

**Modelling Creep in
Toughened Adhesives
for Finite Element
Analysis**

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SUMMARY

A model is introduced for describing the creep behaviour of glassy adhesives. Its application is illustrated using measurements on two rubber-toughened epoxy adhesives. The dependence of creep compliance on applied stress (non-linear creep behaviour) is explained by making the creep relaxation time dependent on the stress level. Creep behaviour under uniaxial tension and uniaxial compression is observed to be different, and this is explained through a dependence of creep relaxation time on an effective stress which is a function of the multiaxial stress state.

Attempts have been made to implement the model in the finite element system ABAQUS so that changes can be investigated in the stress and strain distributions in a bonded joint with time under load. None of the available models can be used for this purpose, but the adhesives creep model can be represented by a generalised model in ABAQUS for time-dependent plasticity. This has enabled changes in distributions of stress and strain in the adhesive in a lap joint to be calculated with approximate values calculated using long-term, isochronous data with an elastic-plastic materials model.

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1 INTRODUCTION

Previous studies on adhesives at NPL have shown how finite element analysis can be used to predict extensions in an adhesively bonded joint that is under an applied load, as well as distributions of stress and strain within the adhesive layer [1]. The purpose behind these studies has been to improve the use of finite element analysis as a tool for designing bonded joints. The results of an analysis can be used to explore variations in the design of the joint in order to reduce stress or strain levels in regions of stress concentration. In conjunction with a valid failure criterion [2], it should be possible to predict the ultimate load that the joint can sustain. Confidence in predictions requires the use of models that accurately describe the deformation behaviour of the adhesive as well as accurate property data for the adhesive. Emphasis has therefore been given in earlier studies to the evaluation of models in finite element packages for describing the non-linear and strain-rate-dependent deformation behaviour of toughened adhesives. Test methods were also developed for characterising deformation behaviour and for determining the properties and parameters required by the models.

This work has demonstrated that the mechanical properties of adhesives vary with strain rate or time under load as expected for viscoelastic materials. This means that under long-term loading, properties will decrease progressively with time, and stress and strain levels in the adhesive will be very different from predictions based on a short-term analysis under monotonic loading. In addition, criteria for failure of the adhesive or parameters in these criteria will probably depend upon factors such as the load history or the time under load.

In order for a stress analysis to assist with the design of bonded joints in these situations, models are needed that describe deformation of the adhesive under creep, intermittent or fatigue loads. The work reported here is the first stage of such a programme. It considers deformation and failure under a long-term load of constant magnitude (creep). Measurement and modelling have been carried out of the creep behaviour of two toughened epoxy adhesives. The onset of non-linear behaviour in creep tests is observed at somewhat lower stresses than obtained in previous work in short-term tests under monotonic loading. The non-linearity arises because of a progressive reduction, with stress level, in the relaxation times of

the molecular relaxation processes associated with the creep deformation. The reduction in relaxation times is related to the stress state as well as the stress magnitude.

This behaviour cannot be modelled using available non-linear creep models in finite element packages so a new model has been developed. An attempt has been made to implement this with the solver in ABAQUS through the preparation of a user-defined subroutine for application with a generalised model in ABAQUS for time-dependent plasticity. This has enabled changes in distributions of stress and strain in the adhesive in a lap joint to be calculated with time under load. These results are compared with approximate values calculated using long-term, isochronous data with an elastic-plastic materials model.

2 MATERIALS

Creep studies have been carried out on two rubber-toughened epoxy adhesives. One material is the one-part adhesive XD 4601 supplied by Dow Plastics. The other is a two-part system with the designation DP460 supplied by 3M Ltd. Sheets of bulk resin with a thickness of approximately 3 mm were cast in a mould. The one-part material was cured at a temperature of 180 °C for 1 hour. The two-part adhesive was left for 24h at a temperature of 23 °C and subsequently post-cured at 100 °C for 30 minutes. Standard tensile specimens [3] were cut from the sheets for the measurement of creep behaviour under uniaxial tensile stress.

3 MODELLING LINEAR CREEP AND PHYSICAL AGEING

The time-dependent, viscoelastic behaviour of polymeric materials may be modelled by combinations of spring and viscous dashpot elements in series and parallel. The simplest model that shows both a short-term, elastic or unrelaxed response as well as a long-term, limiting deformation corresponding to a fully relaxed state is shown in figure 1. For a model consisting of the 3 elements E_0 , E_1 and η_1 , the time-dependent strain response $\epsilon(t)$ to a constant stress σ_0 is

$$\epsilon(t) = \frac{\sigma_0}{E_0} + \frac{\sigma_0}{E_1} \left(1 - \exp \left(-\frac{t}{\tau_1} \right) \right) \quad (1)$$

where the relaxation time τ_1 is given by

$$\tau_1 = \frac{\eta_1}{E_1} \quad (2)$$

This single relaxation time model will not describe actual relaxation processes in polymers which have a very broad distribution of relaxation times. Figure 1 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 now to an applied stress is

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

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. Creep strains can then be described by the more simple expression [4]

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

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, as long as the measurement temperature is not close to the glass transition temperature. 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

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

The magnitude of the parameter t_0 depends on temperature, stress level and stress state. The dependence on stress level gives rise to non-linear creep behaviour and is described in the next section. The magnitude of t_0 also depends on the state of physical ageing of the adhesive at the time of the creep loading [4]. Physical ageing in glassy polymers occurs after the polymer is cooled to temperatures below the rubber-to-glass transition. 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 observed in figure 2. The figure shows experimental data for the creep compliance of the adhesive DP460 with time under a stress of $\sigma_0 = 6.6$ MPa at different states of physical ageing. The age state is determined by the elapsed time t_e after cooling from the post-cure temperature of 100 °C.

The creep behaviour shown in figure 2 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_0 . Figure 3 shows a plot of $\log t_0$ against $\log t_e$ and demonstrates that the increase of t_0 with physical age can be expressed by the equation

$$t_0 = B t_e^\mu \quad (6)$$

where, for DP460, $\mu = 0.87$ and $B = 21.9 \text{ s}^{0.13}$.

Equation (6) shows that changes in t_0 with time become less as the physical age of the adhesive increases. The creep tests described in this section were carried out on specimens that were about 120 days old. The duration of these tests was generally around 10 days or

less. Over this time period, changes in t_0 due to physical ageing will be small so the effects of physical ageing can be neglected in the analysis of creep behaviour.

4 MODELLING NON-LINEAR CREEP UNDER MULTIAXIAL STRESSES

4.1 NON-LINEAR CREEP BEHAVIOUR

Figure 4 shows creep compliance curves for the adhesive DP460 measured under different levels of stress σ_0 . At short creep times, there is a small dependence of the compliance on stress which is consistent with slight curvature in tensile stress/strain curves obtained from constant displacement rate tests at speeds of typically 10 mm/min. This non-linear behaviour is observed to increase significantly with time under load, the limiting stress for linear behaviour being near or below 10 MPa. The effect of stresses above this level is to give a significant reduction in the mean relaxation time t_0 of the creep process and to, thereby, shift curves to shorter times. This can be demonstrated by using equation (5) to obtain best fitting curves to the data in figure 4. The continuous curves in the figure have been obtained with constant values for $D_0 = 0.44 \text{ GPa}^{-1}$ and $m = 0.33$ and selecting values for t_0 that decrease with increasing stress. The variation of t_0 with σ_0 can be described with satisfactory accuracy by the empirical relationship

$$t_0 = A \exp -\alpha \sigma_0^2 \quad (7)$$

as demonstrated by the data for DP460 in figure 5. The parameters A and α in equation (7) can be derived from this figure.

Creep compliance curves showing the non-linear creep behaviour of the adhesive XD4601 under uniaxial tension are shown in figure 6. The variation of the mean relaxation time t_0 for these curves with tensile stress σ_0 is shown in figure 7 and follows the same relationship equation (7) as seen with the adhesive DP460. Values for the parameters in the non-linear creep model represented by equations (5) and (7) are listed in table 1 for the adhesives DP460 and XD4601.

Table 1 Values for parameters for the adhesives DP460 and XD4601 in the model for non-linear creep in adhesives

	DP460	XD4601
D_o (GPa ⁻¹)	0.44	0.34
m	0.33	0.2
A (s)	4.10^7	$10.8 \cdot 10^7$
α (MPa ⁻²)	0.0061	0.0029
λ	1.7	

It should be noted that, although creep behaviour can be modelled to satisfactory accuracy using constant values for the model parameters above, small dependencies of D_o on stress and of t_o , and hence A and α , on the physical age of the adhesive are evident in experimental data.

4.2 EXTENSION OF THE MODEL TO CREEP UNDER MULTIAXIAL STRESSES

Equations (5) and (7) describe creep behaviour under a uniaxial tensile stress. In this section, modifications to the model are proposed that will enable creep behaviour to be described under an arbitrary stress state. Figure 8 compares creep compliance curves for DP460 obtained under uniaxial tensile and compressive stresses of 25 MPa. At low stresses where creep behaviour is linear, compliance curves in tension and compression are expected to be the same. Under the higher stress of 25 MPa where behaviour is non-linear, the results in figure 8 show that the reduction in relaxation time t_o is less under compression than under tension and hence that it is not only the magnitude of the stress that influences t_o but the stress state also. 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

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

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

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

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

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

The parameter λ is a measure of the sensitivity of the mean creep relaxation time to the hydrostatic component of stress.

Combining equations (7) and (8) gives

$$t_o = A \exp -\alpha \bar{\sigma}^2 \quad (11)$$

Thus, under a tensile creep stress σ_o , $\sigma_e = \sigma_o$ and $\sigma_m = \sigma_o/3$ so, from equation (8), $\bar{\sigma} = \sigma_o$.

Under a compressive creep stress σ_c , $\sigma_e = \sigma_c$ and $\sigma_m = -\sigma_c/3$, so $\bar{\sigma} = \frac{1}{\lambda} \sigma_c$. Under the stress $\sigma_c = 25$ MPa, the results of figure 8 show that t_o for the compressive test is 1.10^7 s. Using this value in equation (11) gives $\lambda = 1.7$.

5 APPLICATION OF THE CREEP MODEL IN A FINITE ELEMENT ANALYSIS

5.1 GENERAL

In order to predict stress and strain distributions in an adhesive joint under long-term loading, it is necessary to implement the creep model described in sections 3 and 4 in a finite element system. The most rigorous method to achieve this is through the development of software for a user-defined materials model (UMAT) for application with the solver in a finite element system. Time limitations in Project MMS11 have prevented the use of this approach for the work reported here, and so methods for carrying out approximate analyses are considered next. In section 5.2, the use of available non-linear creep models in the finite element system ABAQUS is explored for describing the type of creep behaviour observed in toughened

adhesives. These models have been developed for creep in metals at high temperatures and are shown in section 5.2 to be unsatisfactory for adhesives. In section 5.3, the use of a generalised model for time-dependent plasticity in ABAQUS is interpreted by means of a user-defined subroutine to represent the equations developed in sections 3 and 4 to model non-linear viscoelasticity of adhesives.

Section 5.4 considers the use of isochronous stress/strain data in an elastic-plastic analysis to enable calculations to be carried out of stresses and strains in the adhesive at some specified time. This analysis uses data (isochronous) corresponding to the specified time under a constant load, but the analysis is unable to account for any evolution of stress and strain distributions with time under load by virtue of the time-dependent deformation behaviour of the adhesive. The results of finite element analyses of the deformation of a lap joint under a sustained load using the methods described in sections 5.3 and 5.4 are compared in section 6.

5.2 MODELLING CREEP IN ADHESIVES USING AVAILABLE FUNCTIONS IN ABAQUS

A creep strain function in ABAQUS that is commonly used to model non-linear creep in metals at elevated temperatures takes the form

$$\varepsilon(t) = \varepsilon_o + C \sigma_e^n t^{m_{eff}} \quad (12)$$

where C , n and m_{eff} are material parameters.

This equation relates the time-dependence under creep to a power law in time. The creep compliance curves in figure 4 can be described with reasonable accuracy using a power law as shown in figure 9. The data in this figure have been modelled using the equation

$$D(t) = D_o \left(1 + \left(\frac{t}{t_{eff}} \right)^{m_{eff}} \right) \quad (13)$$

Note, the parameters m_{eff} and t_{eff} will be numerically different from m and t_o in equation (5).

Under a uniaxial tensile creep stress of σ_o , equation (12) becomes

$$D(t) = D_o + C \sigma_o^{n-1} t^{\text{meff}} \quad (14)$$

This can be identified with equation (13) if

$$\frac{D_o}{t_{\text{eff}}^{\text{meff}}} = C \sigma_o^{n-1} \quad (15)$$

Thus if equation (12) is to be applicable to adhesives, a plot of $\log t_{\text{eff}}$ vs $\log \sigma_o$ should be linear. This plot for the adhesive DP460 is shown in figure 10. It is clear that these results cannot be approximated by a linear plot, and we can conclude that the creep law in equation (12) is not applicable to adhesives.

5.3 A CREEP FUNCTION FOR TIME-DEPENDENT PLASTICITY

A generalised creep function in ABAQUS arising from flow by rate-dependent plasticity takes the form

$$\dot{\epsilon}_{ij}(t) = \dot{\epsilon}_s \left(\frac{\partial \sigma_e}{\partial \sigma_{ij}} \right) + \dot{\epsilon}_{sw} \delta_{ij} \quad (16)$$

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 [1]

$$F = \sigma_e + \mu \sigma_m \quad (17)$$

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

With reference to equation (16), it can be shown that

$$\frac{\partial \sigma_e}{\partial \sigma_{ij}} = \frac{3}{2\sigma_e} (\sigma_{ij} - \sigma_m \delta_{ij}) \quad (18)$$

Substituting this into equation (16) gives

$$\dot{\epsilon}_{ij}(t) = \dot{\epsilon}_s \frac{3\sigma_{ij}}{2\sigma_e} + \left(\dot{\epsilon}_{sw} - \frac{3\sigma_m}{2\sigma_e} \right) \delta_{ij} \quad (19)$$

The creep function for adhesives, equation (4), can be generalised to describe multiaxial creep and takes a form similar to equation (19), thus

$$\dot{\epsilon}_{ij}(t) = \left((1+\nu) D_o \sigma_{ij} - 3\nu D_o \sigma_m \delta_{ij} \right) \frac{m t^{m-1}}{t_o^m} \exp\left(\frac{t}{t_o}\right)^m \quad (20)$$

where ν is Poisson's ratio assumed to be independent of time, and t_o is given by equation (11).

This can be identified with equation (19) if

$$\dot{\epsilon}_s = \frac{2\sigma_e}{3} (1+\nu) D_o \cdot \frac{m}{t_o^m} \cdot t^{m-1} \exp\left(\frac{t}{t_o}\right)^m \quad (21)$$

and

$$\dot{\epsilon}_{sw} = \frac{\sigma_m m}{t_o^m} (1-2\nu) D_o t^{m-1} \exp\left(\frac{t}{t_o}\right)^m \quad (22)$$

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.

Through equations (21) and (22), a model for non-linear viscoelasticity has been associated with a model for time-dependent plasticity. Coding for a user subroutine has been written so that equations (21) and (22) can be implemented in equation (16) in ABAQUS for the solution of creep analyses by finite element methods. The results of an analysis for a bonded lap joint are discussed in section 6.

5.4 ISOCHRONOUS STRESS/STRAIN DATA

The non-linear creep behaviour of adhesives shown in figures 4 and 6 can be displayed as a series of plots of tensile stress against tensile strain values corresponding to different times under load. These are isochronous curves and can be constructed for arbitrary values of stress using the model equations (5) and (7) which give

$$\varepsilon(t) = \sigma_o D_o \exp \left(\frac{t}{(A \exp - \alpha \sigma_o^2)} \right)^m \quad (23)$$

Using values for the model parameters given in table 1, equation (23) can be used to construct the tensile stress/strain curve shown in figure 11 for DP460 at a time under load of $t = 10^6$ s. Isochronous curves corresponding to loading under uniaxial compression can be calculated by combining equations (5) and (11) to give

$$\varepsilon_c(t) = \sigma_c D_o \exp \left(\frac{t}{(A \exp - \alpha \sigma_c^2)} \right)^m \quad (24)$$

where ε_c and σ_c are strains and stresses obtained under a compressive creep stress. The compression curve in figure 11 was calculated using equation (24).

If these data are to be used in an elastic-plastic analysis, it is necessary to present the results in figure 11 as hardening curves of stress against the plastic component of strain ε^p . These values are derived from the tensile stress and strain values in figure 11 using

$$\varepsilon^p = \varepsilon - \frac{\sigma_o}{E} \quad (25)$$

where E is Young's modulus appropriate to a loading time of 10^6 s. An analogous equation relates plastic and total strains in a compressive test. A value for $E = 1510$ MPa was obtained from the linear, small strain region of the curves in figure 11. Hardening data derived using equation (25) are shown in figure 12.

The difference between the tensile and compression curves in figure 12 demonstrate that non-linear deformation under long-term loading is very sensitive to the hydrostatic component of stress. In an elastic-plastic analysis of this non-linear behaviour, this sensitivity to hydrostatic stress can be included through the use of a model, such as the linear Drucker-Prager model, with a yield criterion that is sensitive to hydrostatic stress. In order to carry out a finite element analysis of a lap joint using isochronous data, (see section 6), the plastic strain hardening behaviour of the adhesive is defined by the tensile hardening curve in figure 12. 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 has been calculated from values for the tensile and compressive stresses, σ_o and σ_c respectively, at the same equivalent plastic strain in figure 12 using

$$\mu = \tan \beta = \frac{3(\sigma_c - \sigma_o)}{\sigma_c + \sigma_o} \quad (26)$$

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

$$\sigma_o \epsilon^p = \sigma_c \epsilon_c^p \quad (27)$$

By way of illustration, one pair of equivalent stresses is shown in figure 12 derived using equation (27).

Values for μ obtained from equations (26) and (27) increase slowly with plastic strain and reach a value of 0.6 at plastic strains of around 0.1. 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 non-linear creep in adhesives by an elastic-plastic analysis is only approximate.

6 FINITE ELEMENT ANALYSES UNDER CREEP LOADING

6.1 GENERAL

The results of finite element analyses of a lap joint of the adhesive DP460 under long-term loading are presented in section 6.3. In section 6.3.1, calculations of stress and strain distributions in the adhesive layer have been carried out using the creep model developed in this project together with its implementation in ABAQUS by means of a user subroutine described in section 5.3. Results are shown at loading times of 10s and 10^6 s to demonstrate the evolution of stress and strain distributions with time under load. These results are compared in section 6.3.2 with calculations made using an elastic-plastic model with isochronous hardening data derived using the creep model (see section (5.4)) at 10s and 10^6 s times under load. Two separate analyses are carried out for each time scale, and, as noted earlier, in each analysis the time-dependent behaviour of the adhesive is ignored.

For reasons explained in section 5.1, the first analysis above is considered to be the more rigorous. However, in order to undertake a simple evaluation of the validity of this model and the associated subroutine code, predictions have been made of the creep strain produced in a tensile specimen by step increases in the uniaxial tensile stress applied to the specimen.

6.2 CREEP OF A TENSILE SPECIMEN UNDER STEP LOADING

The stress history is shown in figure 13, and this was applied to the adhesive XD4601. Predicted strains using a finite element analysis with a single element representing the gauge region in the tensile specimen are compared with measured values in figure 14. It can be seen that the analysis predicts the strain history accurately for the initial stress application only. Significant errors are introduced in strain predictions after the application of each additional step in stress. The reason for this is presumably associated with the procedure used by the FE analysis to generate and superpose calculated strains following each application of additional stress. Thus at times $t > t_2$ corresponding to the application of the second additional stress σ_2 , the analysis appears to use the following expression to calculate the time-dependent strain.

$$\begin{aligned}
\varepsilon(t) = & D_o \sigma_o \exp \left(\left(\frac{t_1}{t_o} \right)^m + \left(\frac{t_2 - t_1}{t_o} \right)^m + \left(\frac{t - t_2}{t_o} \right)^m \right) \\
& + D_o (\sigma_1 - \sigma_o) \exp \left(\left(\frac{t_2 - t_1}{t_{o1}} \right)^m + \left(\frac{t - t_2}{t_{o1}} \right)^m \right) \\
& + D_o (\sigma_2 - \sigma_1) \exp \left(\frac{t - t_2}{t_{o2}} \right)^m \quad t > t_2
\end{aligned} \tag{28}$$

where t_o , t_{o1} and t_{o2} are the relaxation times under stresses σ_o , σ_1 and σ_2 , respectively and are given by equation (7). It can be seen that this expression does not include any changes to the relaxation time for the strain contributions from the stress σ_o on application of the additional stresses σ_1 and σ_2 nor for the strain contribution from the stress σ_1 on application of the stress σ_2 . An alternative expression, equation (29), allows the relaxation times to change corresponding to the stress level for the appropriate time interval.

$$\begin{aligned}
\varepsilon(t) = & D_o \sigma_o \exp \left(\frac{t_1}{t_o} + \frac{t_2 - t_1}{t_{o1}} + \frac{t - t_2}{t_{o2}} \right)^m \\
& + D_o (\sigma_1 - \sigma_o) \exp \left(\frac{t_2 - t_1}{t_{o1}} + \frac{t - t_2}{t_{o2}} \right)^m \\
& + D_o (\sigma_2 - \sigma_1) \exp \left(\frac{t - t_2}{t_{o2}} \right)^m \quad t > t_2
\end{aligned} \tag{29}$$

The formulation of the superposition of each strain contribution is also different in equation (29) from that in equation (28). Although the strain prediction given by equation (29) is closer to experimental values, there is still a significant departure following the application of the second increment in stress corresponding to a total stress of 36 MPa. Closer examination suggests that this is due to a temporary reduction in the relaxation time value t_{o2} below that given by equation (7). This interpretation is consistent with other creep studies under intermittent load and is the subject of further research work.

6.3 CREEP OF A LAP JOINT TEST SPECIMEN

6.3.1 A creep analysis

Despite these apparent shortcomings in the implementation of the creep model in ABAQUS, a creep analysis has been carried out using the model to determine the evolution of stress and strain distributions with time under load in a lap joint of the adhesive DP460. The material parameters for this adhesive required by the creep model are given in table 1 of section 4.1. The geometry and dimensions of the joint and the element mesh used in the analysis are shown in figure 15.

The analysis considers a load of 2300 N applied to the lap joint for a duration of 10^6 s. Changes in the distribution of the maximum principal stress near the ends of the adhesive layer after 10s and 10^6 s under load are shown in figure 16. 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 17 shows the evolution of the maximum principal strain in the same region of the adhesive. The strain is seen to increase significantly with time especially in the region of strain concentration.

6.3.2 Analyses using isochronous data

Figures 18 and 19 show predictions, analogous to figures 16 and 17, of stress and strain distributions for times of 10s and 10^6 s under load obtained using isochronous data with an elastic-plastic analysis. Since time-dependent behaviour is not considered in an analysis using isochronous data, two separate analyses were carried out with hardening data appropriate to each time under load. These hardening curves are shown in figure 12 and were derived from the creep functions as explained in section 5.4. These calculations gave values for Young's modulus of 2200 MPa and 1510 MPa, respectively, for the 10s and 10^6 s times under load. Values for Poisson's ratio = 0.41 and for the hydrostatic stress sensitivity parameter $\mu = 0.6$ (see section 5.4) were assumed to be independent of time.

The contour maps in figures 18 and 19 show stresses decreasing and strains increasing with time under load as obtained with the creep analyses (figures 16 and 17 respectively) but the levels of stress and strain are different as would be expected for an approximate analysis.

7 LONG-TERM FAILURE UNDER CREEP LOADING

Figures 20 and 21 show the results of creep tests on bulk specimens at elevated stresses leading in most cases to rupture of the specimen. Just prior to failure in some of the tests, there is a rapid increase in strain. The fall off in the creep deformation observed near the end of the test at 35 MPa in figure 20 is an artefact of this test and arose because the deformation mechanism of the creep machine reached its limit of travel. The test was continued after the limit was adjusted. The specimens in the tests at 27 MPa in figure 20 and at 37 MPa in figure 21 did not fail within the timescale of the tests. In all tests where failure occurred, 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 the bubbles.

Although these results are not sufficiently comprehensive to enable conclusions to be drawn regarding a viable criterion for failure of tough adhesives under long-term loading, they indicate that a criterion based on a critical level of a component of strain deserves further study. Whilst the results show failure at different levels of strain, this can be explained by the stress and strain concentrating effect of air bubbles of different size and location in each specimen. The observation that some specimens in figures 20 and 21 are able to sustain high strains without failure (comparable with strain levels in short-term tests under monotonic loading) suggests that there are no bubbles of significant size in these specimens. The magnitude and the state of strain around a bubble at the instant of failure could be estimated using the creep model developed in this project in conjunction with a finite element analysis that took account of the size and location of the bubble in each specimen. Although not attempted here because of time constraints, such analyses should be carried out as part of the future project on long-term failure of adhesives. A creep analysis would capture the evolution of strain with time under load in a zone of cavitated material 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.

It is appropriate at this stage to contrast these conclusions with the results of failure studies carried out earlier in this project on bulk and joint specimens of toughened adhesives under short-term, monotonic loading. In this work, conclusions were drawn that identified a critical level of the hydrostatic or maximum principal stress as the most plausible criterion to explain failure initiation in the different joint geometries. 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 in bulk specimen tests. 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 each joint geometry are located at the interface, it is difficult to distinguish between adhesion and cohesive failure mechanisms.

8 CONCLUSIONS

The creep behaviour of toughened adhesives can 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

- the stress magnitude
- the stress state
- the physical age of the adhesive

This behaviour cannot be described by any of the creep models in the finite element system ABAQUS. It is however possible to represent the model for creep in adhesives to appear similar to a formulation in ABAQUS for time-dependent plasticity. A subroutine must be written to implement this representation.

A finite element analysis of creep in a lap-joint specimen reveals that the distribution of stress in the adhesive layer decreases with time under load whilst the strain distribution increases with time.

A similar conclusion is obtained using an elastic-plastic analysis with hardening behaviour generated using isochronous stress/strain curves. The redistributions of stress and strain levels in the adhesive obtained by this analysis are qualitatively similar to those obtained using a creep analysis, but the derived levels of stress and strain for a given time under load are different.

9 ACKNOWLEDGEMENT

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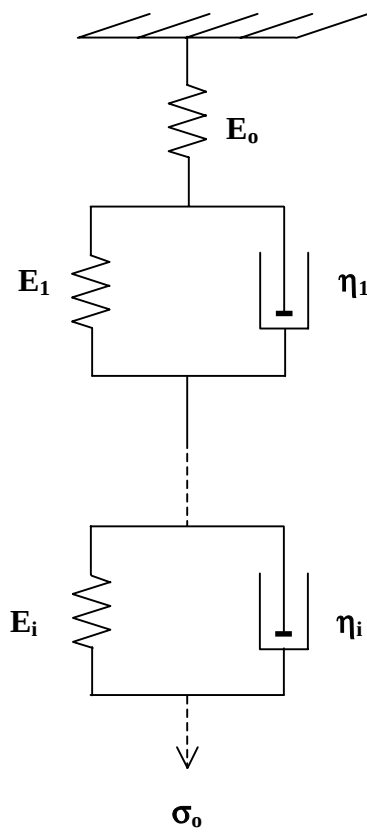


Figure 1 A spring and dashpot model for linear creep in polymers.

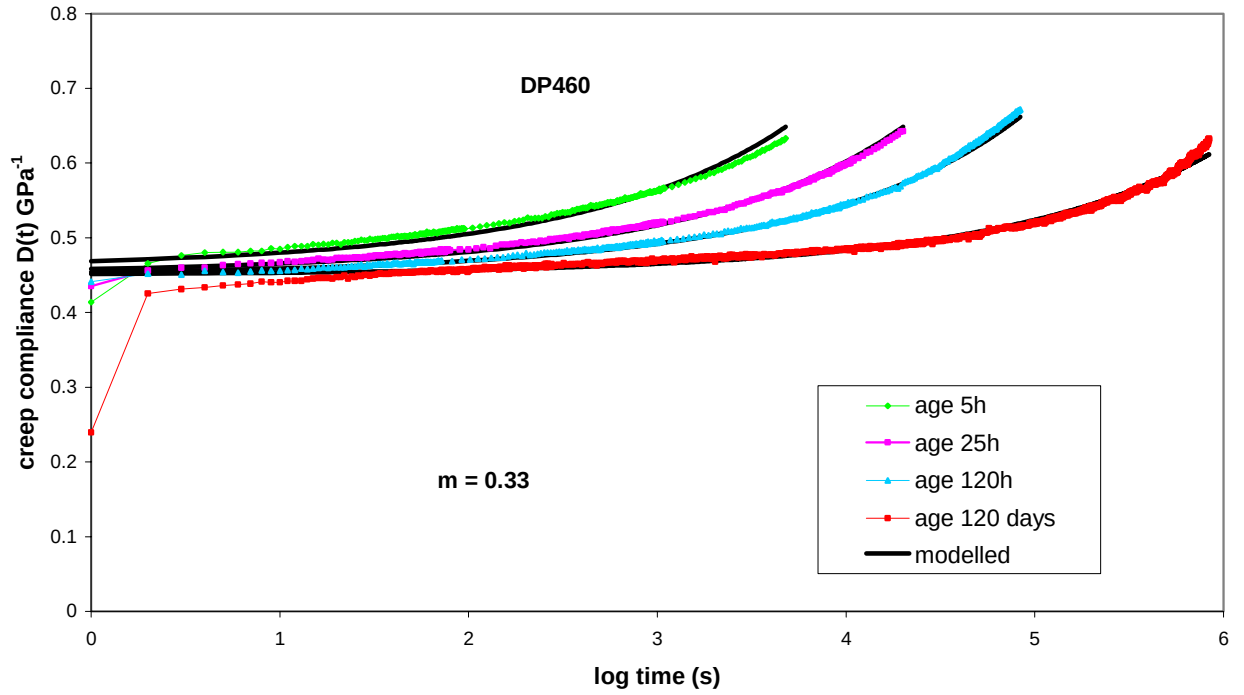


Figure 2 Creep compliance curves for DP460 at a stress $\sigma_0 = 6.6$ MPa and at different states of physical ageing.

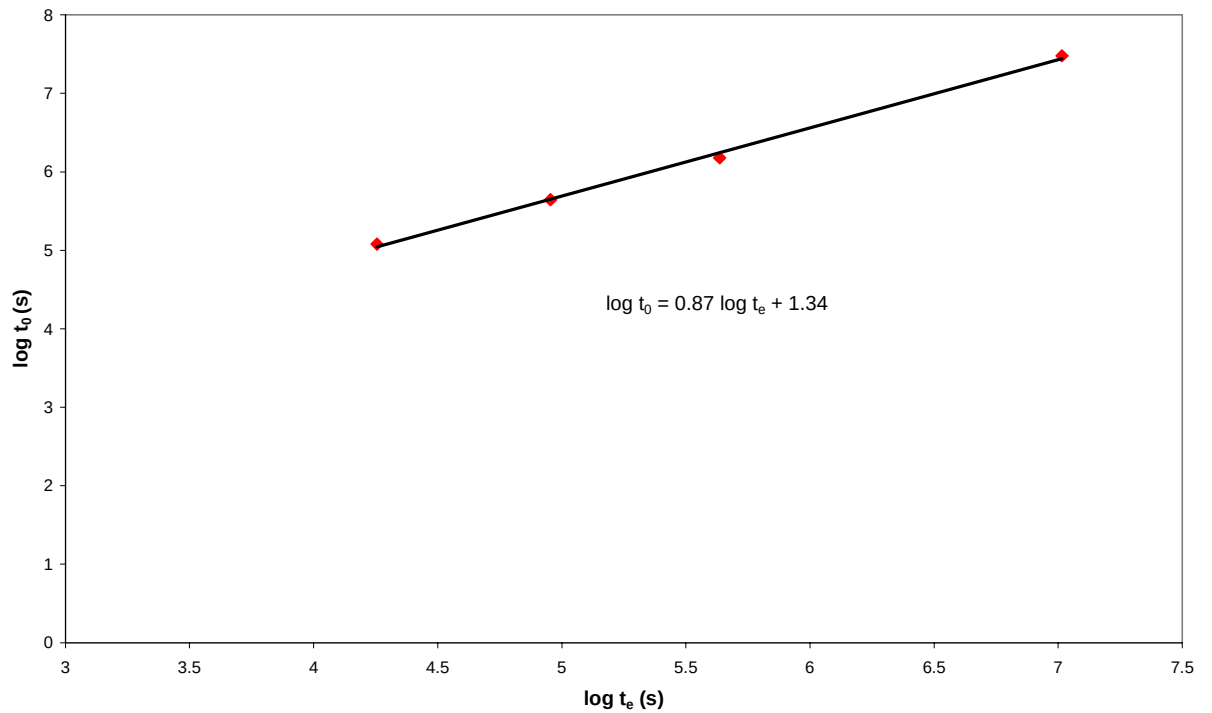


Figure 3 Variation of mean relaxation times t_0 for DP460 with ageing time t_e after curing

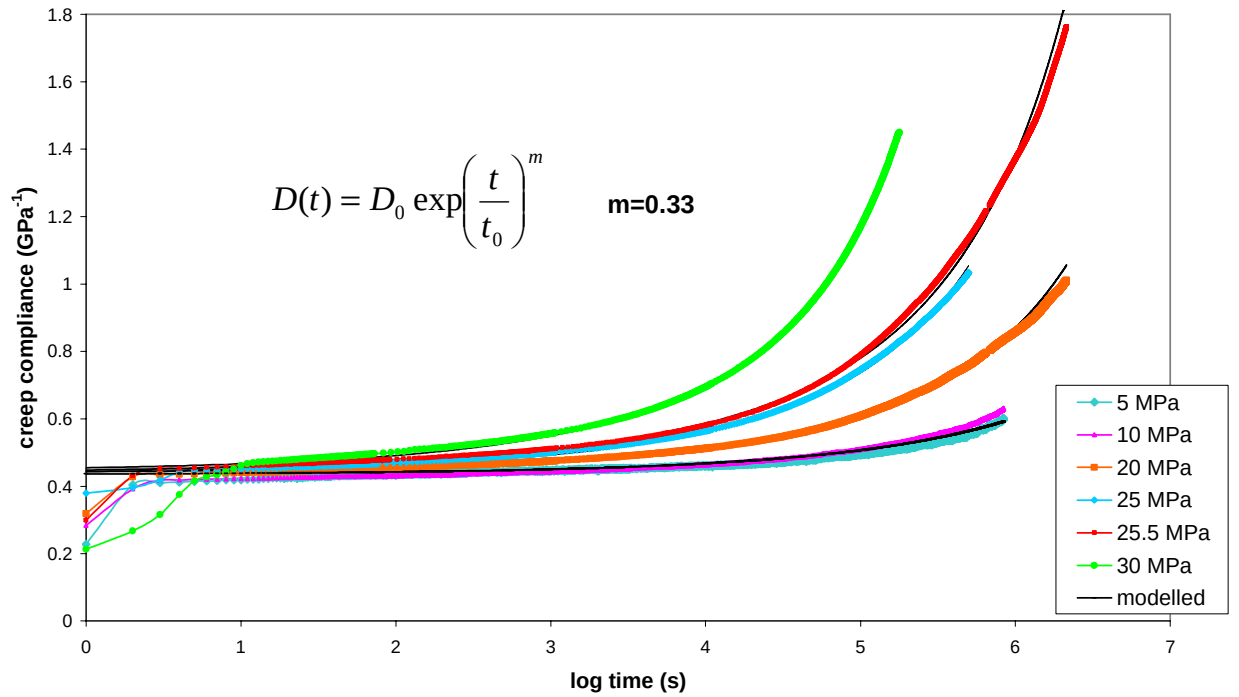


Figure 4 Creep compliance curves for DP460 at different levels of stress

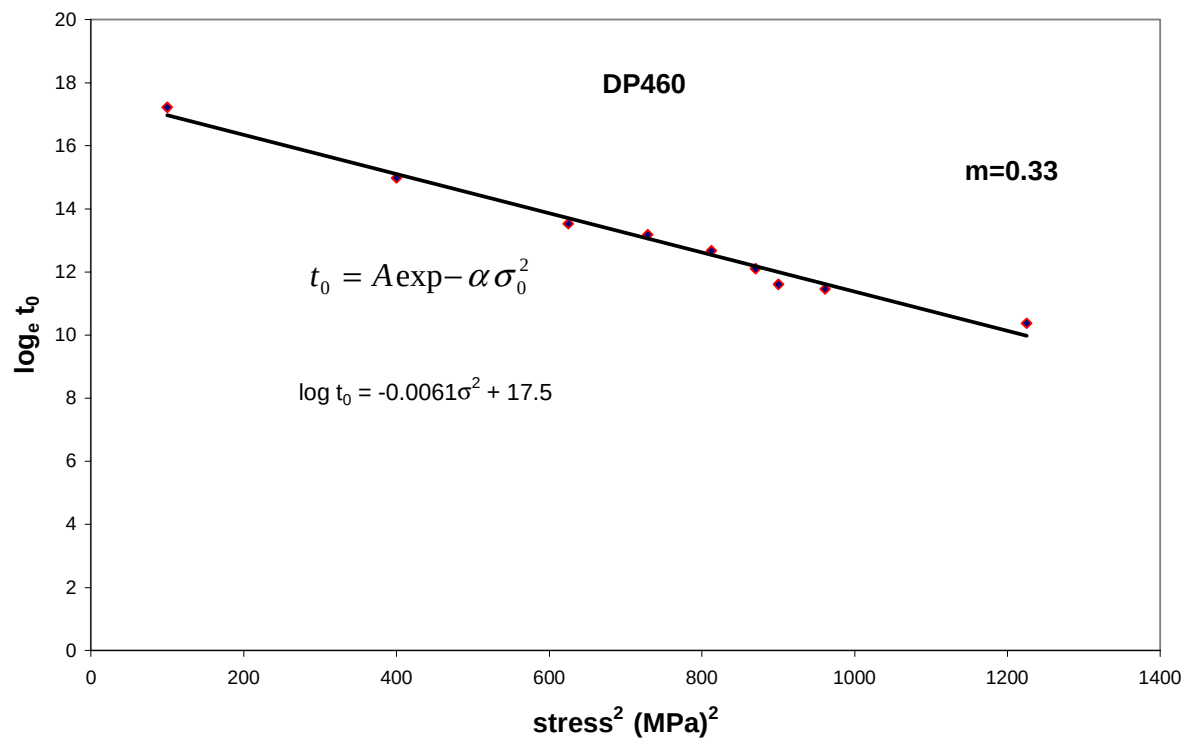


Figure 5 Variation of the mean relaxation time t_0 for DP460 with creep stress σ_0

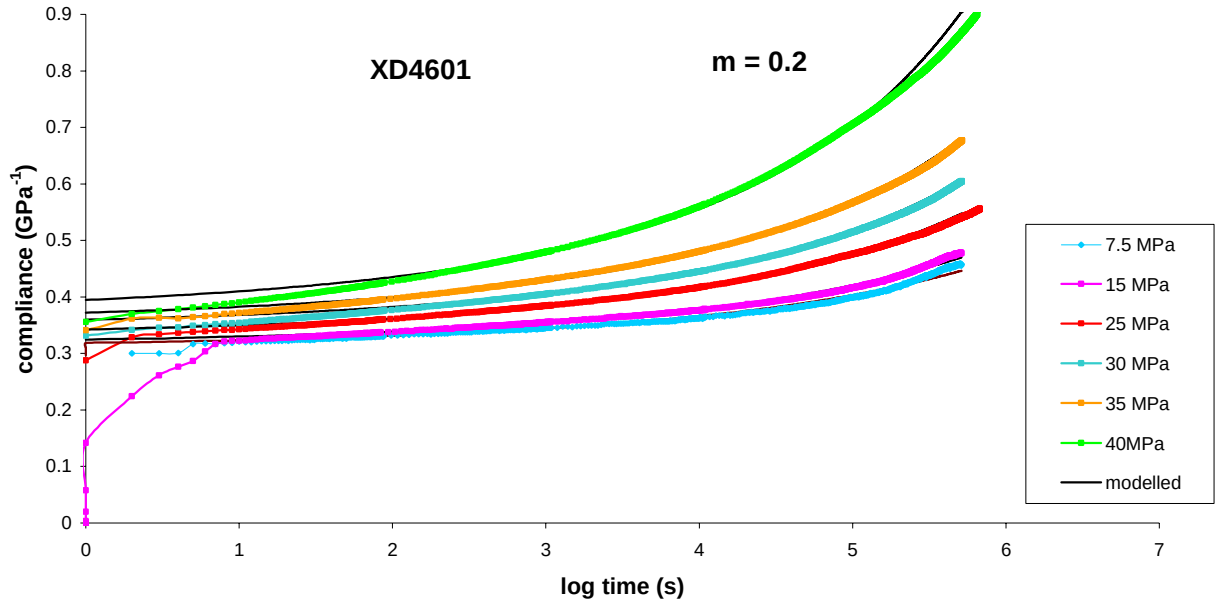


Figure 6 Creep compliance curves for XD4601 at different levels of stress

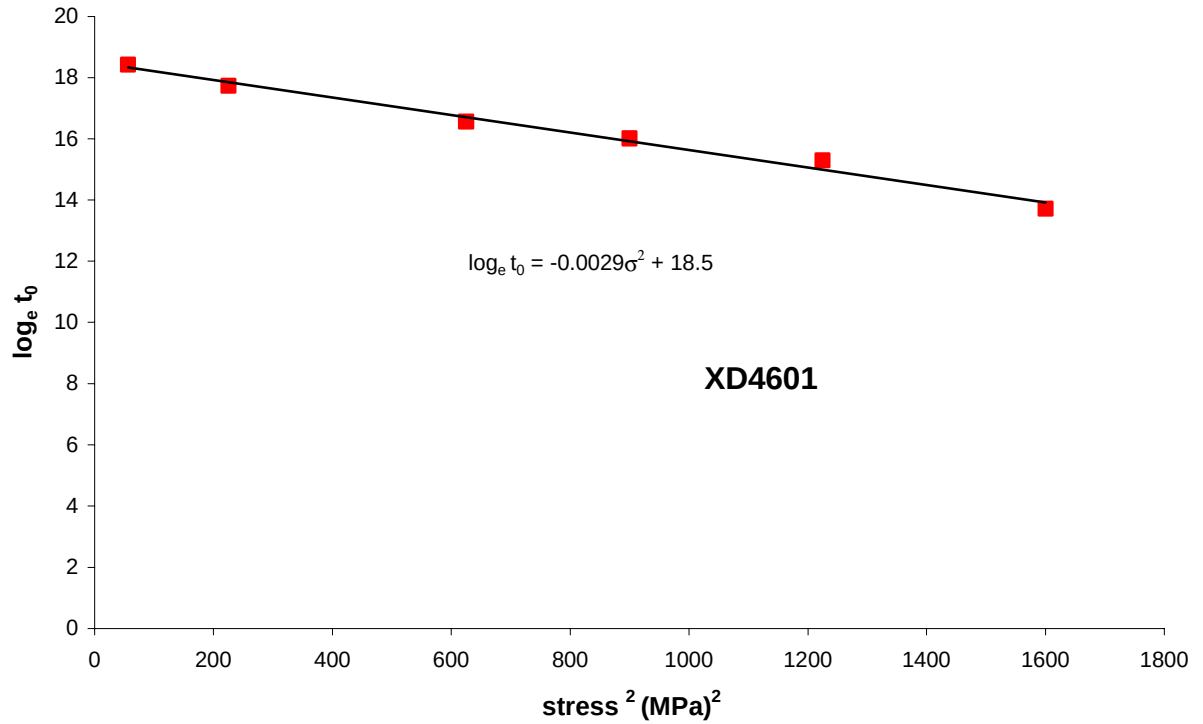


Figure 7 Variation of the mean relaxation time t_0 for XD4601 with creep stress σ_0

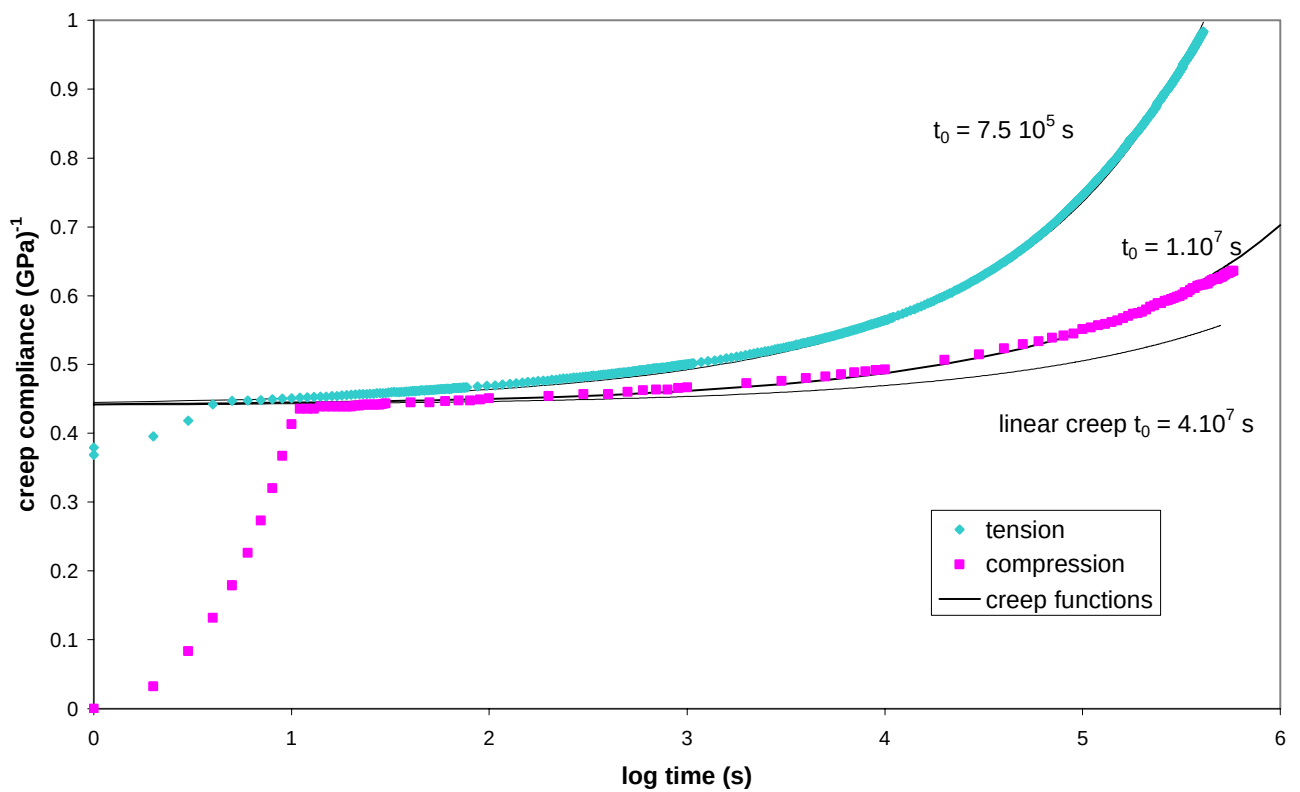


Figure 8 Experimental creep data under tension and compression obtained at a stress of 25 MPa. These have been modelled using different relaxation times t_0 . Also shown for comparison is the predicted curve for a low stress where tensile and compressive behaviour is expected to be the same.

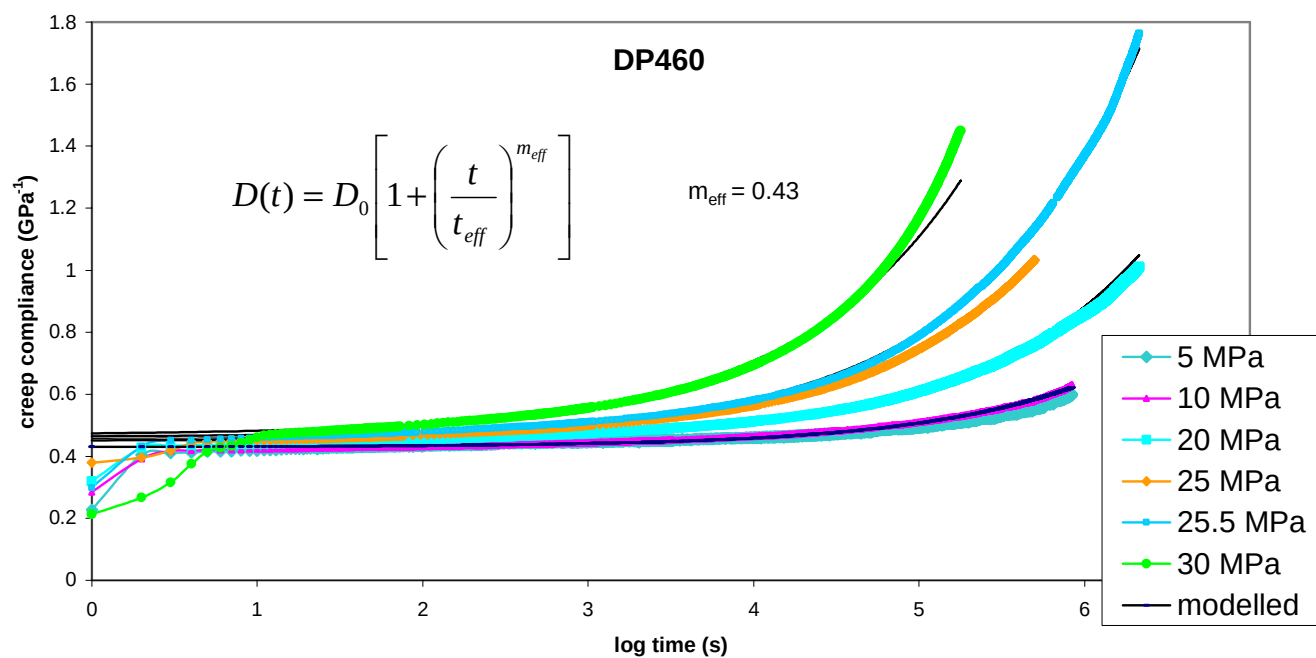


Figure 9 Creep compliance data for DP460 modelled using a power law function in time

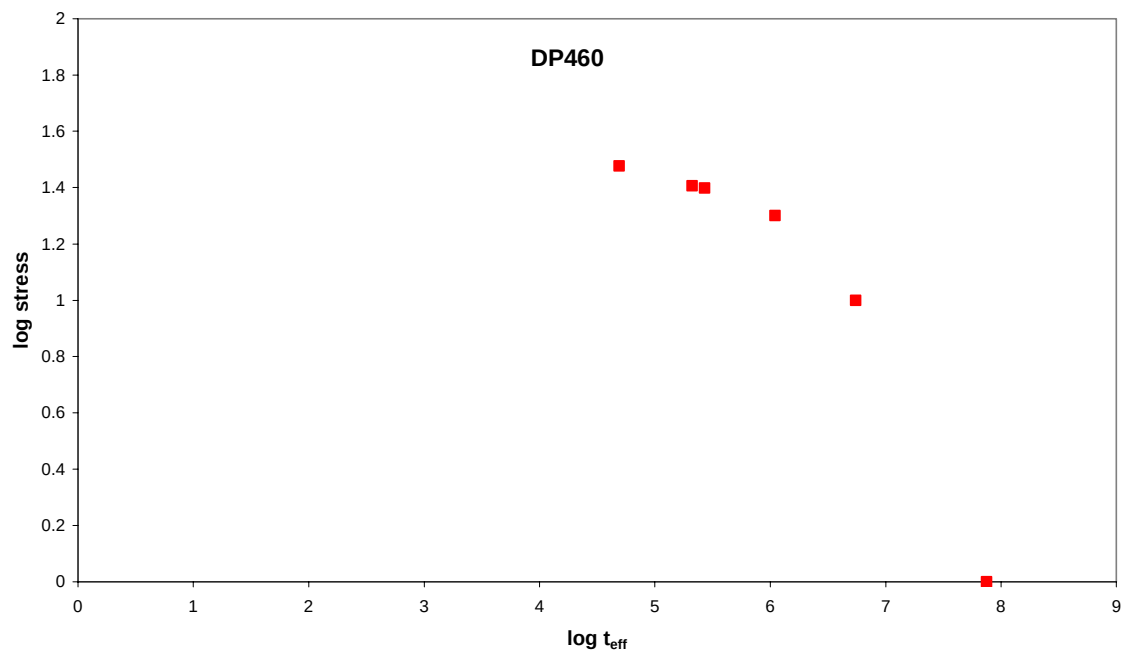


Figure 10 Plot of log stress against log relaxation time for a power law model demonstrating the unsuitability of equation (13) for modelling creep in adhesives

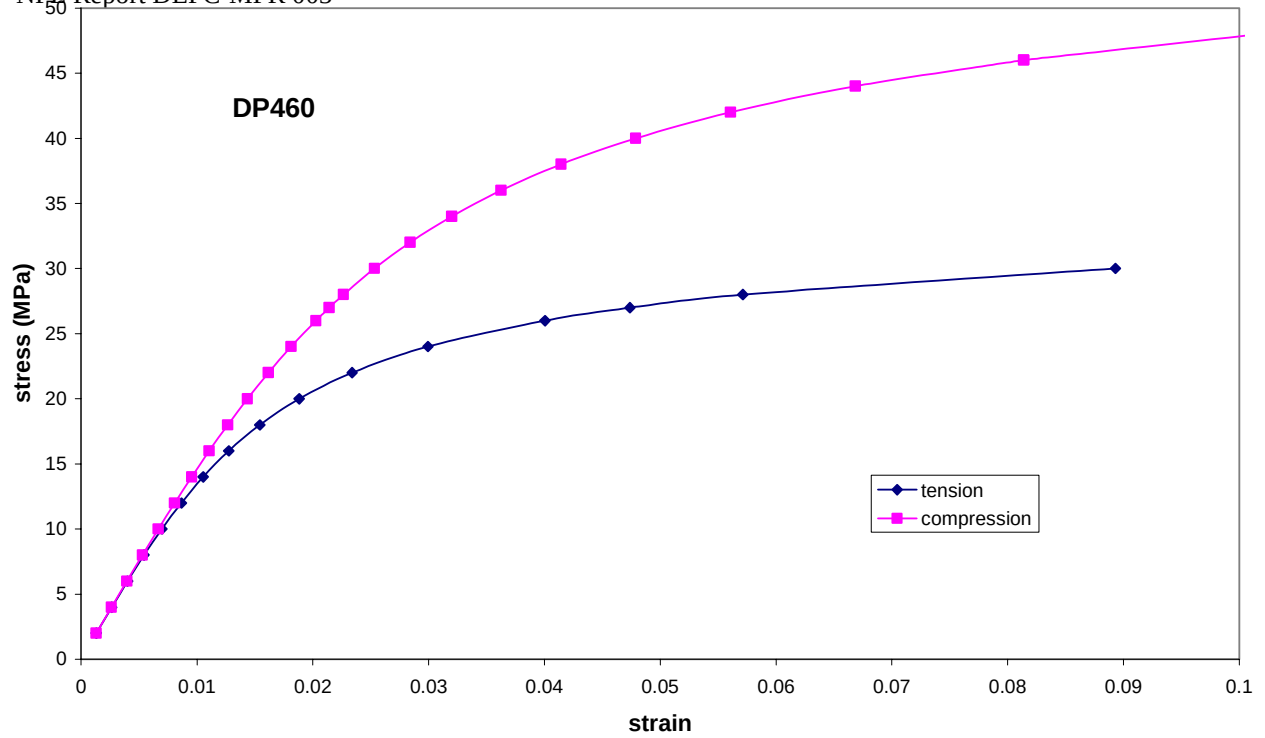


Figure 11 Isochronous stress/strain curves for DP460 derived using the creep model at a constant time under load of 10^6 seconds

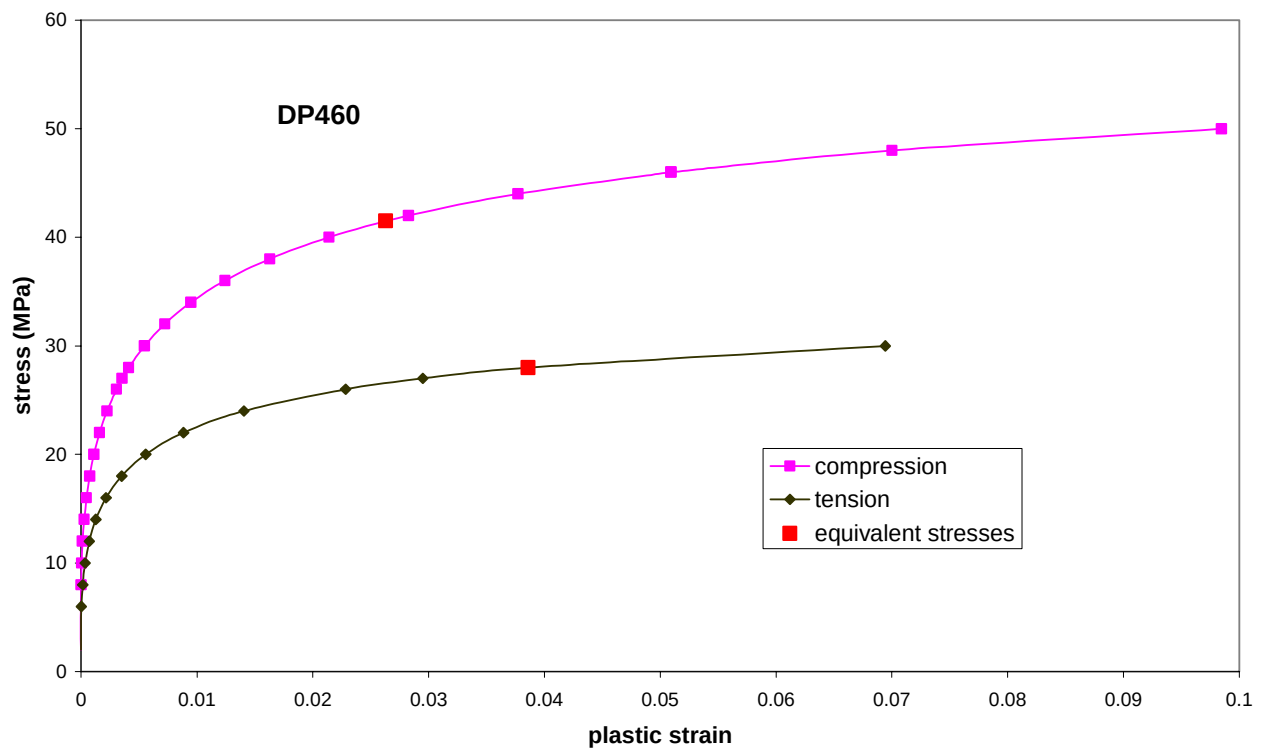


Figure 12 Stress/plastic strain curves derived from the data in figure 11.

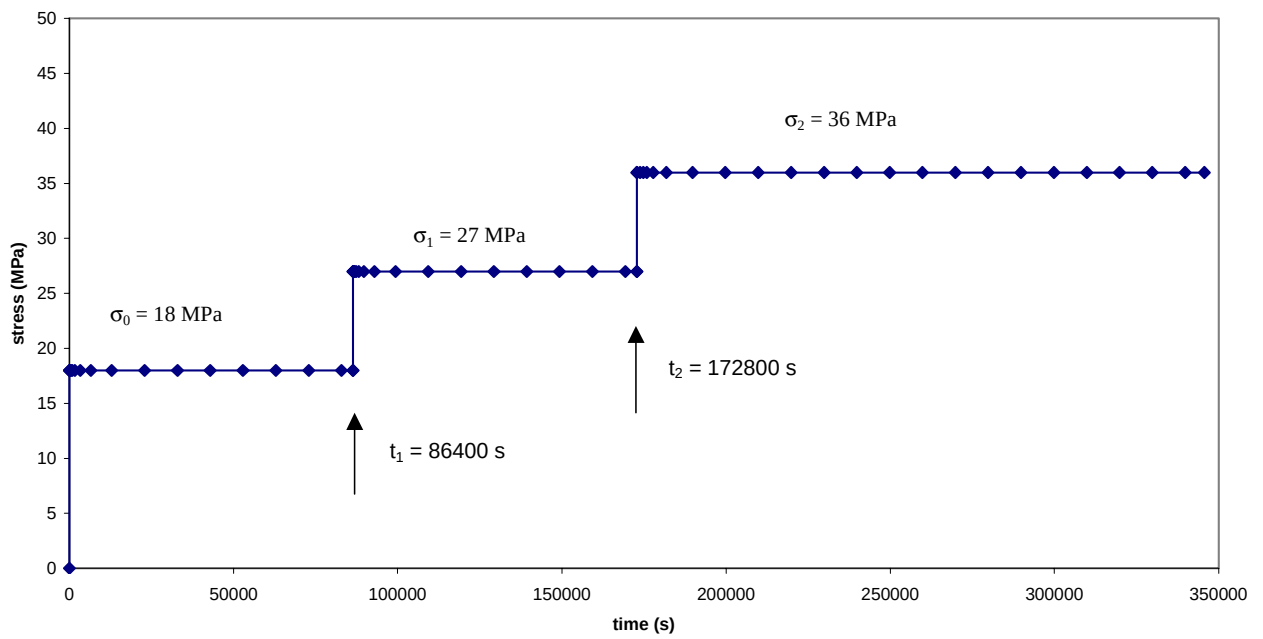


Figure 13 Stress history for a creep test and a finite element analysis on adhesive XD4601 under step increases in stress. See figure 14 for results.

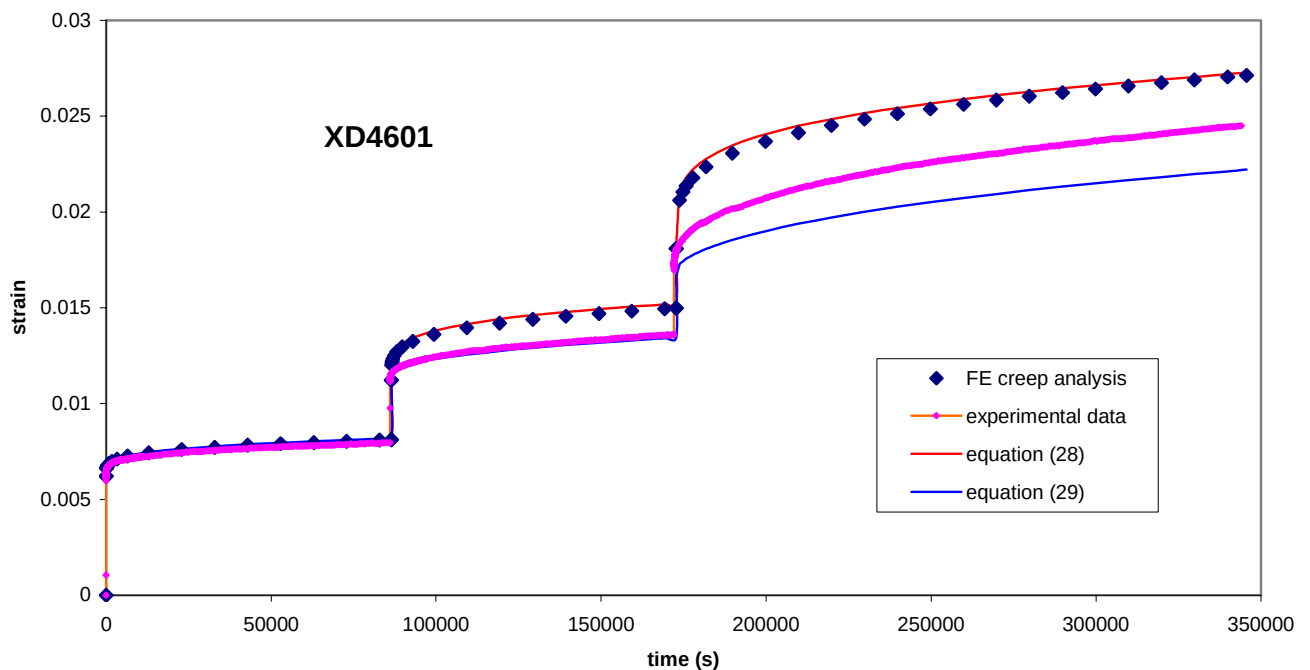


Figure 14 Comparison of creep strains in XD4601 measured under the stress history shown in figure 13 with strains calculated using the creep model expressed as time-dependent plasticity in Abaqus.

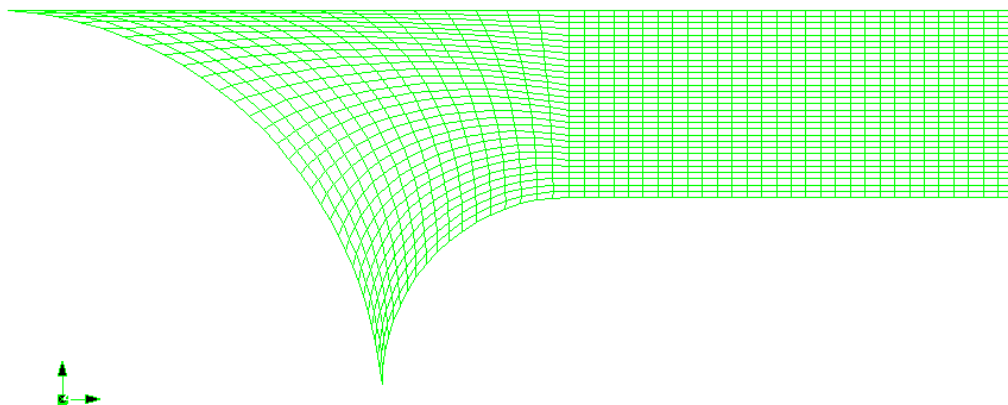
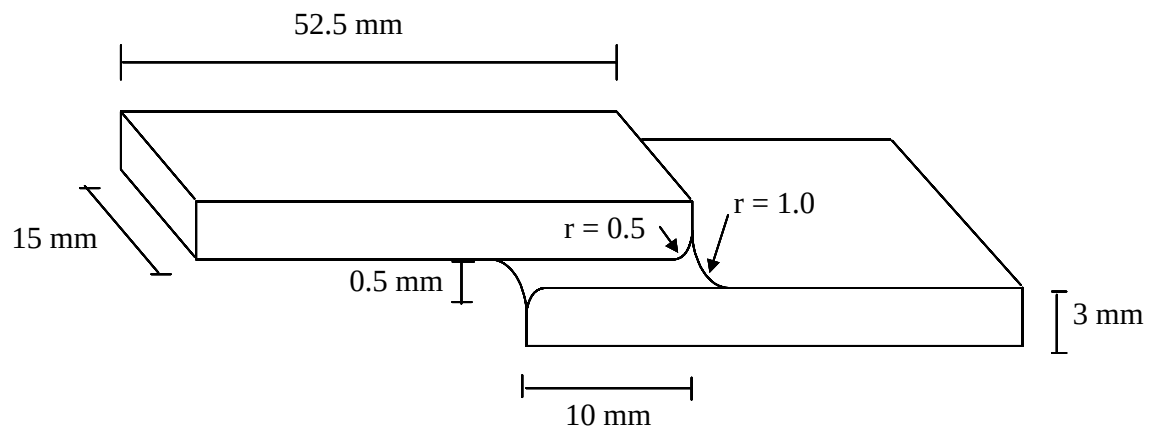


Figure 15 Diagram of the lap joint specimen and the mesh used for the fillet region of the adhesive

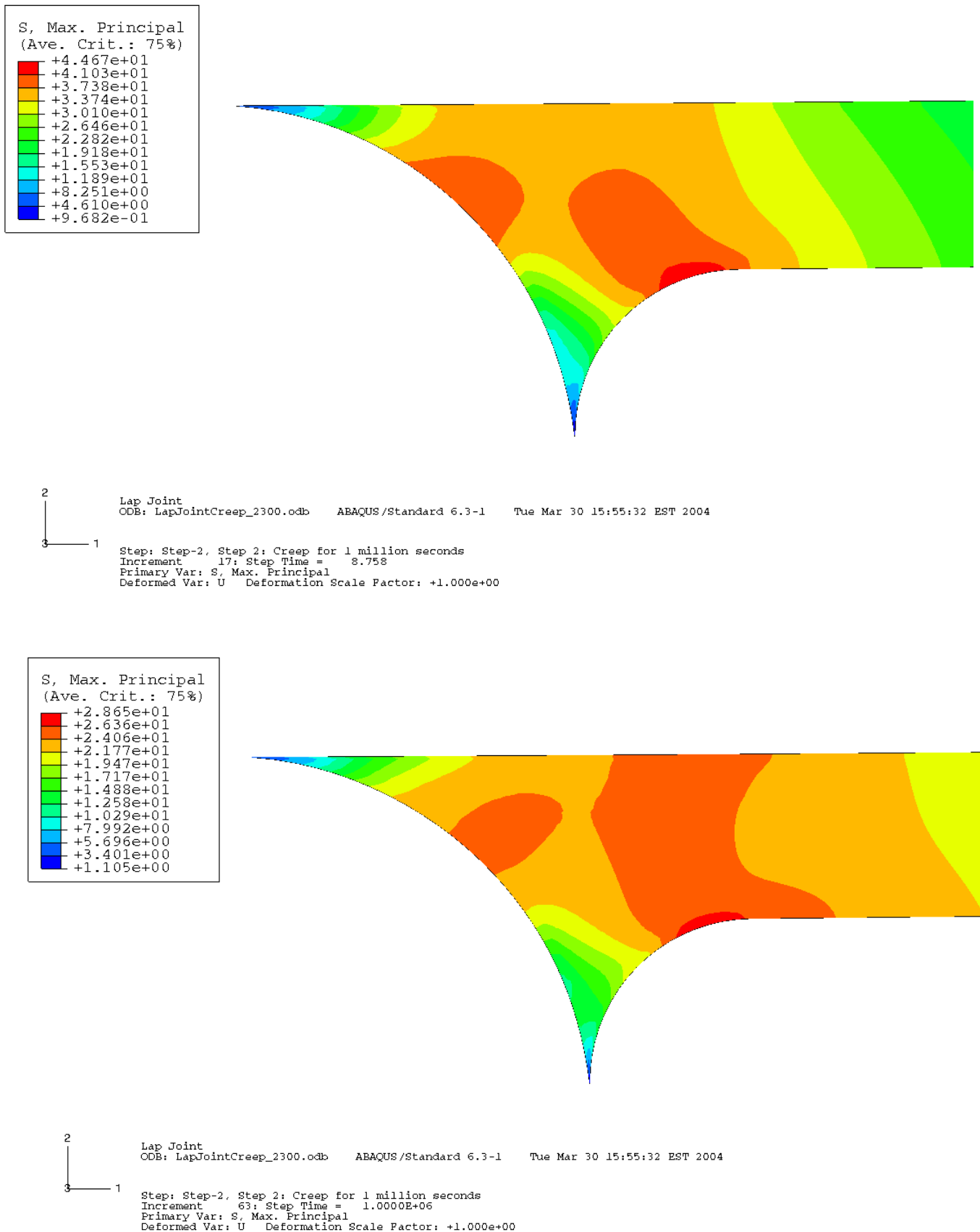


Figure16 Contours of maximum principal stress in the lap joint after 10 s and 10⁶ s under a load of 2300 N predicted using the implementation of the creep model in ABAQUS.

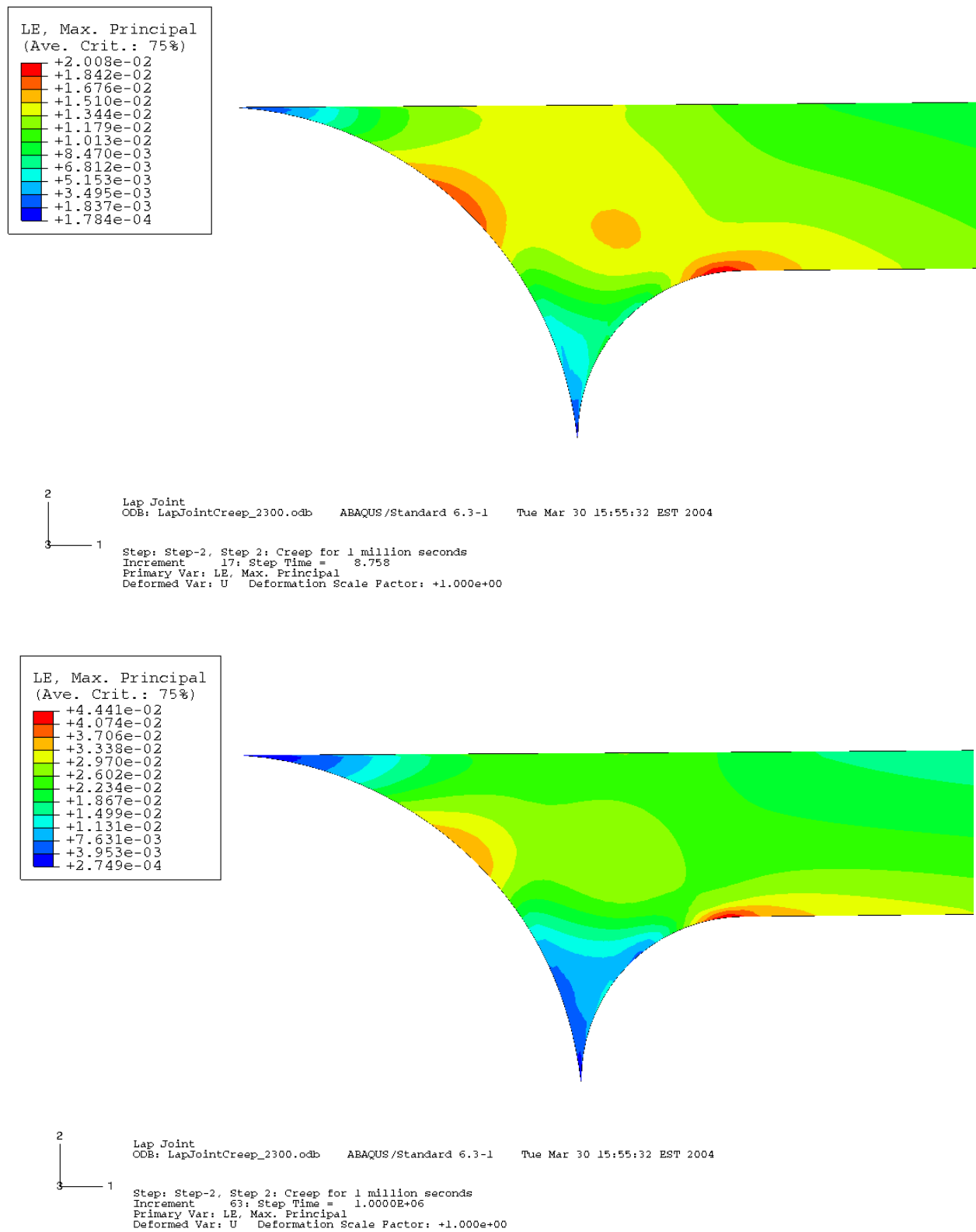


Figure 17 Contours of maximum principal strain in the lap joint after 10 s and 10⁶ s under a load of 2300 N predicted using the implementation of the creep model in ABAQUS.

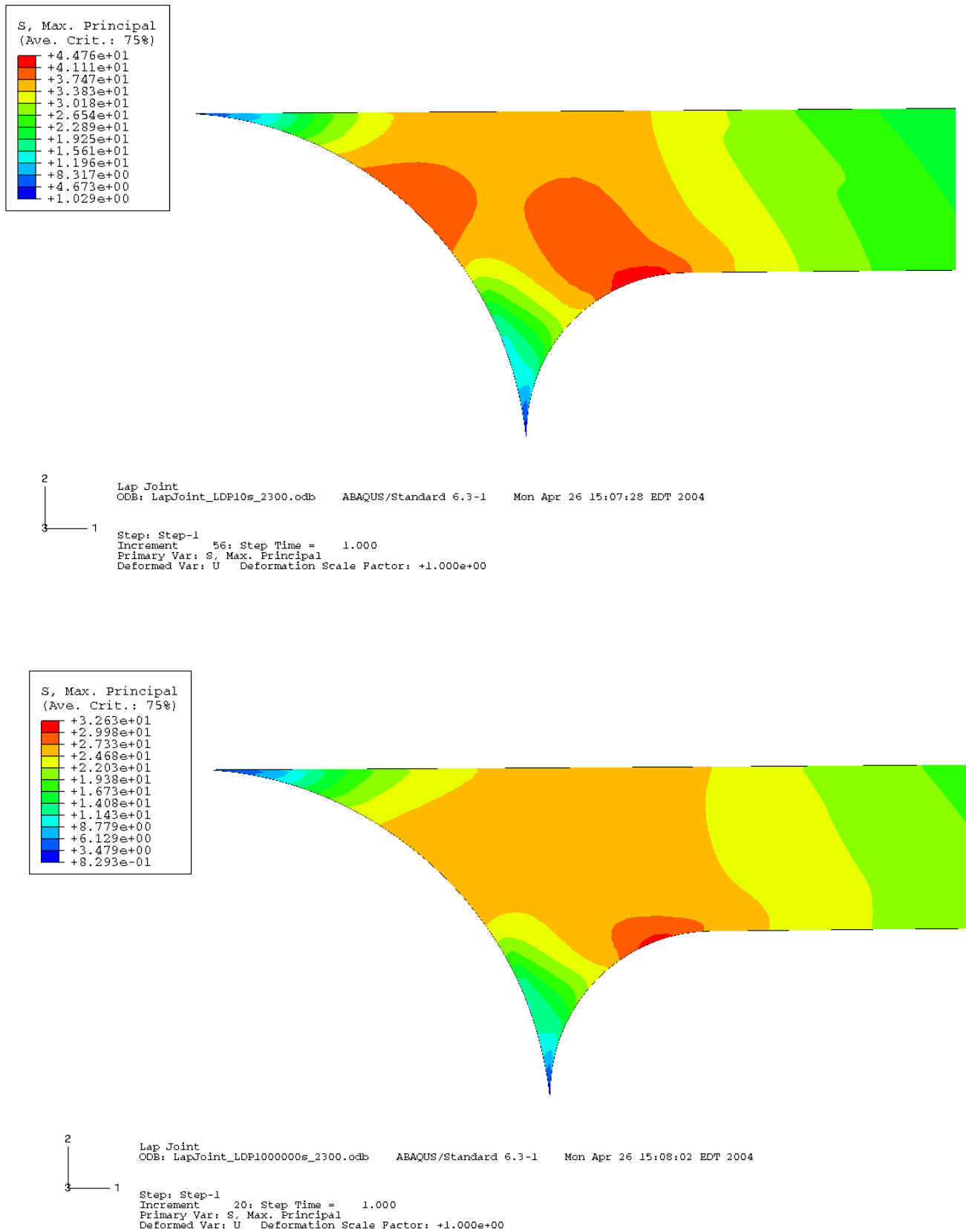


Figure 18 Contours of maximum principal stress in the lap joint after 10 s and 10^6 s under a load of 2300 N predicted using an elastic-plastic model with isochronous data.

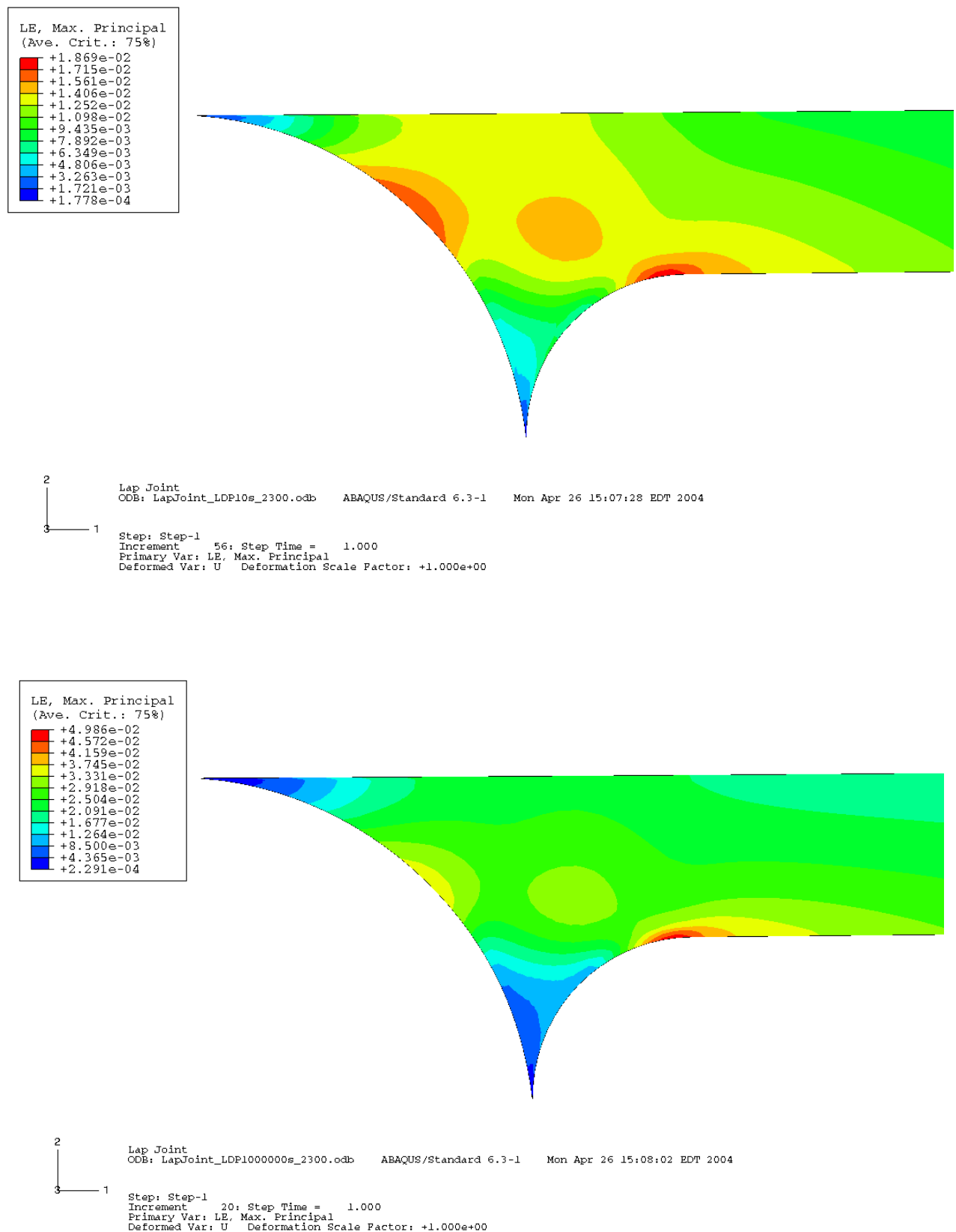


Figure 19 Contours of maximum principal strain in the lap joint after 10 s and 10^6 s under a load of 2300 N predicted using an elastic-plastic model with isochronous data.

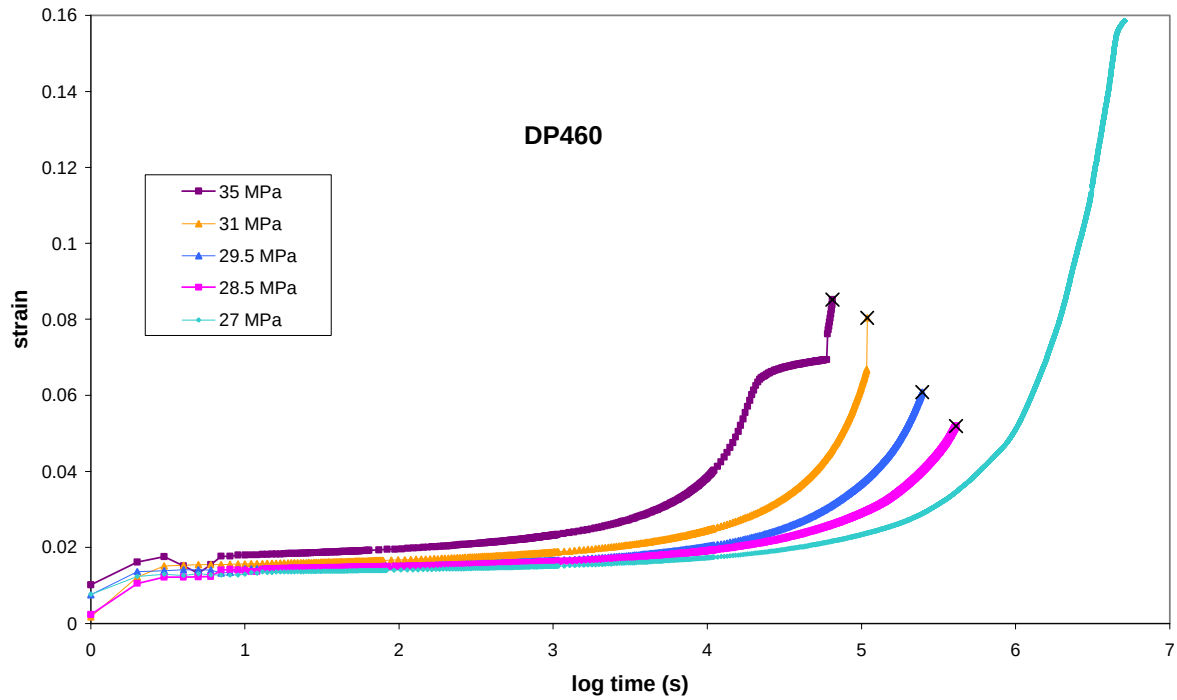


Figure 20 Strain against log time under load for the adhesive DP460 at different stress levels. The symbol x marks the instant of failure in those tests where failure was observed

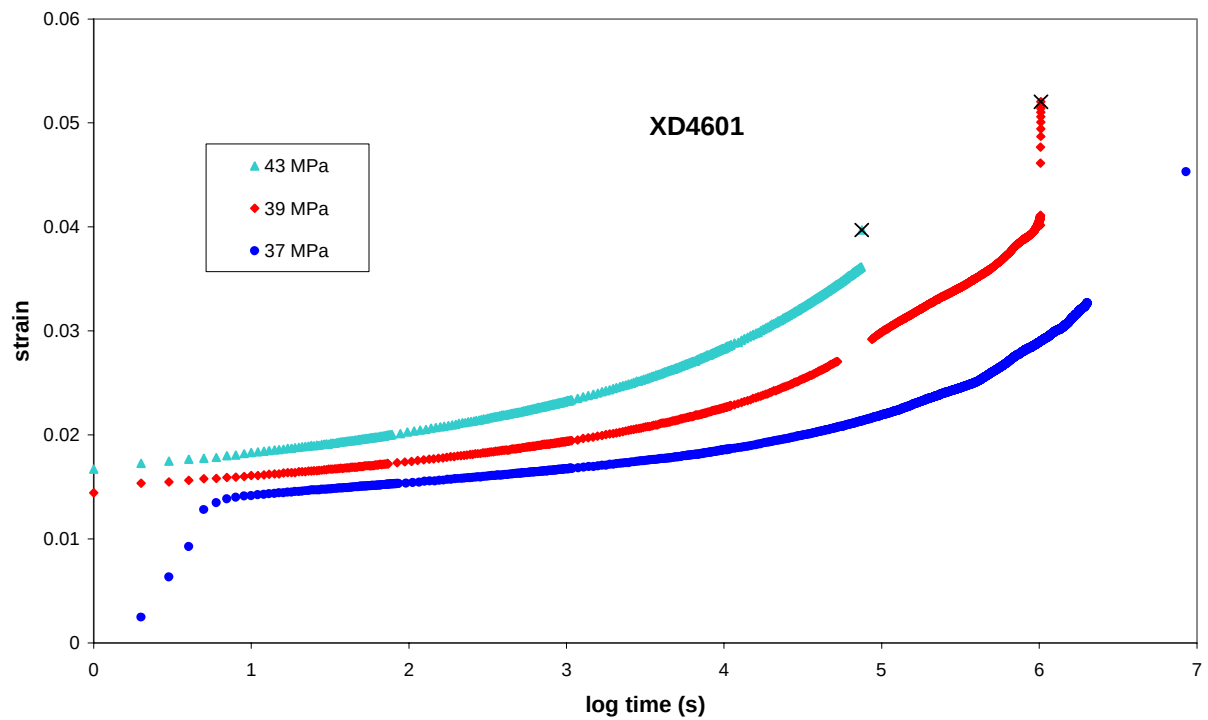


Figure 21 Strain against log time under load for the adhesive XD4601 at different stress levels. The symbol x marks the instant of failure in those tests where failure was observed