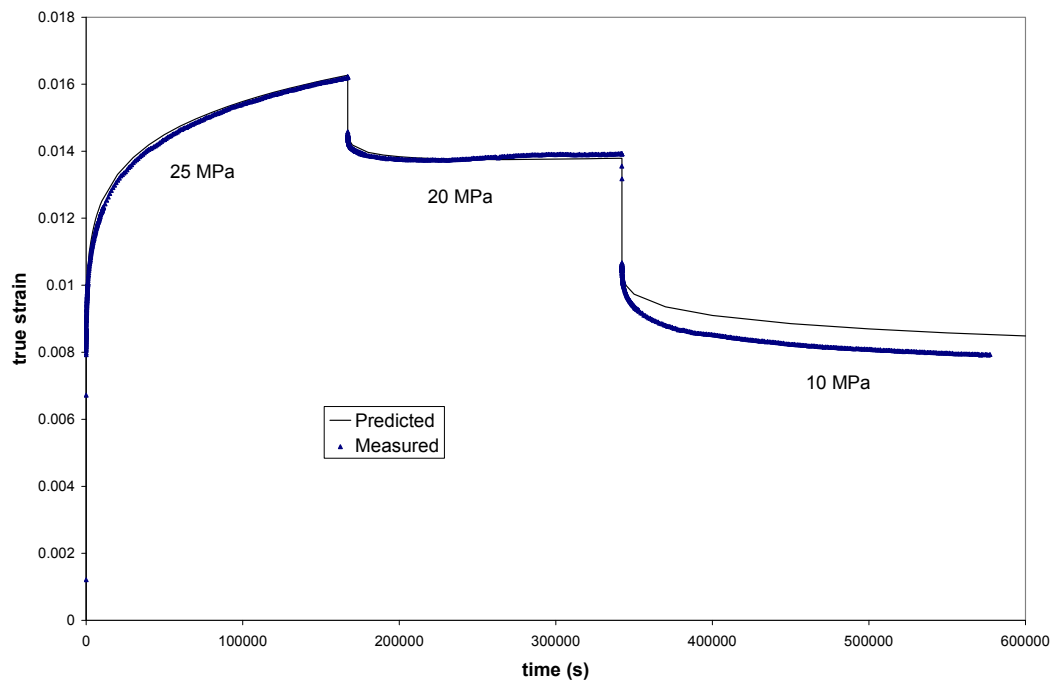


Figure 6. Comparison of measured and predicted values of strain against time for the step-unloading test.

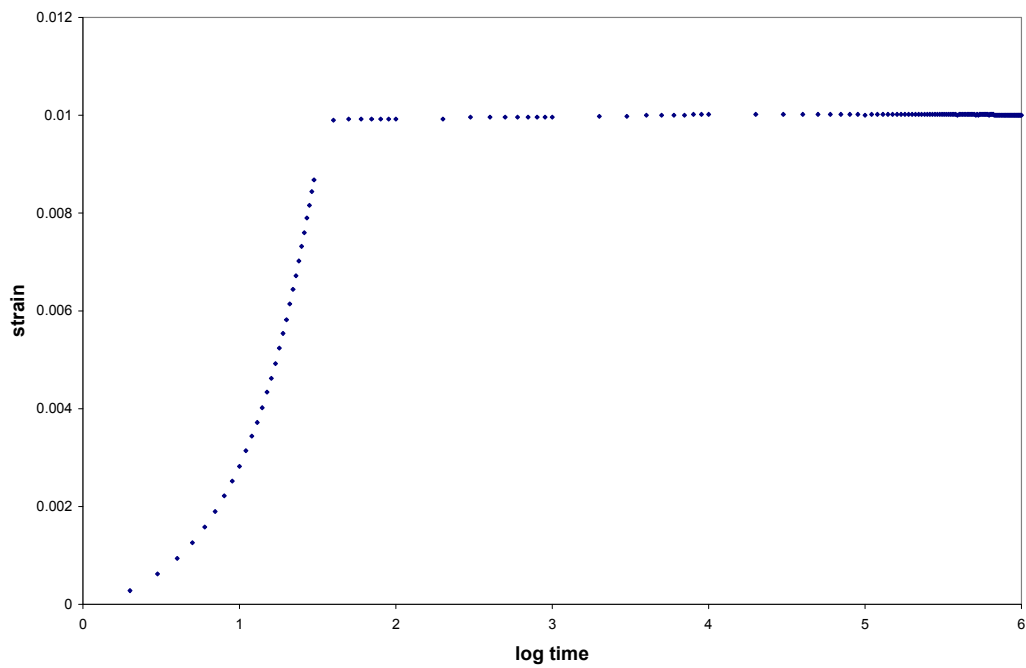


Figure 7. Strain history for the stress relaxation test.

### 5.2.2 Stress relaxation under constant strain

A stress relaxation test was carried out by loading a specimen at a speed of 1 mm/minute to a strain of 0.01 in a universal testing machine and maintaining the strain at this level throughout the remaining duration of the test. The strain in the specimen was measured using extensometers contacting the specimen. The strain history is shown in Figure 7. In Figure 8, the calculated stress is compared with measured values. The agreement is very satisfactory and is further support for the theoretical basis behind the calculations.

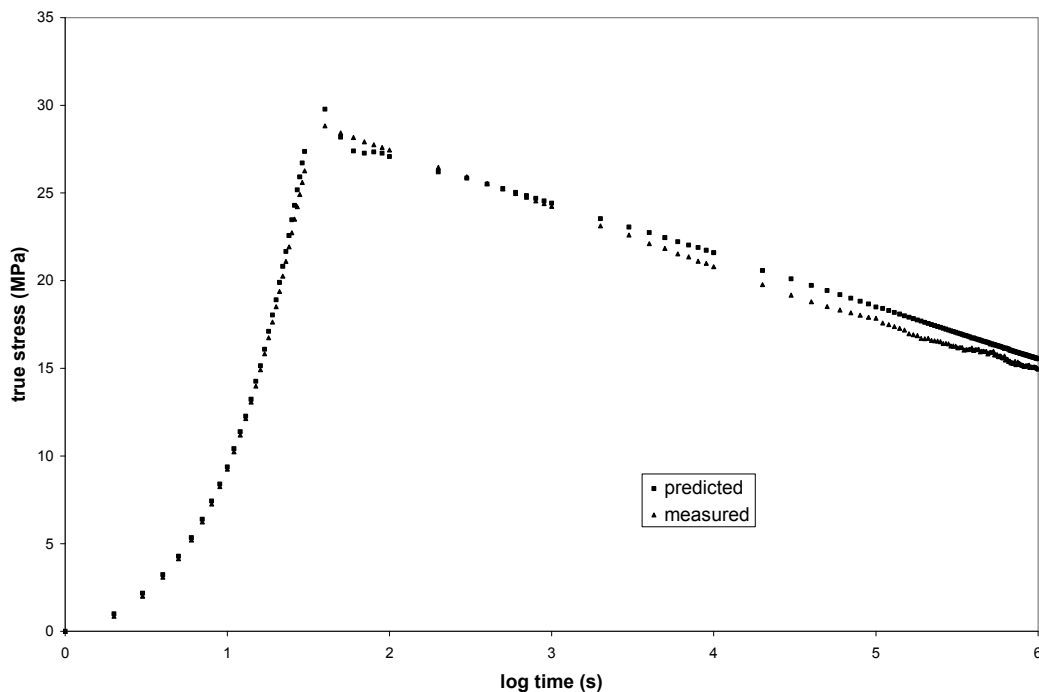


Figure 8. Comparison of measured and predicted values of stress against log time from the stress relaxation test. The applied strain is 0.01.

### 5.2.3 Constant displacement rate

In these tests, a tensile specimen was loaded in a universal testing machine at a constant speed to a stress of 26 MPa. The machine movement was then reversed at the same speed until the load was removed. The strain history was measured using extensometers in contact with the specimen. Tests were carried out at two test speeds of  $2.8 \times 10^{-3}$  mm/s and  $2.8 \times 10^{-5}$  mm/s. The strain rate was not constant during the test because of non-linear deformation of the specimen. Mean strain rates were  $1.6 \times 10^{-5} \text{ s}^{-1}$

and  $1.6 \times 10^{-7} \text{ s}^{-1}$ , respectively. The measured strain histories were input data for the calculation of stress over the time span of the loading and unloading. Plots of the calculated and measured stress against strain are shown in Figure 9. It can be seen that, during the loading phase, the calculations accurately predict the shape of the stress-strain curves and their dependence on loading rate. During reversal of the applied strain, the measured strain is consistently smaller at every reduced stress than that predicted by the theory. The strain recovery is therefore more rapid than predicted and is consistent with the departure of predicted and measured strains in the step unloading experiment in Figure 6. Once again, this suggests that, during unloading, the polymer maintains the mobility associated with the higher stress for some duration rather than immediately adopting the  $t_0$  value for the current stress that is assumed by the theory.

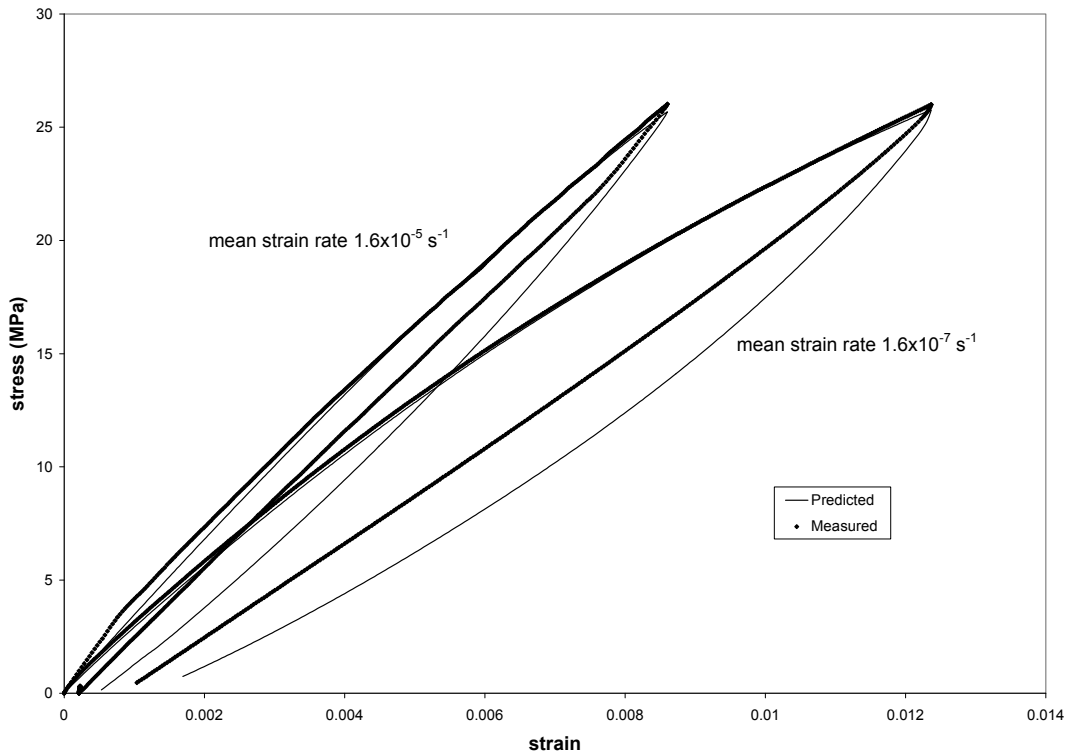


Figure 9. Comparison of measured and predicted stress-strain curves for tensile tests carried out under 2 different constant displacement rates.

It is apparent from Figure 9 that complete recovery of strain would probably occur if additional time had been allowed in the experiment, as would be expected for deformation that is viscoelastic. At significantly higher levels of applied stress, plastic

deformation will contribute to the creep process, and complete recovery would then not be expected.

#### 5.2.4 Creep recovery

In order to explore further the nature of the errors produced when the model and theory are used to predict recovery, a series of creep and recovery tests has been carried out. In these tests, the strain has been measured following total removal of the stress at the end of a creep experiment. Calculated and measured strains are compared for tests at low stress levels in Figure 10.

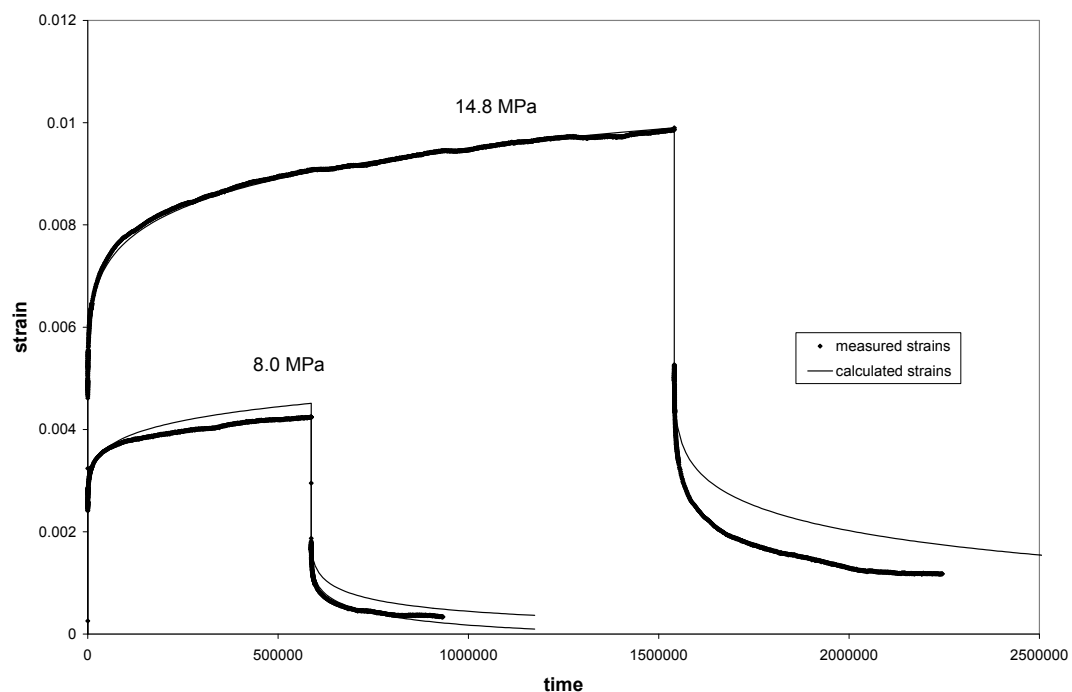


Figure 10. Comparison of measured and calculated creep strains in creep and recovery tests carried out at low and intermediate stress levels.

At the lower stress of 8.0 MPa, creep strains under load predicted by the model, with the parameters given in Table 1, show a small but progressive departure from measured strains, as observed in the creep results in Figure 1. At this stress level, there is therefore a discrepancy in the predicted and measured strains at the start of the recovery period. If this is taken into account, there is very close agreement between predicted and measured strains during recovery. This is presumably due to there being little difference in the  $t_0$  values for stress levels of zero and 8 MPa since 8 MPa is only just outside the

stress range for linear behaviour (see Figure 3). At 14.8 MPa, there is a clear discrepancy between measured and predicted strains during recovery, which increases progressively for larger creep stresses as shown in Figure 11.

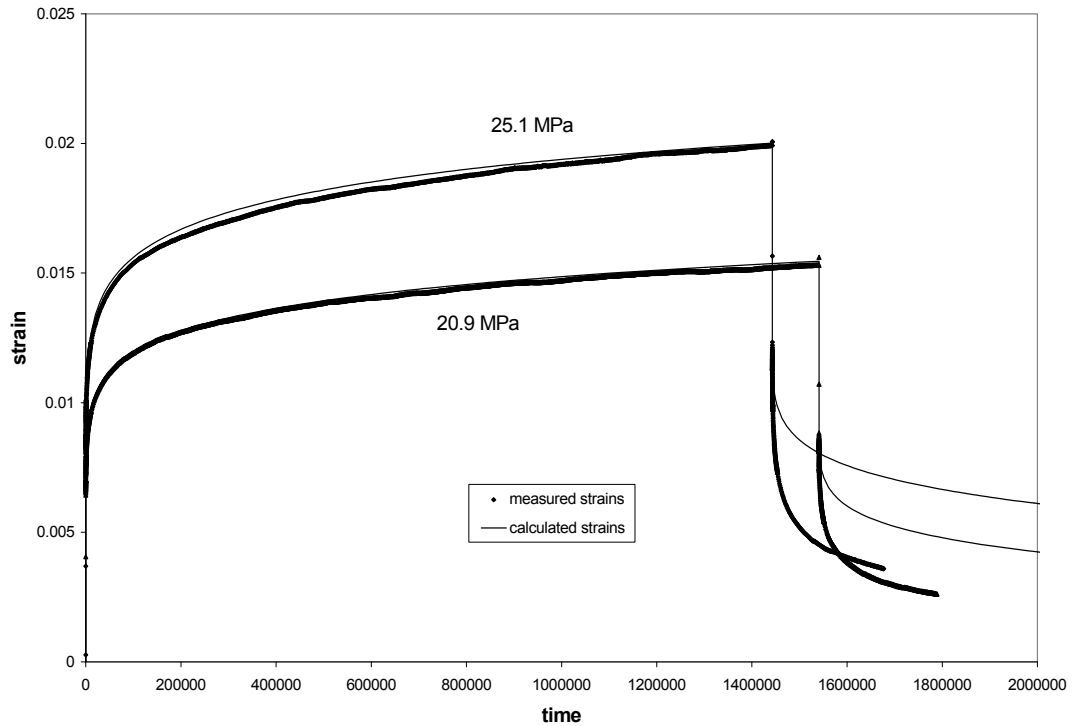


Figure 11. Comparison of measured and calculated creep strains in creep and recovery tests carried out at high stress levels.

During recovery, the stress is zero so the calculation will adopt a large  $t_0$  value. The results in Figures 10 and 11 indicate that, in the initial stages of recovery,  $t_0$  is much smaller and possibly close to its value when under stress. In Figure 12, the recovery strains for the tests in Figure 11 have been calculated assuming a  $t_0$  value that is the same as that during loading. Predicted strains are now much closer to measured values, although with increasing time, there is evidence that  $t_0$  does increase progressively.

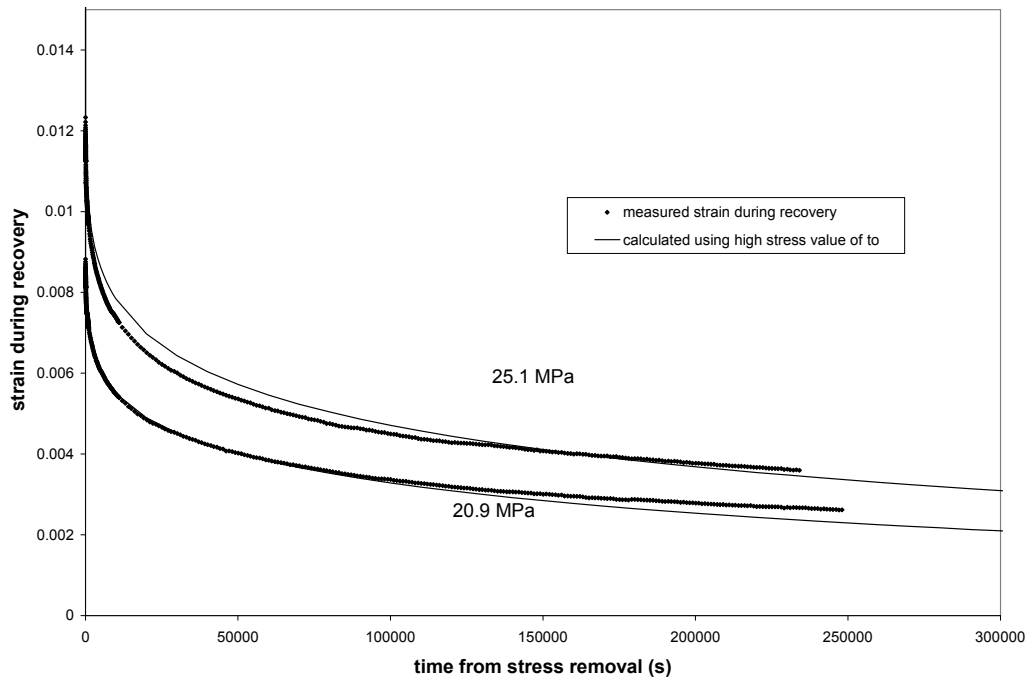


Figure 12. Comparison of the measured strain following stress removal for the tests in Figure 11 with calculated strains obtained assuming no change in the values for the time parameters following stress removal.

## 6 DISCUSSION

The results in Figures 6, 9, 10, 11 and 12 demonstrate that on reduction or removal of a stress high enough to produce non-linear deformation, the time parameter  $t_0$  does not take up the value appropriate to the lower stress instantaneously but maintains the high stress, more mobile, value for a significant period of time. Recovery times and the factors that influence these times have not been investigated here, but it is clear that the retardation in the recovery of  $t_0$  on stress removal is most evident for large and rapid stress changes as seen in Figures 10 and 11. For low rates of stress reduction, the changes in  $t_0$  with stress are small and predictions are close to experiment as seen in Figures 6, 8 and 9. The problem with predicting creep recovery from stresses in the non-linear region of deformation has been reported by other workers. The interpretation given by Read [12] and Santore [13] support the observations made here in terms of a delay in the increase in the creep retardation time parameter  $t_0$  on stress reduction, but their work indicates that the time parameter actually decreases initially below the value

at the high stress. It subsequently increases progressively towards a value appropriate to the lower stress. This conclusion may be supported by the results shown in Figure 12 where, by using a  $t_0$  value below that for the higher stress, slightly better agreement could be obtained between calculated and experimental strains during recovery. The results in Figure 12 and the precision of our creep model and parameters are, however, probably not sufficiently high to verify such an interpretation. Furthermore, these referenced studies were carried out on young material so that significant changes to the parameter  $t_0$  were taking place during the tests through physical ageing. As noted in Section 2, the tests on polyoxymethylene reported here were carried out on well-aged material, but issues regarding whether elevated stresses reduce the physical age of a specimen still need to be resolved.

## 7 CONCLUSIONS

The tensile creep behaviour of the polyoxymethylene material studied here can be modelled using a stretched exponential function with four material parameters. For creep times up to  $10^6$  s and at ambient temperature, three of the parameters are, to a good approximation, independent of stress. The fourth parameter, which represents a mean retardation time for the relaxation process responsible for creep, decreases with increasing stress and gives rise to non-linearity in creep behaviour. Non-linearity in creep behaviour in polyoxymethylene is evident at stress levels above around 8 MPa.

Creep tests under uniaxial compression reveal that the magnitude of the retardation time parameter depends on the stress magnitude as well as the stress state. A relationship has been proposed relating the time parameter to an effective stress that is a linear combination of the shear and hydrostatic components of the stress. This has enabled expressions to be proposed relating components of strain and stress under a multi-axial creep stress. Where accurate measurements of creep behaviour have only been made under a uniaxial tensile stress, derivation of the expressions for creep under multi-axial stress will require the assumption that either the Poisson's ratio or the bulk modulus does not change with time.

Theoretical expressions relating time-dependent stresses and strains under a general stress or strain history have been developed and give predictions that compare well with

experiment for some simple stress and strain histories under uniaxial tension. Predictions depart from experiment under conditions where the stress is reduced. These departures are small for small and slow reductions of stress, but become more significant as the stress decrease becomes greater and more rapid. This observation is consistent with a delay in the increase in the time parameter  $t_0$  when the stress is decreased which is not included in the theory. This delay does not appear to be significant when the stress is increased.

## ACKNOWLEDGEMENT

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