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A Review of Creep Modelling for Toughened Adhesives and Thermoplastics

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ABSTRACT

This report provides an overview of predictive models for determining deformation of toughened adhesives and thermoplastic materials under creep loading conditions. The report includes a new model developed at the National Physical Laboratory for characterising the non-linear creep behaviour of rubber-toughened adhesives. The model, which has been adopted for use with the finite element software package ABAQUS, has been used to predict extensions in both bulk adhesives and bonded joints. It is intended in future work to assess the applicability of the creep model for thermoplastic materials, such as polypropylene and polyethylene.

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1 INTRODUCTION

Creep is the increase in strain or deformation of a material with time when the material is subjected to a constant load for an extended period of time (i.e. time-dependent deformation). The change of strain at any time increases with load, temperature and relative humidity. Viscoelastic materials, such as adhesives and thermoplastics, can undergo creep deformation at relatively low stress levels (well below the ultimate strength of the material) and low temperatures (i.e. room temperature - referred to as cold flow). This can lead to considerable reduction in life expectancy of the component. The present cost to UK industry of plastic failures is estimated at £300M per annum of which 10% can be attributed to creep/relaxation failures [1]. Costs and inconvenience attributable to adhesive failures can be expected to be similar in magnitude (if not higher) with the added concern that bonded structures are often used in primary load-bearing applications.

It is therefore important to be able to predict (and design for) the effects of long-term loading on deformation and failure behaviour. Confidence in predictions requires the use of models that accurately describe the deformation behaviour of the polymeric materials - accounting for non-linear creep under multi-axial stresses. The non-linearity arises because of a progressive reduction, with stress level, in the relaxation times of molecular relaxation processes, which account for the creep deformation.

This report examines models used to describe creep deformation behaviour of thermoplastics and structural adhesives. It includes a new model developed at the National Physical Laboratory for describing the creep behaviour of glassy adhesives. The creep model can be represented as a generalised model for time-dependent plasticity in the finite element analysis (FEA) package ABAQUS, which enables changes in the stress and strain distributions with time under load to be calculated for the adhesive layer of bonded joints. It is intended in future work to assess the creep models applicability to thermoplastic materials, such as polypropylene and polyethylene. The review forms part of the *Measurements for Performance Programme* project “Prediction of the Lifetime of Adhesive Joints Under Sustained Loading” funded by the United Kingdom Department of Trade and Industry.

2 MODELLING LINEAR CREEP

Combinations of spring and viscous dashpot elements in series and parallel (e.g. Maxwell and Voigt models) may be used to model the time-dependent, viscoelastic behaviour of polymeric materials. The spring and dashpot configuration shown in Figure 1 accommodates a short-term, elastic or unrelaxed response as well as a long-term, limiting deformation corresponding to a fully relaxed state [2].

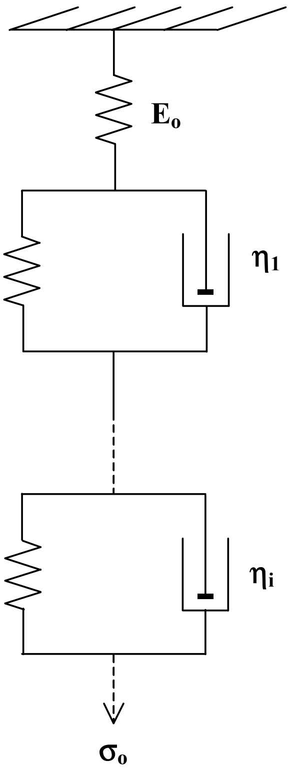

Figure 1: A spring and dashpot model for linear creep in polymers

For a model consisting of the 3 elements $\mathbf{E_o}$, $\mathbf{E_1}$ and $\boldsymbol{\eta_1}$, the strain response $\boldsymbol{\varepsilon}\mathbf{(t)}$ to a constant stress $\boldsymbol{\sigma_o}$ is:

$$\varepsilon(t) = \frac{\sigma_o}{E_o} + \frac{\sigma_o}{E_1}\left(1 - \exp -\left(\frac{t}{\tau_1}\right)\right) \tag{1}$$

where the relaxation time $\boldsymbol{\tau_1}$ is given by:

$$\tau_1 = \frac{\eta_1}{E_1} \tag{2}$$

The single relaxation time model given above is too simplistic and is unable to describe the actual relaxation processes that occur in polymers, which have a very broad distribution of relaxation times. This model can be extended, through the incorporation of additional spring and dashpot (Voigt) elements in series to broaden the spectrum of relaxation times, and hence the time span of the relaxation process being modelled. The strain response with the inclusion of these additional elements to an applied stress is:

$$\varepsilon(t) = \frac{\sigma_o}{E_o} + \sigma_o \sum_{i=1}^{n} \frac{1}{E_i}\left(1 - \exp -\left(\frac{t}{\tau_i}\right)\right) \tag{3}$$

where there are **n** Voigt elements in the model.

The large number of parameters that need to be determined in this model is inconvenient and is usually not necessary for modelling creep in glassy polymers at temperatures well below the glass-to-rubber transition temperature (**T_g**). Creep strains for glassy polymers can be described by a simpler expression:

$$\varepsilon(t) = \frac{\sigma_o}{E_o} \exp\left(\frac{t}{t_o}\right)^m \tag{4}$$

This function will only model the short-time tail of the relaxation function given by Equation (3), but this is usually a valid approximation, even for extended periods under load, provided the measurement temperature is not close to **T_g**. In Equation (4), the exponent **m** characterises a broad spectrum of relaxation times whose mean or effective value is **t_o**. The equation can also be expressed as a creep compliance function **D(t)** where [3]:

$$D(t) = \frac{\varepsilon(t)}{\sigma_o} = D_o \exp\left(\frac{t}{t_o}\right)^m \tag{5}$$

The magnitude of the parameter **t_o** is dependent on temperature, stress level and stress state. The dependence on stress level gives rise to non-linear creep behaviour (see Section 3). The magnitude of **t_o** also depends on the state of physical ageing of the adhesive at the time of the creep loading. Creep tests within the current programme have been carried out on well-aged specimens so as to minimise the effects of physical ageing during the creep tests. For longer term loading situations (i.e. years), the effects of physical ageing on creep would need to be included for high accuracy predictions (see Appendix 1).

For semi-crystalline polymers, such as polypropylene and polyethylene, where the temperature is close to **T_g**, an alternative approach is to employ the Williams-Watts equation [4]:

$$D(t) = D_o + \Delta D\left(1 - \exp\left[-\left(\frac{t}{t_o}\right)\right]^m\right) \tag{6}$$

It should also be noted that the Williams-Watt function might not fit experimental data for all semi-crystalline polymers.

3 NON-LINEAR CREEP UNDER UNIAXIAL STRESSES

At short creep times, the compliance curves exhibit slight non-linearity. This non-linear behaviour increases significantly with time under load. At higher stresses, the creep curves are observed to shift to shorter times. Figures 2 and 3 show compliance curves for DP460 (two-part epoxy adhesive cured for 24 hours at 23ºC and subsequently post-cured at 100ºC for 30 minutes - supplied by 3M Ltd) and XD4601 (a one-part adhesive cured at 180°C for 1hr - supplied by Dow Plastics).

The shift to shorter times is attributed to an increase in molecular mobility brought about by the application of elevated stresses that results in a reduction in the mean relaxation time $\mathbf{t_0}$. It is the enhanced mobility that gives rise to non-linear creep behaviour.

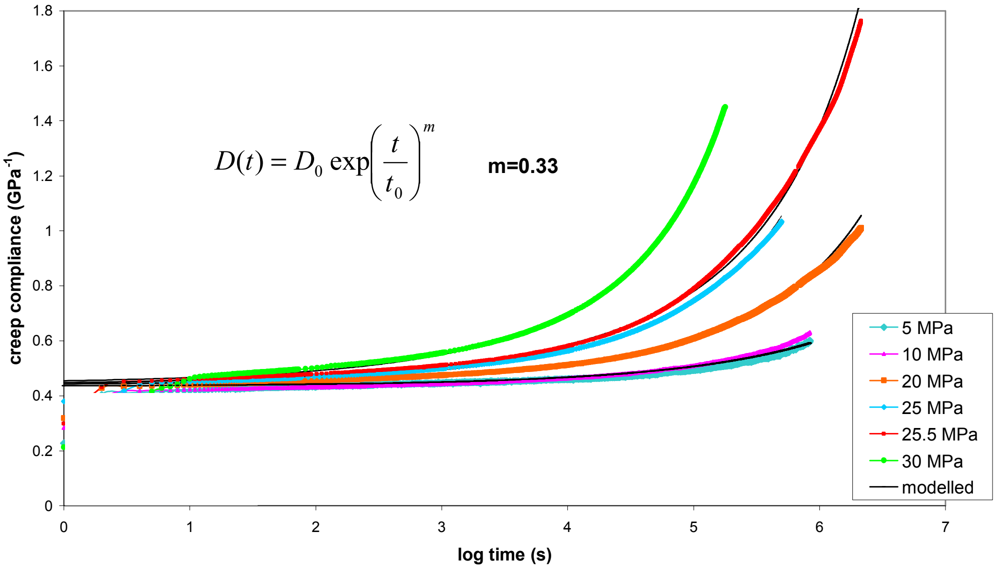

Figure 2: Creep compliance curves for DP460 at different levels of stress

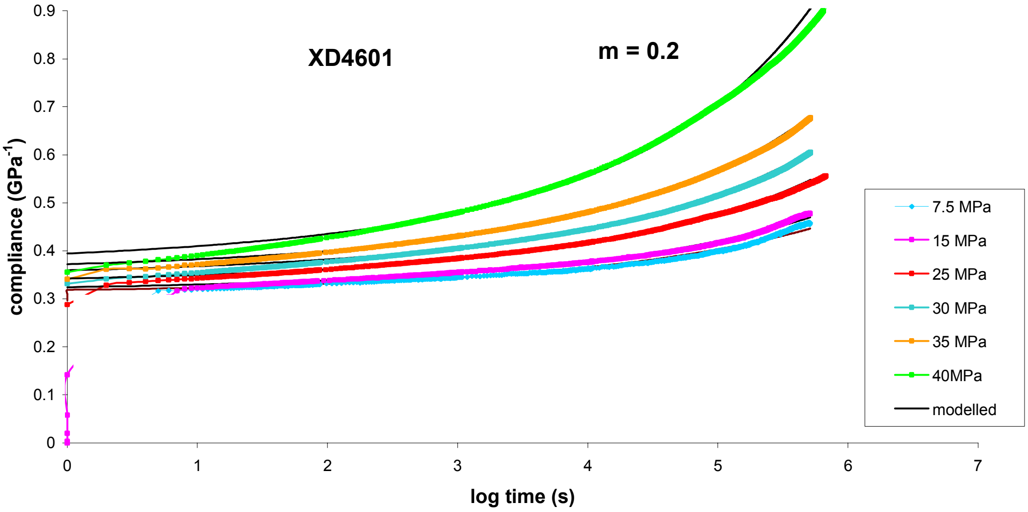

Figure 3: Tensile creep compliance curves for XD4601 at different levels of stress

The variation of $\mathbf{t_o}$ with $\boldsymbol{\sigma_o}$ can be described with satisfactory accuracy by the empirical relationship:

$$\mathbf{t_o = A \exp -\alpha\sigma_o^2} \tag{7}$$

The parameters **A** and **α** can be derived from a linear regression fit to the plot of $\mathbf{\log_e t_o}$ versus $\boldsymbol{\sigma_o}^2$ (see Figures 4 and 5, and Table 1).

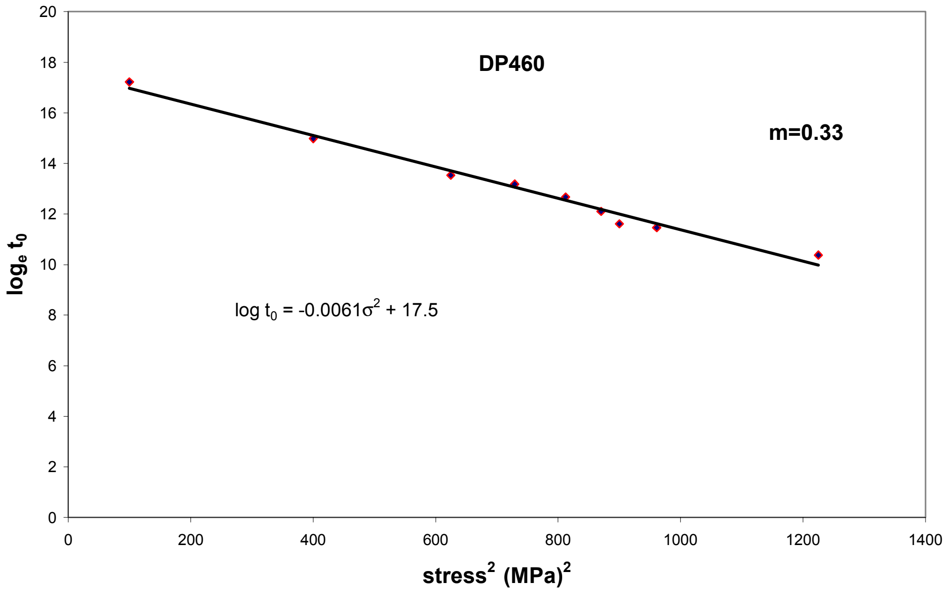

Figure 4: Mean relaxation time t_o for DP460 as a function of creep stress

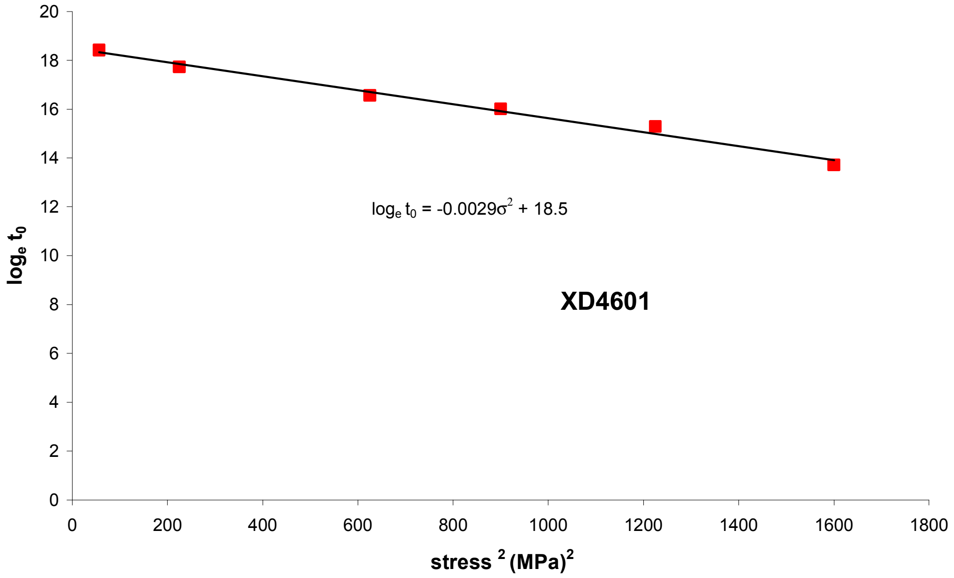

Figure 5: Mean relaxation time t_o for XD4601 as a function of creep stress

Table 1: Non-Linear Creep Model Parameters for DP460 and XD4601

	DP460	XD4601
D_o (GPa^{-1})	0.44	0.34
m	0.33	0.20
A (s)	4.0×10^7	10.8×10^7
α (MPa^{-2})	0.0061	0.0029
λ	1.7	N/A

It should be noted that, although creep behaviour can be modelled to satisfactory accuracy using constant values for the model parameters shown in Table 1, small dependencies of $\mathbf{D_o}$ on stress and of $\mathbf{t_o}$, and hence **A** and $\boldsymbol{\alpha}$, on the physical age of the adhesive are evident in experimental data.

4 NON-LINEAR CREEP UNDER MULTIAXIAL STRESSES

As previously mentioned, creep behaviour of viscoelastic materials is not only dependent on stress level, but also the stress state. Figure 6 compares creep compliance curves for DP460 obtained under uniaxial tensile and compressive stresses of 25 MPa. It can be seen clearly that there are significant differences in deformation rate with the material deforming more rapidly with time under tension (i.e. reduction in relaxation time $\mathbf{t_o}$ is less under compression than under tension) - see also [5].

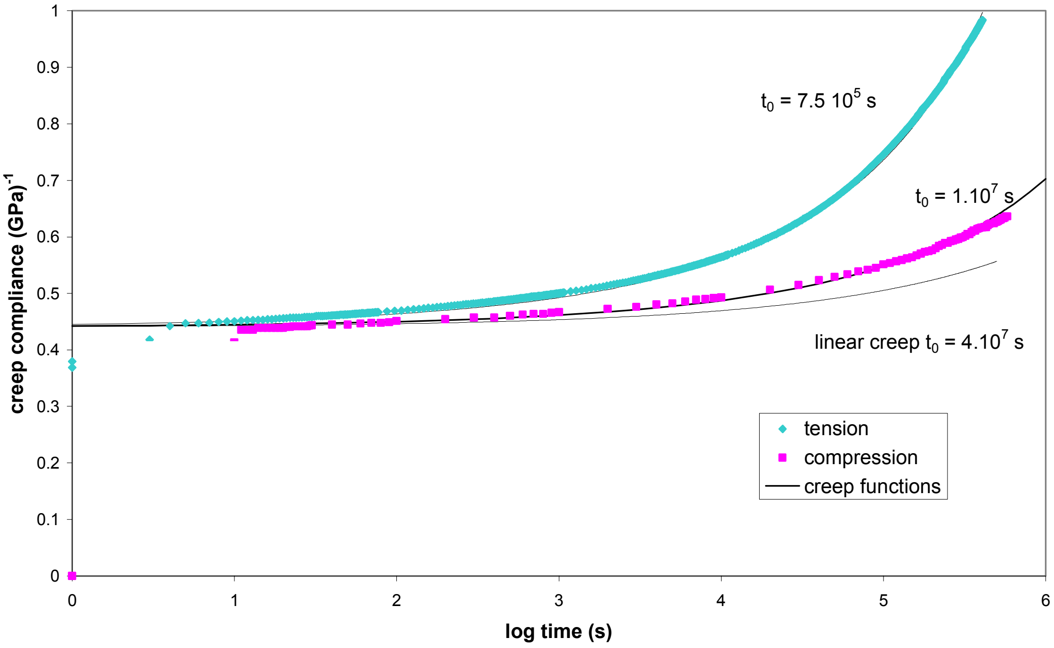

Figure 6: Tensile and compressive creep data for DP460 at a stress of 25 MPa (Modelled using different relaxation times t_o)

Also shown for comparison is the predicted curve for a low stress where tensile and compressive behaviour is expected to be the same. The results indicate that the stress in Equation (7) should be replaced by an effective stress $\bar{\sigma}$ that is a function of both the shear and hydrostatic components of the creep stress. The simplest function to consider is:

$$\bar{\sigma} = \frac{(\lambda+1)}{2\lambda}\sigma_e + \frac{3(\lambda-1)}{2\lambda}\sigma_m \tag{8}$$

where σ_e is the effective shear stress given, in terms of principal components of the applied creep stress, by:

$$\sigma_e = \left[\frac{1}{2}\left[(\sigma_1-\sigma_2)^2+(\sigma_2-\sigma_3)^2+(\sigma_1-\sigma_3)^2\right]\right]^{1/2} \tag{9}$$

and σ_m is the hydrostatic component of the creep stress given by:

$$\sigma_m = \frac{1}{3}(\sigma_1+\sigma_2+\sigma_3) \tag{10}$$

σ_1 , σ_2 and σ_3 are the principal stresses and λ is a measure of the sensitivity of the mean creep relaxation time to the hydrostatic component of the stress.

Combining Equations (7) and (8) gives:

$$t_o = A\exp -\alpha\bar{\sigma}^2 \tag{11}$$

Under tensile creep stress σ_o:

$$\sigma_e = \sigma_o \text{ and } \sigma_m = \sigma_o/3 \text{ so } \bar{\sigma} = \sigma_o. \tag{12}$$

Under compressive creep stress σ_c:

$$\sigma_e = \sigma_c \text{ and } \sigma_m = -\sigma_c/3 \text{ so } \bar{\sigma} = \frac{1}{\lambda}\sigma_c \tag{13}$$

For a compressive creep stress σ_c of 25 MPa, the t_o for DP460 is 1.10^7s (based on the data shown in Figure 6), which gives a sensitivity value of λ of 1.7 when substituted into Equation (11) - see also Table 1.

5 A CREEP FUNCTION FOR TIME-DEPENDENT PLASTICITY

Non-linear creep is generally modelled in FEA by time-dependent plasticity. A creep strain function that is commonly used to model non-linear creep in metals at elevated temperatures (and included in FEA packages) is:

$$\varepsilon(t) = \varepsilon_o + C\,\sigma_e^{\,n}\, t^{m_{eff}} \tag{14}$$

C, **n** and **m** are material parameters. This relationship, however, has been shown to be unsatisfactory for modelling creep behaviour of adhesives (see [2]).

A generalised creep function used in FEA packages, such as ABAQUS, arising from flow by rate-dependent plasticity takes the form:

$$\dot{\varepsilon}_{ij}(t) = \dot{\varepsilon}_s \left(\frac{\partial \sigma_e}{\partial \sigma_{ij}} \right) + \dot{\varepsilon}_{sw}\, \delta_{ij} \tag{15}$$

This expression can be derived from the flow law in a model for deformation by plasticity where the flow potential has contributions from the shear and hydrostatic stress given by the linear Drucker-Prager model [6]:

$$F = \sigma_e + \mu \sigma_m \tag{16}$$

where μ is the flow parameter. The terms $\dot{\varepsilon}_s$ and $\dot{\varepsilon}_{sw}$ can then be associated with contributions to the creep strain rate arising from shear and dilatational (swelling) flow processes.

With reference to Equation (15), it can be shown that:

$$\frac{\partial \sigma_e}{\partial \sigma_{ij}} = \frac{3}{2\sigma_e}\left(\sigma_{ij} - \sigma_m\, \delta_{ij} \right) \tag{17}$$

Substituting this into Equation (15) gives:

$$\dot{\varepsilon}_{ij}(t) = \dot{\varepsilon}_s\, \frac{3\sigma_{ij}}{2\sigma_e} + \left(\dot{\varepsilon}_{sw} - \frac{3\sigma_m}{2\sigma_e} \right) \delta_{ij} \tag{18}$$

The creep function for adhesives, Equation (4), can be generalised to describe multiaxial creep and takes a form similar to Equation (18), thus:

$$\dot{\varepsilon}_{ij}(t) = \left((1+\nu) D_o\, \sigma_{ij} - 3\nu\, D_o\, \sigma_m\, \delta_{ij} \right) \frac{m\, t^{m-1}}{t_o^{\,m}}\, \exp\left(\frac{t}{t_o} \right)^m \tag{19}$$

where ν is Poisson's ratio assumed to be independent of time, and $\mathbf{t_o}$ is given by Equation (11).

This can be identified with Equation (18) if:

$$\dot{\varepsilon}_s = \frac{2\sigma_e}{3}(1+\nu)\, D_o \,.\, \frac{m}{{t_o}^m} \,.\, t^{m-1} \exp\left(\frac{t}{t_o}\right)^m \tag{20}$$

and

$$\dot{\varepsilon}_{sw} = \frac{\sigma_m\, m}{{t_o}^m}\,(1-2\nu) D_o\; t^{m-1} \exp\left(\frac{t}{t_o}\right)^m \tag{21}$$

where t_o is a function of σ_e and σ_m given by Equations (11) and (8). The model parameters in these equations have been determined experimentally and are shown in Table 1.

Use of this model in a stress analysis of an adhesively bonded joint would reveal a redistribution of stress and strain levels through the bond with time under load resulting from non-linear creep in the adhesive. Appendix 2 considers FEA of a single-lap joint under creep loading.

6 LONG-TERM FAILURE UNDER CREEP LOADING

Creep rupture will occur if sufficient load is applied (or time allowed). In the case of bulk adhesive tensile specimens there is often a rapid increase in strain prior to failure. In tests conducted on DP460 and XD601 epoxy adhesives [2] where failure was obtained, an air bubble was observed in the fracture surface. These are presumably the sites of fracture initiation arising from raised stress and strain levels in the vicinity of air bubbles. Failure at different levels of measured creep strain can probably be explained by the stress and strain concentrating effect of air bubbles presumably of different size and location in each specimen. The observation that some specimens were able to sustain high measured creep strains without failure (comparable with strain levels in short-term tests under monotonic loading) suggests that there were no bubbles of significant size present in these specimens.

Although the results were not sufficiently comprehensive to enable conclusions to be drawn regarding a viable criterion for failure of tough adhesives under long-term loading, the results indicate that a strain-based criterion warrants further investigation. The stress and strain distributions around a bubble at the instant of failure will be estimated using the creep model developed at NPL in conjunction with FEA. The creep analysis would aim to predict the evolution of strain with time under load in a zone of cavitated material (as in the case of rubber-toughened epoxies) around a bubble. A critical strain failure criterion would then be consistent with a failure mechanism involving the growth and coalescence of cavities in this zone to some critical level necessary for failure.

Previous work relating to bulk and joint specimens of toughened adhesives under short-term, monotonic loading indicated a critical level of the hydrostatic or maximum principal stress as the most plausible criterion to explain failure initiation in bonded joints. This was an unexpected conclusion since a critical strain criterion would be considered more reasonable for a tough material that exhibits significant plastic deformation under essentially constant stress prior to failure.

The presence of defects tends to lend itself to a fracture mechanics approach with crack growth occurring when a critical fracture energy value is exceeded. Critical fracture energy may be time-dependent, and hence a more appropriate approach for predicting crack propagation and failure under creep loading conditions.

It may be however, that failure in some of the joint tests originates not from rupture of the adhesive (a cohesive failure), but by loss of adhesion at the adhesive-adherend interface. Since the sites for stress and strain concentration in the bonded joints tended to be located at the interface, it is difficult to distinguish between adhesion and cohesive failure mechanisms. Efforts were made to ensure that the surface treatment was conducive to cohesive failure of the adhesive. This may not have precluded the possibility of failure occurring close to the interface (i.e. interphase).

7 CONCLUSIONS

The creep behaviour of thermoplastics and structural adhesives will need to be modelled using an exponential function of time, which involves a parameter that represents a mean relaxation time for the creep process. The relaxation time depends on:

- Stress magnitude
- Stress state
- Physical age of the adhesive

Indications are that creep behaviour of these materials cannot be described accurately by any of the creep models available in FEA packages, such as ABAQUS. It is however possible to represent the model for creep in adhesives in a form that is similar to the formulation in ABAQUS for time-dependent plasticity. A subroutine has been written to implement this representation.

Creep results from tests conducted on bulk polymer specimens (i.e. toughened epoxy) reveal that the existing creep models tend to underestimate creep compliance at high stresses. It is highly probable that the creep description breaks down at higher stresses due to additional processes, such as cavitation and crazing, which occur when the stress exceeds a critical level, which is less than the yield stress of the material. In practical situations where a component or structure is expected to sustain loads for long periods with minimal deformation, the stress should be less than the critical stress. The formation of either cavities or crazes can be expected to contribute to accelerating the creep behaviour at these elevated stresses, which will require an extension of the creep model.

ACKNOWLEDGEMENT

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APPENDIX 1: PHYSICAL AGEING

Physical ageing in glassy polymers occurs after the polymer is cooled to temperatures below **T_g**. At an elevated temperature, where a polymer is in the rubbery state, the structure of the material, as determined by molecular conformations, is in equilibrium. As the temperature is cooled through the glass transition temperature, conformational changes that are needed to maintain an equilibrium structure are restricted by the increase in the relaxation times (reduced mobility) of molecular rearrangements at the lower temperatures. These non-equilibrium structures have a relatively high mobility to relaxation processes under creep loading, and this gives rise to relatively rapid creep at short elapsed times after cooling.

Despite the low temperature of the glassy polymer, there is sufficient molecular mobility for structural changes to take place with subsequent elapsed time (physical ageing) leading to structural states that become progressively closer to equilibrium for the low temperature. These ageing processes give rise to a reduction in molecular mobility under creep and a shift in creep curves to longer creep times as shown in Figure A1.1. Figure A1.1 shows the effect of different states of physical ageing on the creep compliance of DP460 with time under a stress of σ_o = 6.6 MPa. For this material, the age state is determined by the elapsed time **t_e** after cooling from 100°C (post-cure temperature).

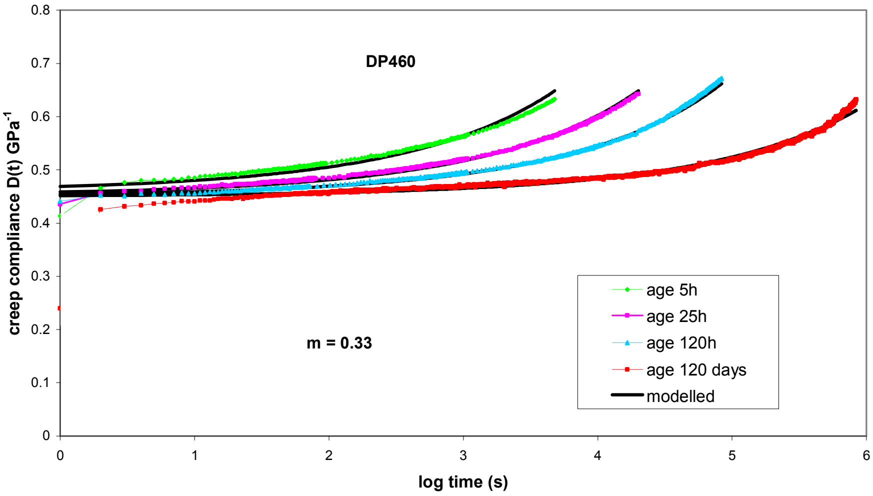


Figure A1.1: Creep compliance curves for DP460 at σ_o = 6.6 MPa and at different states of physical ageing

The tensile creep behaviour shown in Figure A1.1 can be modelled using Equation (5). The continuous lines are best fits to data at each elapsed time. The only parameter to change in each curve is the mean relaxation time **t_o**. Figure A1.2 shows a plot of **$\log t_o$** against **$\log t_e$** and demonstrates that the increase of **t_o** with physical age can be expressed by the following equation [2]:

$$\mathbf{t_o = B\, t_e^{\mu}} \tag{A1.1}$$

The values of μ and **B** are experimentally derived from creep tests conducted at different ageing times. For DP460, μ = **0.87** and **B = 21.9 $s^{0.13}$**.

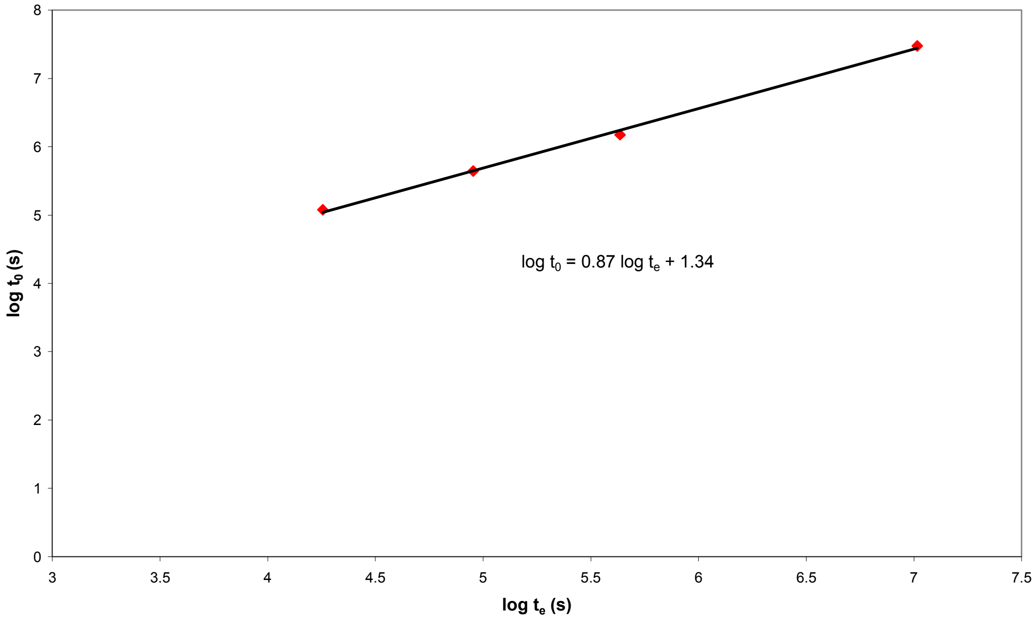


Figure A1.2: Relaxation time t_o for DP460 versus ageing time t_e after curing

Equation (A1.1) shows that changes in $\mathbf{t_o}$ with time become less as the physical age of the polymer increases. The implication is that for tests conducted 3-6 months after the material has been manufactured, changes in $\mathbf{t_o}$ due to physical ageing will be small so the effects of physical ageing can be neglected in the analysis of creep behaviour for test durations around a month. NPL Measurement Good Practice Guide No 2 [7] provides guidance on creep testing and de-ageing of polymeric materials of unknown thermal history.

The question arises as to the effect of different states of physical ageing on creep behaviour under predominantly compressive or shear stresses or other multi-axial stress states (e.g. in bonded joints). For PVC, the influences of elapsed time and elevated stress on creep rates follow the same trends for both tensile and compressive tests [5]. Compliance curves shift to longer times with increasing age and to shorter times with increasing applied stress. The shift with stress is however significantly smaller in compression than in tension. There was no indication of differences due to age on the creep compliance under either tensile or compressive stresses for PVC.

Other authors [4, 8, 9] have developed creep models to accommodate physical ageing for polymeric materials similar to the modelling described above. These have been employed with varying degrees of success, particularly at high stresses.

APPENDIX 2: FEA OF A BONDED LAP JOINT UNDER CREEP LOADING

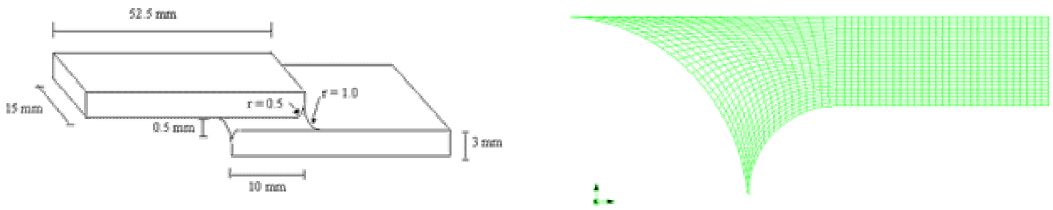
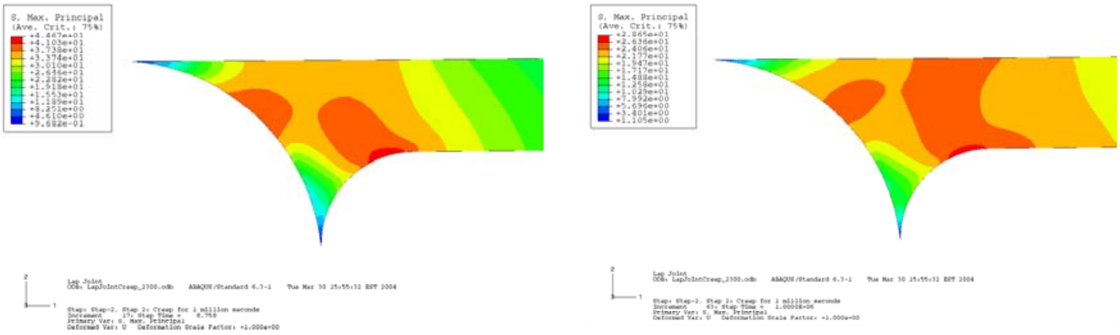


Figure A2.1: Lap joint and mesh used for the fillet region of the adhesive

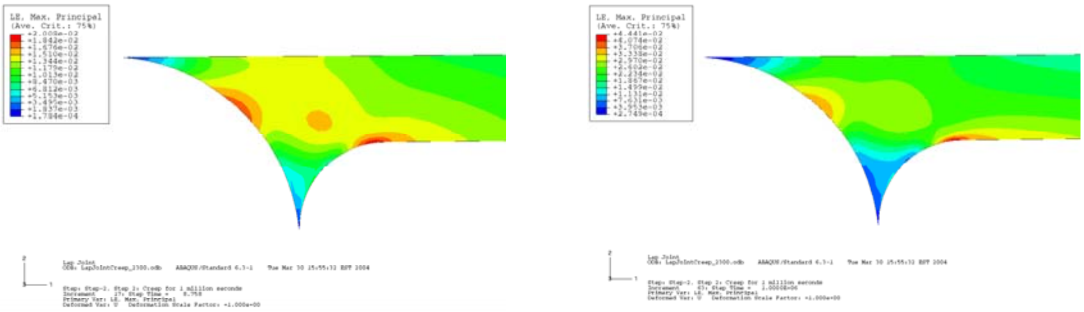
A creep analysis has been carried out to determine the evolution of stress and strain distributions with time under load in a lap joint bonded with the adhesive DP460. Equation (25) has been implemented as a user-defined materials model (UMAT) within the FEA package ABAQUS. The lap joint mesh was created using linear plain strain elements. The mesh was highly refined in the region of high stresses and strains (i.e. fillet region of the adhesive). Specimen geometry and dimensions, and the element mesh used in the analysis are shown in Figure A2.1. The analysis considers a load of 2300 N applied to the lap joint for a period of 10^6 seconds. Changes in the distribution of the maximum principal stress near the ends of the adhesive layer after 10 seconds and 10^6 seconds under load are shown in Figure A2.2.



10 seconds **10^6 seconds**

Figure A2.2: Maximum principal stress contours in the joint at a load of 2300N

These results show a reduction in the level of stress in the adhesive in the region of stress concentration, which is attributable to the non-linear creep behaviour of the adhesive. In contrast, Figure A2.3 shows the evolution of the maximum principal strain in the same region of the adhesive. The strain can be seen to increase significantly with time especially in the region of strain concentration.



10 seconds **10^6 seconds**

Figure A2.3: Maximum principal strain in the joint at a load of 2300N

The non-linear creep behaviour of adhesives shown in Figures 2 and 3 (see Section 3) can be displayed as a series of tensile stress against tensile strain values corresponding to different times under load (known as isochronous curves). Isochronous curves can be generated for both tension and compression, providing long-term (10^6 s) stress/strain data for use with FEA (see [2]). In order to carry out FEA of bonded joints using isochronous data, the plastic strain hardening curves for tension and compression need to be generated. The compression curve is then used to derive the hydrostatic stress sensitivity parameter $\mu = \tan\beta$ in the linear Drucker-Prager model. A value for the parameter μ can be calculated from values for the tensile and compressive stresses, σ_o and σ_c respectively, at the same equivalent plastic strain using (see previous sections):

$$\mu = \tan\beta = \frac{3(\sigma_c - \sigma_o)}{\sigma_c + \sigma_o} \tag{A2.1}$$

Equivalent plastic creep strains ε^P and ε_c^P in tension and compression are related by the equation:

$$\sigma_o \, \varepsilon^P = \sigma_c \, \varepsilon_c^P \tag{A2.2}$$

Values for μ obtained from Equations (A2.1) and (A2.2) increase slowly with plastic strain. This variation of μ with plastic strain and the associated uncertainty with the derivation of a value for μ illustrates that any attempt to model long-term behaviour in adhesives by an elastic-plastic analysis is only approximate.