# Viscoelastic Characterization of a Glass - epoxy Composite

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*The aim of the present work is to investigate the effect of different stress levels on the creep and recovery behaviour of polymer-matrix composites. Four hours creep followed by two hours recovery tests were performed to woven GFRP specimens at five different stress levels corresponding to five different percentages of ultimate static strength. Since the behaviour was linear viscoelastic, a four element viscoelastic model was applied. In addition, the viscoelastic behaviour observed was correlated with the degree of damage generated due to creep and its variation with the applied stress levels.*

*Keywords: creep, recovery, linear viscoelastic behaviour, four parameter model.*

Glass fibers reinforced polymer composites [1, 2] are widely used in aeronautical applications and due to the polymer matrix, they exhibit significant viscoelastic behaviour. As a result, it is advisable to consider time dependent degradation of composite materials when dealing with engineering designs.

The time-dependent change in the dimensions of a polymer when subjected to a constant stress is called creep. As a result of this phenomenon the modulus of a polymer is not a constant, but provided its variation is known then the creep behaviour of polymers can be allowed for using accurate and well established design procedures.

For most traditional materials, the objective of the design method is to determine stress values which will not cause fracture. However, for polymers it is more likely that excessive deformation will be the limiting factor in the selection of working stresses, [3-12].

The mechanism of creep is not completely understood but some aspects can be explained based on structural considerations. For example, in a glassy polymer a particular polymer chain is restricted from changing its position as a result of attractions and repulsions of adjacent chains. It is generally considered that for a chain to change its position it must overcome an energy barrier and the probability of it achieving the necessary energy is improved when a stress is applied.

If the stress is removed, either partially or entirely, the strain decreases or "recovers" as a function of time, in other words, there is a delayed recovery. Polymers also have the ability to recover when the applied stress is removed and to a first approximation this can often be considered as a reversal of creep. The amount of the time-dependent recoverable strain during recovery is generally a very small part of the time-dependent creep strain for metals, whereas for polymers it may be a large portion of the time-dependent creep strain which occurred. Some polymers may exhibit full recovery if sufficient time is allowed for recovery. The strain recovery is also called delayed elasticity.

Stresses can be applied in both the linear and nonlinear viscoelastic regions. Data obtained in the linear viscoelastic region can be treated by the Boltzmann Superposition Principle; one of the simplest and most powerful principles of polymer physics. Creep recovery tests, which directly follow the creep test, also give insight into the processing behaviour of polymer melts. Recovery tests following simple-extension creep tests were used in the past in many cases for the study of validity of the superposition principle.

Creep recovery tests have been underutilized by the polymer processing industry. This is very unfortunate because these tests can produce material functions that are fast, accurate, precise and relevant to polymer processing. Creep stresses can be applied within the elastic limit and above the elastic limit to determine yield stress of polymer melts more accurately than any other rheological test.

If a cross-linked polymer is subjected to a loading cycle composed by a creep deformation up to a time $t_c$, where an equilibrium compliance is established, followed by a recovery deformation curve, follows a mirror image of the creep curve and no permanent flow phenomena appear. If the creep curve did not attain a steady state, the recovery curve will be derived from the subtraction of the corresponding values taken from the extended creep curve at times $t_r$ and $t_r + t_{cf}$ where $t_r$ is the recovery time and $t_{cf}$ the final time of creep loading.

When the stress is removed a continuously decreasing strain follows an initial elastic recovery. If the stress is too high, then the strain that does not disappear after removal of the stress is called the inelastic strain or plastic strain. Plastic strain is defined as time independent although some time dependent strain is often observed to accompany plastic strain.

In the present study, creep and recovery behaviour of glass-epoxy composite reinforced with glass fibers are investigated, (4 h creep followed by 2 hours recovery tests were performed to specimens at five different stress levels corresponding to five different percentages of ultimate strength). It was found that the variation of the reduced creep modulus of the materials considered with applied load follows an exponential decay law.

Since the observed viscoelastic behaviour was linear, a four-element viscoelastic model was used for the description of the viscoelastic behaviour and the variation of all the four elements with applied load gave a better insight into the observed behaviour.

A polymeric matrix composite can develop time - dependent damage such as debonding at the fiber-matrix interface or permanent set, which can reflect on the intrinsic creep resistance of the composite.

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Table 1
MECHANICAL PROPERTIES OF GRP-SHEETS

| Flexural strength | 340 MPa |
|---|---|
| Flexural Modulus | 22 GPa |
| Tensile strength | 240 MPa |
| Density | 2.0 g/cm³ |

In the present investigation, the observed viscoelastic behaviour was correlated with the degree of damage and its variation with applied load. It was found that damage rapidly increases with the increase of the applied load reaching a plateau where saturation of micro-damage is attained.

## Experimental part
*Materials*
The material tested was a glass-fiber polymer matrix composite, supplied in the form plates (fig. 1). The GRP-plate was 2 mm in thickness having 40 layers of glass fabric 80gsm with a layer thickness of 0.05 mm in a 0°/90° fiber orientation. Fiber volume fraction was 60%. The main mechanical properties of GRP-sheets are presented in table 1.
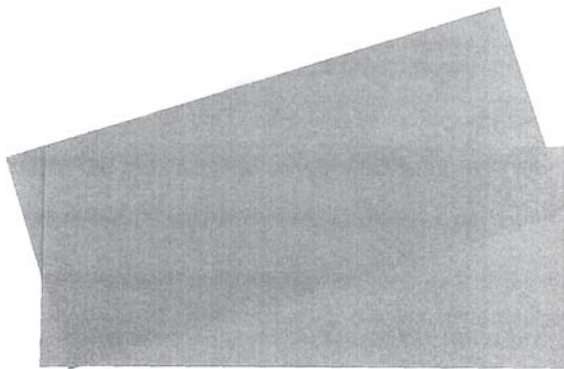


Fig. 1. Glass-fiber plate

*Creep tests*
Creep tests were performed in an arm lever creep test machine that has a ratio of load capacity ranging from 1:1 up to 5:1. Strain measurements were obtained using an LVDT displacement transducer of RDP Electronic Ltd. An A/D converter of National instruments Co was used to acquire the measurements while the treatment of signals was performed with LabView v.6.0 commercial software package.
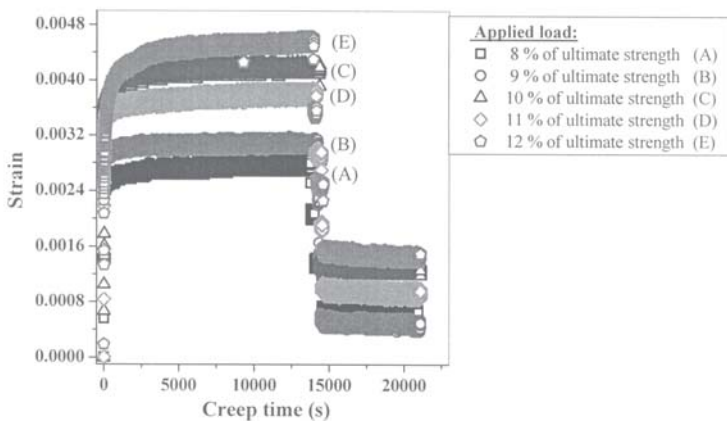
Creep tests (4 h creep followed by a 2 h recovery time) were performed on specimen's with dimensions 130mm . 10mm . 2mm, at five different stress levels; namely 27.2, 30.6, 34, 37.4, and 40.8 MPa, corresponding to 8, 9, 10, 11 and 12 % of the static strength, respectively.

## Results and discussions
The results presented herein represent average values from at least three specimens. Figure 2, shows a family of creep-recovery curves at five different stress levels. It can be seen that strains are increasing with the increase of applied load, as expected.

Next, based on the creep curves shown in figure 2, five different isochronous curves, corresponding to five different creep times are plotted in figure 3.
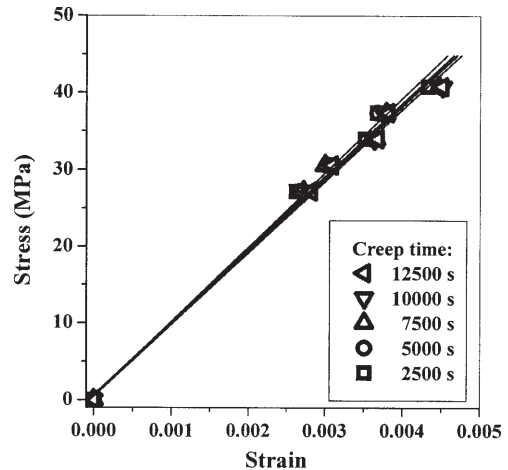


Fig. 3. Construction of isochronous curves from creep curves

From this figure, it is observed, that all isochronous curves are linear, meaning that the material behaves in a linear viscoelastic manner. Creep modulus is calculated from the inclination of the isochronous curves, and its variation with creep time is shown in figure 4. As expected, a general decrease of the modulus with time was found.
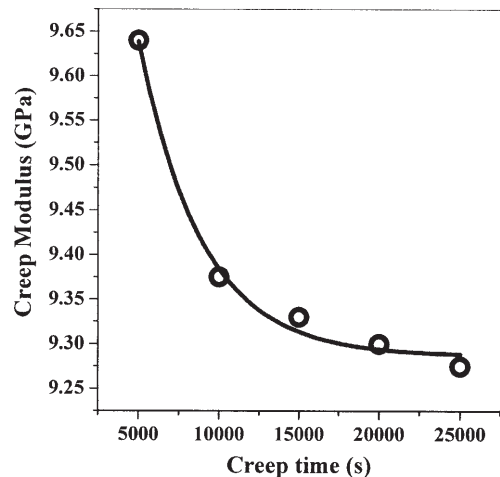


Fig. 2. Creep and recovery curves at four different stress levels



Fig. 4. Creep Modulus variation with time

*Creep Damage:*

The observed modulus decrease implies the gradual development of structural damage in the material and a measure of this damage is the damage factor, D(t), defined as:

$$D(t) = 1 - \frac{E(t)}{E_0}$$

where E(t) is the creep modulus at time t and $E_0$ is the creep modulus at t = 0.
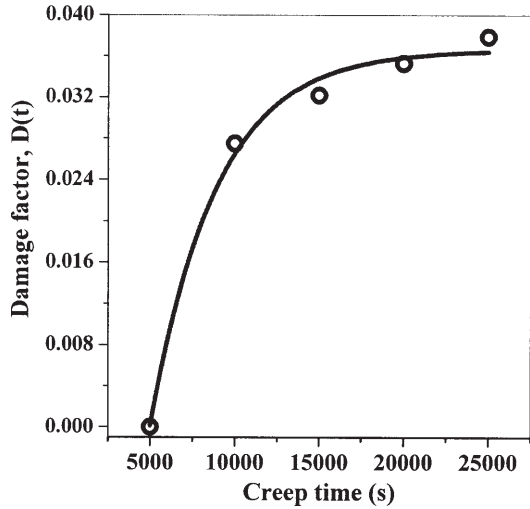


Fig. 5. Variation of the damage factor with creep time

The compound of the intrinsic damage is the influence of extrinsic damage, which refers to the matrix cracks that develop in the weaker laminae in a multidirectional laminate. A weaker lamina in a multidirectional laminate would fail when its load carrying limit is exceeded, but the failure would be constrained to this weaker lamina by the outlying stronger laminae, thus preventing the crack from spanning the entire laminate. These constrained cracks in the weaker laminae in a given lay-up are denoted as matrix cracks, since they are observed in the matrix running parallel to the reinforcing fibers. This damage can develop with time, since the weak laminae continue to share the applied stress even after fracturing due to the constraint of the strong laminae preventing the laminate from the total separation [11].

Figure 5 shows the variation of the damage factor with time. From this figure it is clear that the damage factor increases with time at a decreasing rate. More precisely, at the initial stages of application of the constant load the highest amount of intrinsic damage develops within the material and this is reflected through an almost abrupt increase in the damage factor. In the sequence, damage factor increases with time at a decreasing rate up to a time where damage saturation is attained and no further increase in the damage factor is observed.

*The Equilibrium Recoverable Compliance (ERC)*

On removing the load at the end of a test, the polymeric material can recover most if not all the strain, provided it has not yielded, but the recovery process takes time. Recovery can be rapid and complete under short-term creep, but is over delayed and may not realize completion under long-term creep or high-stress creep (or both).

Experimental recovery data can be expressed using two concepts; fractional recovery strain, FRS = strain recovered / maximum preceding creep strain, i.e. unity represents complete recovery; and reduced time, $t_R$ = recovery time / preceding creep time, i.e. unity means equal creep and recovery times.

A measure of elasticity that can be obtained from a creep-recovery experiment is the Equilibrium Recoverable Compliance (ERC), Jer. When all reversible deformation is recovered, the ERC can be calculated as:

$$J_{er} = \frac{(\varepsilon_c - \varepsilon_r(t))}{\sigma}$$

where $\varepsilon_c$ represents the strain at final time of creep loading and $\varepsilon_r$ represents the strain at the recovery time. The lower is the ERC, the higher will be the elasticity.
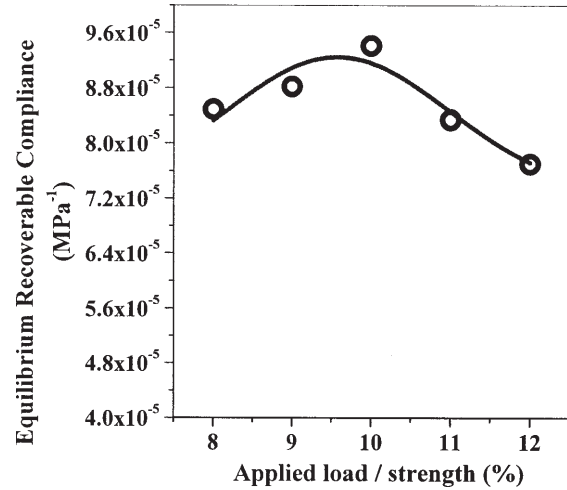


Fig. 6. Variation of the Equilibrium Recoverable Compliance (ERC) at the time of unloading, $t = t_r$, with applied load

The variation of (ERC) as measured at the time of unloading $t_r$ with applied load is shown in figure 6. From this figure a general decrease of ERC with the applied load is observed. This is due to the fact that as the applied load is increased, the abrupt jump of the strain exhibited upon unloading, which represents the elastic part of the viscoelastic behaviour of the material, is increased.

*Modelling*

To describe the viscoelastic response of a material, a constitutive model is required [13, 14]. There is a variety of equations that have been used to model the viscoelastic behaviour of polymeric materials. A simple model, known as the Burgers or four-element model consists of a Maxwell and a Kelvin model in series, as shown in figure 7. The model can be applied under the condition of linear viscoelastic behaviour. The characteristic equations for the prediction of creep behaviour are given below.

*Burgers or Four Element model*

The Burgers model is shown in figure 7a where a Maxwell and a Kelvin model are connected in series. The constitutive equation for a Burgers model can be derived by considering the strain response under constant stress of each of the elements coupled in series as shown in figure 7a.
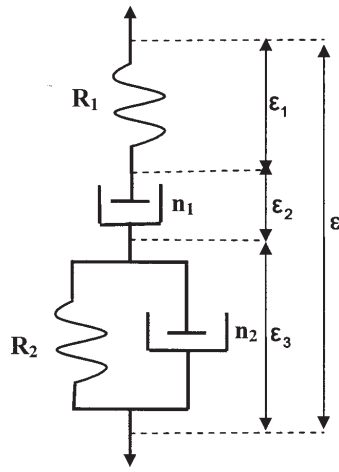
The total strain at time t will be the sum of the strain in the three elements, where the spring and dashpot in the Maxwell model are considered as two elements:

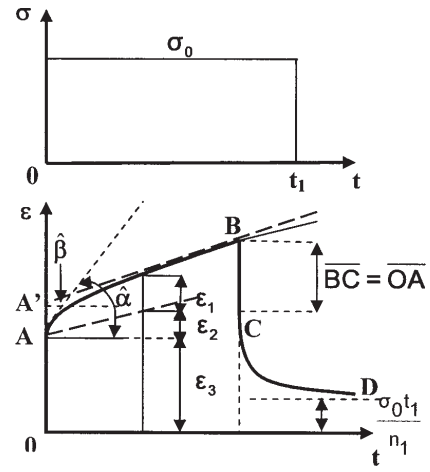$$\varepsilon = \varepsilon_1 + \varepsilon_2 + \varepsilon_3, \qquad (1)$$

Where $\varepsilon_1$, is the strain of the spring:

$$\varepsilon_1 = \frac{\sigma}{R_1}, \qquad (2)$$

$\varepsilon_2$ is the strain in the dashpot:

a - Burgers Model



b - Creep and Recovery

Fig. 7. Behaviour of a Burgers Model

$$\dot{\varepsilon}_2 = \frac{\sigma}{n_1}, \tag{3}$$

and $\varepsilon_3$ is the strain in the Kelvin unit:

$$\dot{\varepsilon}_3 + \frac{R_2}{n_2}\varepsilon_3 = \frac{\sigma}{n_2} \tag{4}$$

Equations 1-4 contain five unknowns $\varepsilon$, $\sigma$, $\varepsilon_1$, $\varepsilon_2$, $\varepsilon_3$ where $\varepsilon$ and $\sigma$ are external variables and $\varepsilon_1$, $\varepsilon_2$, $\varepsilon_3$ are internal variables. In principle, $\varepsilon_1$, $\varepsilon_2$, $\varepsilon_3$ can be eliminated from these four equations to yield a constitutive equation between $\sigma$ and $\varepsilon$ for the Burgers model with the following result:

$$\sigma + \left(\frac{n_1}{R_1} + \frac{n_2}{R_2} + \frac{n_3}{R_2}\right)\dot{\sigma} + \frac{n_1 n_2}{R_1 R_2}\ddot{\sigma} = n_1\dot{\varepsilon} + \frac{n_1 n_2}{R_2}\ddot{\varepsilon} \tag{5}$$

The creep behaviour of the Burgers model under constant stress $\sigma_o$ can be obtained from (5) by solving this second order differential equation with two initial conditions:

$$\varepsilon = \varepsilon_1 = \frac{\sigma_0}{R_1}, \varepsilon_2 = \varepsilon_3 = 0, t = 0 \tag{6}$$

$$\dot{\varepsilon} = \frac{\sigma_0}{n_1} + \frac{\sigma_0}{n_2}, t = 0 \tag{7}$$

The creep behaviour may be found to be as follows and as illustrated in figure 7b:

$$\varepsilon(t) = \frac{\sigma_0}{R_1} + \frac{\sigma_0}{n_1}t + \frac{\sigma_0}{R_2}\left(1 - e^{-\frac{R_2 t}{n_2}}\right) = \frac{\sigma_0}{R_1} + \frac{\sigma_0}{R_2}\left(1 - e^{-\frac{t}{\tau}}\right) + \frac{\sigma_0}{n_1}t \tag{8}$$

where $\sigma_o$ is the initially applied stress and $\tau = n_2/R_2$ is the retardation time taken to produce 63.2% or $(1 - e^{-1})$ of the total deformation in the Kelvin unit.

The creep characteristics of Burgers model as described by relation (8) can be depicted as follows. The first term is constant and describes the instantaneous elastic deformation; the second one is delayed elasticity of the Kelvin unit and dominant in the earliest stage of creep, but soon goes to a saturation value close to $t_o/R_2$; the viscous flow then increases nearly linearly in the third term after a long enough period of loading.

Differentiating (8) yields the creep rate $\dot{\varepsilon}$ of the Burgers model:

$$\dot{\varepsilon} = \frac{\sigma_0}{n_1} + \frac{\sigma_0}{n_2}e^{-\frac{R_2 t}{n_2}} = \frac{\sigma_0}{n_1} + \frac{\sigma_0}{n_2}e^{-\frac{t}{\tau}} \tag{9}$$

Thus the creep rate starts at $t = 0^+$ with a finite value:

$$\dot{\varepsilon}(0^+) = \left(\frac{1}{n_1} + \frac{1}{n_2}\right)\sigma_0 = \tan\alpha, \tag{10}$$

(fig. 7b) and approaches asymptotically to the value:

$$\dot{\varepsilon}(\infty) = \frac{\sigma_0}{n_1} = \tan\beta, \tag{11}$$

It may also be observed from figure 7b, that $\overline{OA} = \frac{\sigma_0}{R_1}$ and $\overline{AA'} = \frac{\sigma_0}{R_2}$. Thus in theory the material constants $R_1$, $R_2$, $n_1$ and $n_2$ may be determined from a creep experiment by measuring $\alpha$, $\beta$, OA and AA' as in figure 7b.

If the stress $\sigma_o$ is removed at time $t_1$, the recovery behaviour of the Burgers model can be obtained from (8) and the superposition principle by considering that at $t = t_1$ a constant stress $\sigma = -\sigma_o$ is added. According to the superposition principle the recovery strain $\varepsilon(t), t \succ t_1$ is the sum of these two independent actions:

$$\varepsilon(t) = \frac{\sigma_0}{R_1} + \frac{\sigma_0}{n_1}t + \frac{\sigma_0}{R_2}\left(1 - e^{-\frac{R_2 t}{n_2}}\right)$$
$$- \left[\frac{\sigma_0}{R_1} + \frac{\sigma_0}{n_1}(t - t_1) + \frac{\sigma_0}{R_2}\left(1 - e^{-\frac{R_2(t - t_1)}{n_2}}\right)\right], t \succ t_1$$

or

$$\varepsilon(t) = \frac{\sigma_0}{n_1}t_1 + \frac{\sigma_0}{R_2}(e^{\frac{R_2 t_1}{n_2}})e^{-\frac{R_2 t}{n_2}}, \tag{12}$$

Recovery is also illustrated in figure 7b. The recovery has an instantaneous elastic recovery followed by creep recovery at a decreasing rate, as shown in (12). The second

term of (12) decreases toward zero for large times, while the first term represents a permanent strain due to viscous flow of $n_1$. Thus the recovery approaches asymptotically to $\varepsilon(\infty) = \dfrac{\sigma_0}{n_1} t_1$ as approaches infinite.

*Calculation of constants*

$R_1$, $R_2$, $n_1$ και $n_2$ parameters are calculated as it is described below:

*Constant $R_1$:* The spring element constant, $R_1$, for the Maxwell model may be obtained from the instantaneous strain, $\varepsilon_1$. Thus, $R_1 = \dfrac{\sigma_0}{\varepsilon_1} = \dfrac{\sigma_0}{OA}$, where $\sigma_0$ = applied stress, and $\varepsilon_1$ = instantaneous strain, (fig. 7).

Variation of $R_1$ versus applied load is shown in figure 8. A small increase of $R_1$ with applied load is observed until it reaches a saturated level and then it remains almost constant. The observed increase reflects a small stiffness increase with applied load.
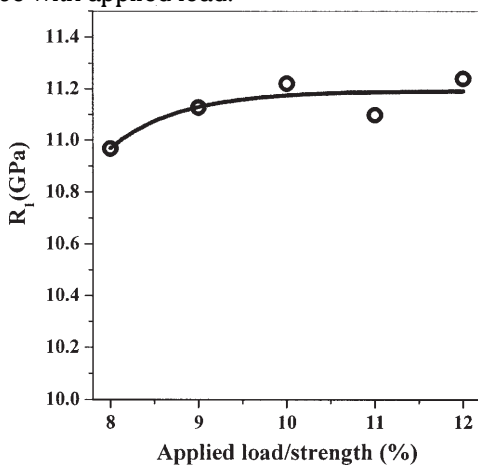


Fig. 8. Variation of $R_1$ with applied load / strength(%)

*Constant $n_1$:* The dashpot constant, $n_1$, for the Maxwell element is obtained from equation (3), as it is described in figure 7.

Variation of $n_1$ versus applied load is shown in figure 9. First, for lower values of applied load, $n_1$ increases until it reaches its maximum value and then decreases with applied load. Since $\dot\varepsilon(\infty) = \dfrac{\sigma_0}{n_1} = \tan\beta$, and a high variation of the applied load does not exists, an initial decrease in $\dot\varepsilon(\infty)$ followed by a subsequent increase will appear. This fall starts exactly at $\sigma = 10\% \ \sigma_u$ a value at which all the three parameters; namely $R_1$, $R_2$ and $n_2$ attain a constant value (figs. 8, 10 and 11).

*Constant $R_2$:* The spring constant, $R_2$, for the Kelvin – Voigt element is obtained from the equation: $R_2 = \dfrac{\sigma_0}{AA'}$.

Variation of $R_2$ versus applied load is shown in figure 10. As it is clear, $R_2$ is decreasing constantly with the applied load.

*Constant $n_2$:* The dashpot constant, $n_2$, for the Kelvin – Voigt element may be determined by selecting a time and corresponding strain from the creep curve in a region where the retarded elasticity dominates (i.e., the knee of the curve in fig. 7) and substituting into equation (9). It is calculated from equation (9) for a point in the curve.

Variation of $n_2$ versus applied load is shown in figure 11 and it is decreasing continuously with the increase of applied load.

Results showed that both $R_2$ and $n_2$ were considerably
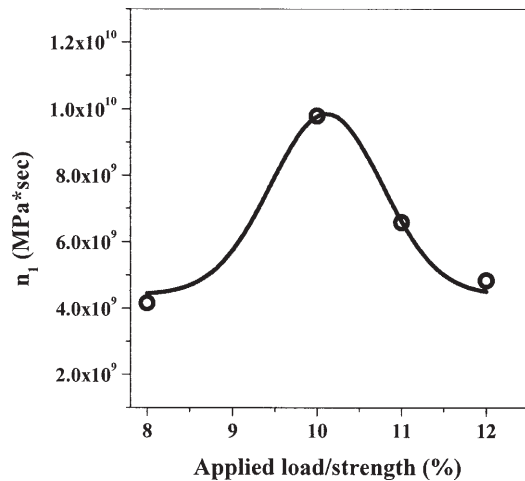


Fig. 9. Variation of $n_1$ with applied load / strength (%)
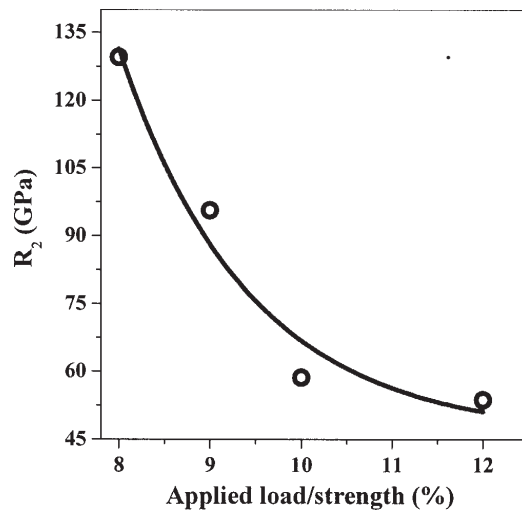


Fig. 10. Variation of $R_2$ with applied load / strength (%)

decreased with increasing load, showing high stress dependency. This indicated the fact that the materials with high bulk modulus deformed very small under low stress, which showed the Kelvin unit behaved with extremely high modulus and very difficult viscous flow. With increasing stress, the orientation movement of amorphous chains including elastic deformation and viscous flow became exaggerate, resulting in a reduced $R_2$ and $n_2$. The consistent change of $R_2$ and $n_2$ with stress led to a nearly constant retardation time, $\tau$ for each specimen ($\tau \cong 400\,s$).
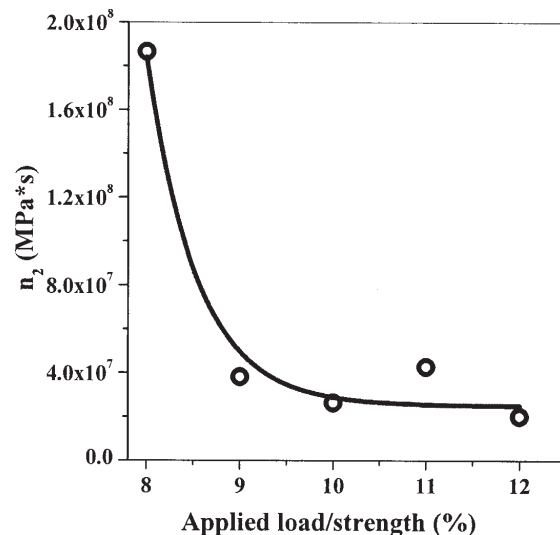


Fig. 11. Variation of $n_2$ with applied load / strength (%)

*Comparison between predicted values and experimental results*

Having thus determined the constants for the model, the strain may be predicted at any selected time or stress level assuming that these are within the linear viscoelastic region where the model is applicable.

Figures 12-16 show experimental and predicted creep curves for all the stress levels applied (8, 9, 10, 11 and 12 % of maximum strength). It is clear that there is a fair agreement between experimental and predicted values from the application of four parameter model.
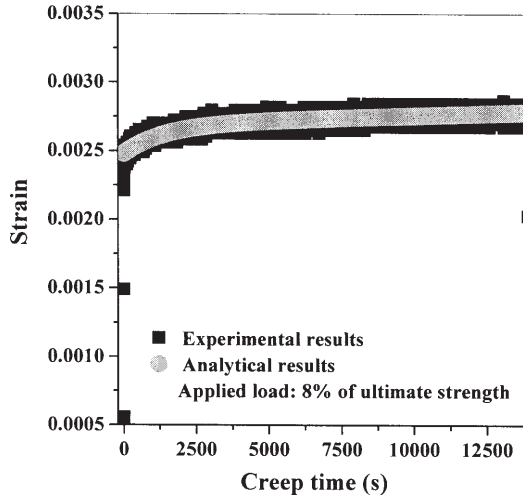


Fig. 12. Comparison between experimental and predicted values from the application of the parameter model at 8% of ultimate strength applied load
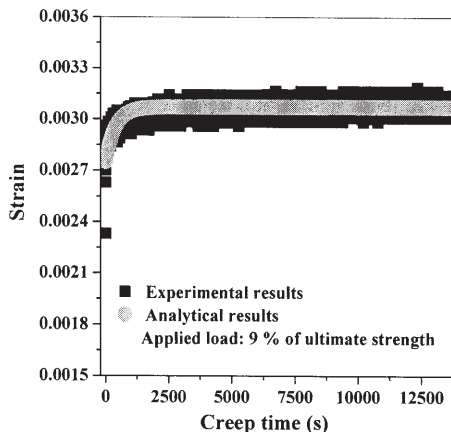


Fig. 13. Comparison between experimental and predicted values from the application of the parameter model at 9% of ultimate strength applied load
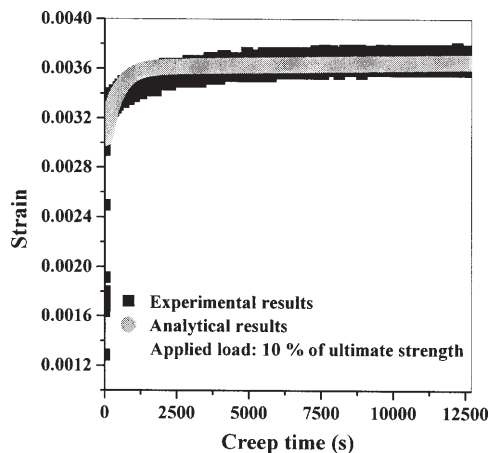


Fig. 14. Comparison between experimental and predicted values from the application of the parameter model at 10% of ultimate strength applied load
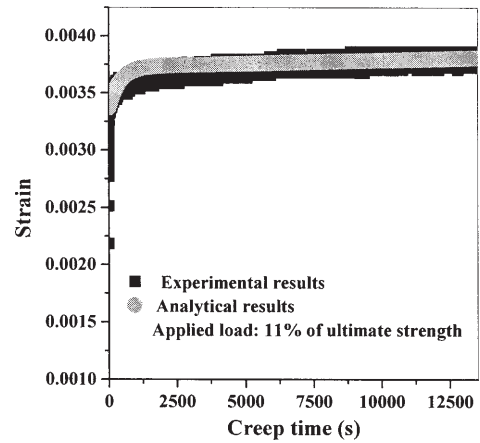


Fig. 15. Comparison between experimental and predicted values from the application of the parameter model at 11% of ultimate strength applied load
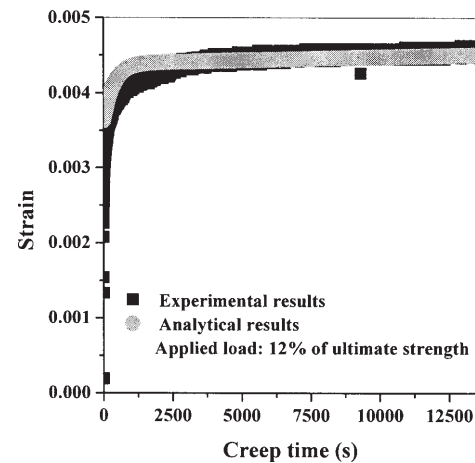


Fig. 16. Comparison between experimental and predicted values from the application of the parameter model at 12% of ultimate strength applied load

**Conclusions**

In the present work the effect of creep and recovery behaviour of a polymer-matrix composite was investigated. For this purpose, 4 h creep followed by 2 h recovery tests were performed to specimens for five different stress levels, (8, 9, 10, 11 and 12 % of the static ultimate strength).

The variation of the reduced creep modulus of the material considered with the different applied stress levels follows an exponential decay law.

In order to better understand such a behaviour, a four parameter viscoelastic model was applied and the variation of all the four parameters with the five different applied stress levels gave a better insight into the observed behavior.

A very good agreement between experimental and analytical results for all stress levels was observed, testifying that we are in the linear viscoelastic region and that the experimental procedure was almost perfect.

A general decrease of the Equilibrium Recoverable Compliance, ERC, with the applied load is observed. This is due to the fact that as the applied load is increased, the abrupt jump of the strain exhibited upon unloading, which represents the elastic part of the viscoelastic behaviour of the material, is increased.

In addition, the viscoelastic behaviour observed was correlated with the value of the degree of damage and its variation with the applied stresses.

It was found that damage increases rapidly with applied stress reaching a plateau where saturation of micro-damage is attained.

Results showed that both $R_2$ and $n_2$ were considerably decreased with increasing load, showing high stress dependency. The consistent change of $R_2$ and $n_2$ with stress led to a nearly constant retardation time, $\tau$ for each specimen ($\tau = 400s$).

## References

1.HULL, D., CLYNE, T.W., An Introduction to Composites Materials, Cambridge Solid State Science Series, Second Edition, 1996

2.HULL, DEREK, "An introduction to composite materials", Cambridge University Press, 1981

3.GUEDES, M., TAVARES, CM.L., FERREIRA, AJ.M., Experimental and theoretical study of the creep behaviour" Composites Science and Technology, **64**, 2004, p. 1251

4.RAGHAVAN, J., MESHII M., Creep of Polymer Composites, Composites Science and Technology, **57**, 1997, p. 1673

5.ZHENG, SF, WENG, GJ., European Journal of Mechanics A/Solids, , **21**, 2002, p. 411

6.PAPANICOLAOU, G.C, XEPAPADAKI, A.G, KARAGOUNAKI, K., ZAOUTSOS, S.P., Journal of Applied Polymer Science, **108**, 2008, p. 640

7.G.C. PAPANICOLAOU, S.P. ZAOUTSOS, A.H. CARDON, Further development of a data reduction method for the nonlinear viscoelastic characterization of FRPs, Composites, Part A, **30,** 1999, p. 839

8.S. P. ZAOUTSOS, G. C. PAPANICOLAOU, A. H. CARDON, On the non-linear viscoelastic behaviour of polymer-matrix composites, Composifes Science und Technology, **58**, 1998, p. 883

9.G.C. PAPANICOLAOU , S.P. ZAOUTSOS, A.H. CARDON, Prediction of the non-linear viscoelastic response of unidirectional fiber composites, Composites Science and Technology, **59**, 1999, 1311±1319

10.BONIFACE, I., OGIN, S.I., SMITH, P.A., Damage Development in Woven Glass Fibre/Epoxy Laminates Under Tensile Loading, The Institute of Materials. Proceedings of the 2nd International Conference on Deformation and Fracture of Composites, Manchester, 29-31 March, 1993, p.33-1

11.LEADERMANN, H., Elastic and Creep Properties of Filamentous Materials and Other High Polymers, Textile Foundation ,Washinghton DC, USA, 1943

12.HOUSHYAR, S., SHANKS, R.A., HODZIC, A., Tensile creep behaviour of polypropylene fibre reinforced polypropylene composites, Polymer Testing, **24**, 2005, p. 257

13.RAGHAVAN, J., MESHII, M., Creep rupture of Polymer Composites, Composites Science and Technology, **57**, 1997, 375

14.PANG, F., WANG, C.H., Activation theory for creep of woven composites, Composites: Part B: **30,** 1999, p. 613

15.W.N.FINDLEY, J.S.LAI, K.ONARAN, Creep and Relaxation of nonlinear Viscoelastic Materials With an introduction to Linear Viscoelasticity, Dover Publications, INC, New York, 1989