Viscoelastic behavior of carbon epoxy composites by creep tests

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Abstract (Times New Roman 12 pt bold, 6 pt space before and after titles)

One of the main requirements for the use of fiber-reinforced polymer matrix composites in structural applications is their evaluation during service life, which demands a study of the time dependent behavior due to viscoelastic response of the polymeric matrix. In the present study, creep and creep rupture test in constant load were performed in specimens of unidirectional carbon fiber-reinforced epoxy composites with fibers orientations of 90º, at temperatures of 25 and 70 ºC. The aim is the viscoelastic characterization of the material through the creep curves to some levels of constant load during periods of 1000 h, the attainment of the "creep rupture envelope" by the creep rupture curves and the determination of the transition from linear to non-linear behavior through isochronous curves. In addition, comparisons of creep compliance curves with a viscoelastic behavior prediction model based on Schapery equation were also performed. It was noted a modification of the material behavior regarding the strength, stiffness and deformation, showing that these properties were affected for the time and tension level, especially above the room temperature. The prediction model was capable to represent the creep behavior, however the equations components may change with the applied tension and the elapsed time of test.

1. Introduction

For polymeric materials, by applying a step load that remains constant over time, an elastic action occurs due to the loading, followed by creep, with a slow and continuous increase of strain at a decreasing rate of change of strain with time, in other words, a slowing of the increase of strain When the stress is removed there is a fast elastic recovery, followed by a recovery at a continuously decreasing rate. However, if after a long time a measurable portion of this strain does not disappear, with this residual strain being affected by the load time, then this strain is called viscoplastic. Viscoelastic solids may or may not show viscoplasticity [1].

The concept of linear viscoelastic behavior comes in if the stress is proportional to the strain at a certain time and the linear superposition principle, called the Boltzmann superposition principle, occur. This principle, though simple is important of physics of polymers, because it defines the creep compliance creep in relating the tensile stress with strain time in a creep test, but is valid only for low levels of stress and strain [2]. If the strain exceeds the value of one or two percent, or even lower values of strain, in most viscoelastic material the behavior exhibits non linearity [3].

1.1. Model of creep behavior

The constitutive equation of single integral developed by Schapery [4] from the thermodynamic theory is among the most widely applied in creep behavior studies for polymeric composites, which can accurately describe the non linear time-dependent behavior in many types of polymers and polymer composites. The Schapery equation uses linear (modulus or compliance) and non linear (four functions dependent on the strain or stress) viscoelastic properties. For uniaxial load, this equation presents a very similar form to the Boltzmann´s integral superposition, which is used in the linear viscoelastic theory, resulting in a simple application method for materials characterization [5]. The Schapery constitutive equation that describes the non linear behavior under constant uniaxial loading and an isothermal condition, is shown in Equation 1:

$$\varepsilon(t) = g_0 D_0\, \sigma_0 + g_1 \int_0^t \Delta D(\psi - \psi') \frac{d(g_2 \sigma)}{d\tau} d\tau \qquad (1)$$

Using a series of considerations [5, 6, 7], Equation 1 can be reduced as shown in Equation 2, with the transient and instantaneous components of creep strain (ε_c) being described according to the applied stress (σ_0):

$$\varepsilon_c = \left[g_0 D_0 + C \frac{g_1 g_2}{a_\sigma^n} t^n \right] \sigma_0, \quad \text{to:} \quad 0 < t < t_a \qquad (2)$$

For low level stress, composite materials can exhibit a linear response, in which the functions g_0, g_1, g_2 and a_σ are assumed equal to unity. Thus Equation 2 can be reduced to:

$$\varepsilon_c = \left[D_0 + C t^n \right] \sigma_0 \qquad (3)$$

Grégory et. al. [8] used the Equation 3, rewriting it to the non linear regime as follows:

$$\varepsilon_c = \left[D'_0 + C' t^n \right] \sigma_0 \text{, with } D'_0 = g_0 D_0 \text{ and } C' = \frac{g_1 g_2}{a_\sigma^n} C \qquad (4)$$

These authors used the Equations 3 and 4, in the form of a power equation, $y=a+bx^c$, to describe the composite material behavior in the linear and non linear range, respectively. The curves from these equations were adjusted to the creep compliance curves, obtained from the practices creep curves, obtaining the terms a, b and c for each level of applied stress, in other words, D_0, C and n for the Equation 3 and D'_0, C' and n for the Equation 4. Making a comparison of these results with the experimental data, the authors concluded that for linear regime both terms C and n show a very close values, enable its to use as constants, as specified by the Schapery model. However, for non linear regime, they found the terms C' and n like dependent on the stress, with the first growing exponentially and second decreasing linearly with increasing applied stress. Thus, proposed rewrite Equation 4 in the form:

$$\varepsilon_c = \left[D'_0 + C(\sigma_0)\; t^{n(\sigma_0)} \right] \sigma_0 \qquad (5)$$

With the values of the constants C and n and the terms $C(\sigma_0)$ and n, Grégory et al. constructed curves to show the variation of this constants and terms due to applied stress, with the purpose of prediction the creep behavior.

To facilitate the interpretation and presentation of the results, there was a manipulation in Equations 3 and 5, in order to make them respectively as shown in Equations 6 and 7, moving the applied stress (σ_0) to the left side of the formula in both equations and considering that $D(t) = \varepsilon(t)/\sigma_0$ during the period under creep. Thus, for the linear range, the Equation 3 became:

$$D(t) = D_0 + Ct^n \tag{6}$$

For the non linear range, Equation 5 became:

$$D(t) = D'_0 + C(\sigma_0)\, t^{n(\sigma_0)} \tag{7}$$

2. Experimental

A unidirectional carbon/epoxy composite was manufactured wound the fiber onto a spindle-shaped flat rectangular mandrel with two working faces. The reinforcement used, considering the intended performance and the filament winding technique, was the HT300 carbon fiber. This carbon fiber was supplied in continuous roving with 6000 filaments, each filament with an average diameter of 7 µm. The matrix, whose viscosity and pot life should be appropriate to manufacture components by the filament winding process, was an epoxy DGEBA resin, a methyltetrahydrophthalic anhydride as hardener and a benzildimethylamine as accelerator. The matrix cure was carried out in an oven at atmospheric pressure and temperatures up to 150 ºC, with slow cooling in the air at oven. After cooling, the plates were released from mold and the cut was performed using a diamond saw. According to the purpose of this study, the plates were cut perpendicular to the winding direction in order to get specimens at 90º, considering as 0º de fiber direction, as illustrated by FIG 1. The specimens geometry and dimensions were obtained in accordance ASTM D3039/3039M [9] and were used cardboard tabs of 1.3 mm thick.

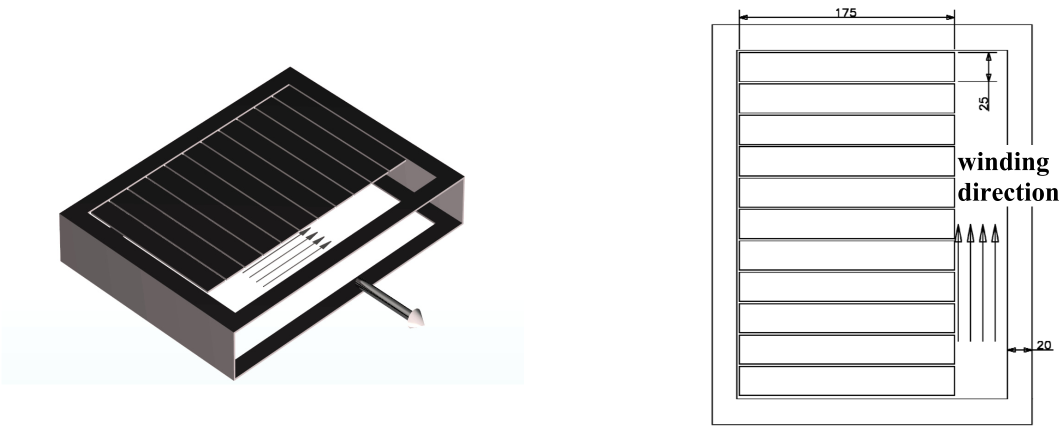


Figure 1 – Carbon/epoxy plate sketch and cutting of the specimens

Composite viscoelastic behavior at 25 and 70 °C was gotten by creep and creep rupture tests by tensile at a constant load, following ASTM D2990-95 [10], which recommends measurements of the strain versus time in specimens subjected to constant load and temperature. For the tests, it was designed a dead weight equipment with a load factor of 8:1, as illustrated by FIG 2. The load applied was a fraction of the ultimate tensile strength obtained from the tensile tests at the temperatures of 25 and 70 ºC. The tests were performed in specimens for 1000 hours or until the occurrence of rupture, featuring a creep rupture test. It was used a specimen for each different stress and temperature. The procedure was to heat the chamber that contained the specimen until the test temperature was reached and maintained the specimen without load for at least 1 hour [11].

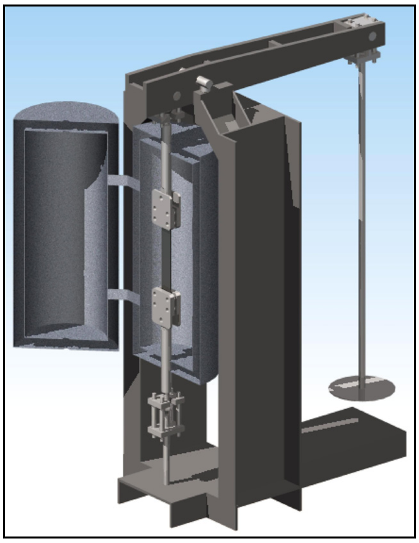


Figure 2 – Illustration of the dead weight equipment.

3. Results and discussion

3.1. Specimen characterization

Static tensile tests were performed at temperatures of 25 and 70 °C and the values were, respectively, for static rupture stress of 62.5 MPa and 54.0 MPa for the fracture strain of 0.56% and 0.55% and the calculated modulus of 10.04 GPa and 9.58 GPa.

3.2. Creep rupture

The rupture of the specimens performed at 70 °C, shown in Figure 3 in logarithmic scale, followed a linear trend with the stress decrease, as presented by Brinson [12] that related the tests to the R-W theory. This theory suggests the existence of a stress plateau which below it the fault does not occur. The curve in Figure 3 did not show the slope would indicate the existence of this plateau. However, it is important to mention that for stresses below the line, specimens did not break during the test.

The graphic presentation of the creep rupture test at 25 ºC was unrepresentative because the variation in time to rupture. A variation of results was also found by Raghavan and Meshii [13]. In the present study there was not specimens break before the 1000 h to a stress corresponding to the 55% of σ_{rupt}, Above this value to σ_{rupt}, were observed rupture of the specimens.

For the creep rupture tests, the influence of the temperature on the composites behavior can be observed, once specimens to 90° showed stress reduction that led to rupture before 1000 h for both static rupture fraction as for absolute stress, when the temperature rose from 25 to 70 ºC.

Also, the increase in temperature caused higher strain values at rupture when comparing the similar ratio of σ_{rup} values.

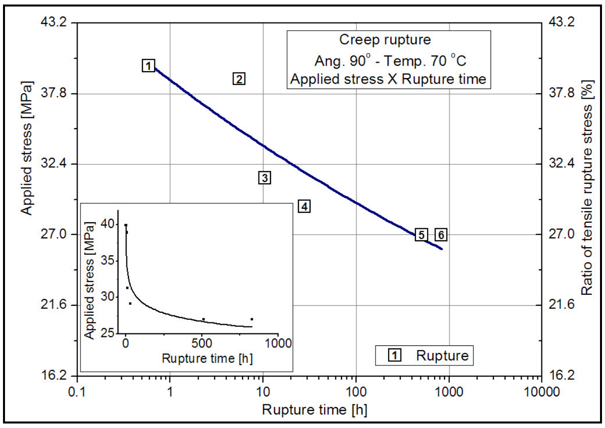
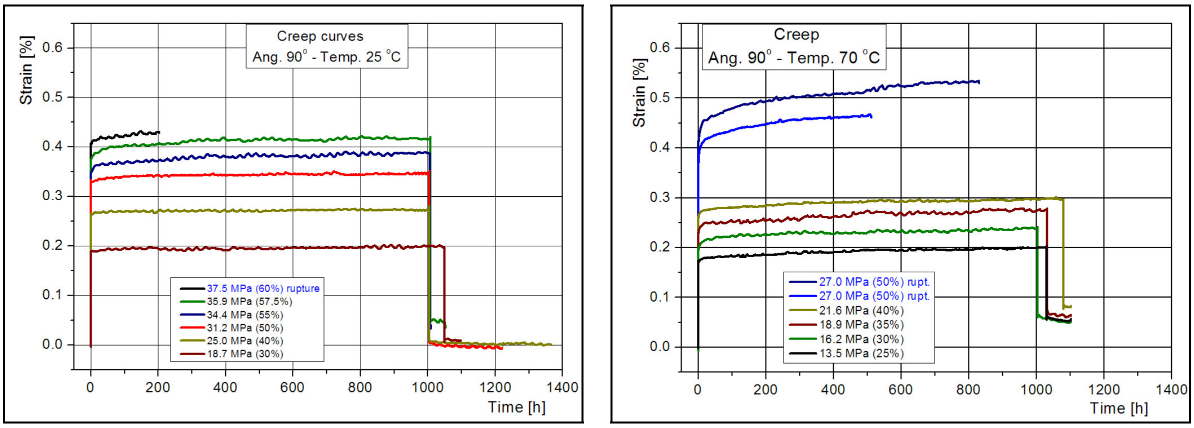


Figure 3 – Testing of creep rupture for the specimens to 90° in 70 °C represented time to rupture like a function of applied stress.

3.3. Creep

3.3.1. Creep curves

The curves of strain versus time, shown in Figure 4, are the results of creep tests for the composite oriented at 90° at 25 and 70 ºC. In the figure, additional creep rupture curves are shown, although rupture had occurred before 1000 h, the curves provide a comparison in the analysis of the compliance results.



a) b)

Figure 4 – Creep and creep rupture tests for specimens at 90°: a) 25 °C; b) 70 ºC.

In the tests, it was observed a time-dependent strain in response to the applied load, with higher strain values with the increasing stress and temperature. Additionally, strain rates decreasing with time and the absence of the second and third stage of creep.

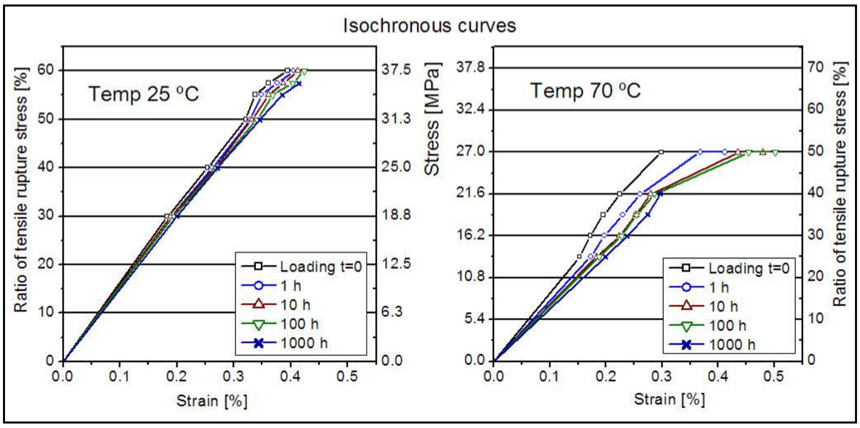
Concluded the scheduled time for the creep tests and accomplished the removal of loading, the curves showed the instantaneous elastic recoveries values close to those obtained at the time of loading, called the instantaneous elastic strain, which is reversible and disappears with the removal of the load. This is attributed to the proximity of instantaneous elastic strain and instantaneous elastic recovery, in the linear behavior, due to the absence of damage and crack growth. [14]

A comparison between temperatures was made for the strain values. Considering absolute applied stress of same magnitude, could be found that creep behavior is function of the increasing temperature, there are higher values of time-dependent strain, i.e., viscoelastic, in the instant time just before the removal of loading, at the temperature of 70 °C. However, when one compares similar ratio of σ_{rupt} between two temperatures, there were no significant changes in the viscoelastic strain at the time just before the removal of loading,

3.3.2. Isochronous curves

The transition from linear to non linear was demonstrated through the isochronous curves plotted from strain values in equal times at various stress levels, shown in Figure 5. To allow a better comparison values between the temperatures, the graphs show the same scale in the ordinate axis, which indicate the applied stress. For verification of this proportionality, it was used a graphical method from a linear approximation curve, that was constructed from the strain value in a fixed time, for various levels of loading.

For the composites at 25 °C, the transition from linear to non linear behavior occurred for stress above 34.4 MPa (55% of σ_{rupt}), while at 70 °C this transition occurred above 21.6 MPa (40% of σ_{rupt}). If one looks isochronous curves, it could see again that the increase in temperature affects the composite behavior by reducing the values of the stresses transition from linear to non linear behavior, both with respect to absolute stress as compared to the fraction of the static rupture stress (σ_{rupt}).



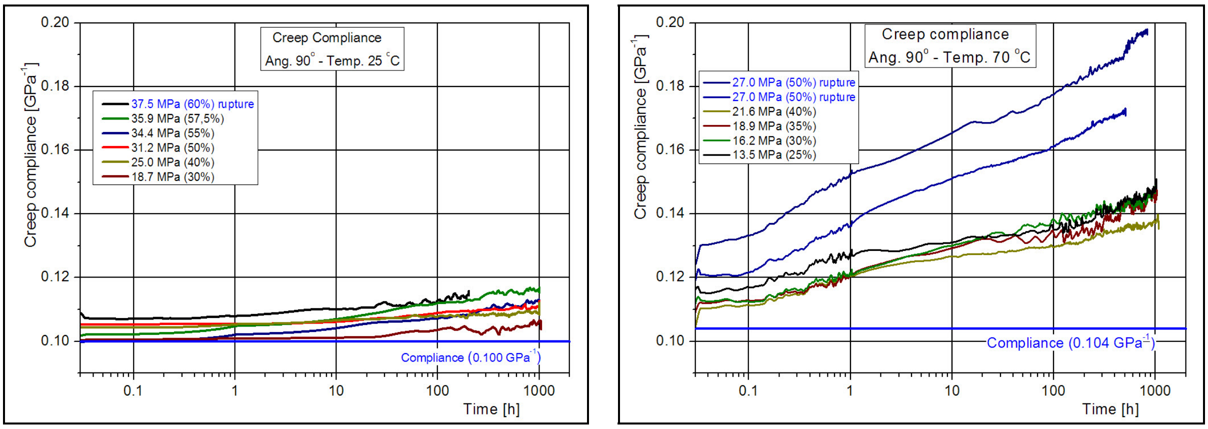
a) b)

Figure 5 – Isochronous creep curves for specimens at 90°: a) 25 °C; b) 70 °C.

3.3.3. Creep compliance

Another analysis that was done in this work was the determination of the transient creep compliance in tensile, calculated by the relationship between the viscoelastic strain and the applied stress, as shown in Figure 6. To allow a better comparison values between the temperatures, the graphs show the same scale in the ordinate axis, which indicate the creep compliance.

First, there was a growing trend of creep compliance over time for all test conditions and more pronounced variations occur for the tests to 70 °C, showing a temperature dependence of this property. As reported by Ahci and Talreja [15], the creep compliance curves in the linear range remained close to each other and could be coincidental if there were no occurrence of cracks. The curves obtained in the present study demonstrates a proximity and coupling between them during the linear range, however with an increasing of applied stress and the transition to the non linear range the separation and detachment of the curves occurred, which were in agreement with those reported by the authors cited. This grouping of the curves in the linear range could be seen in the curves for 25 ºC between 30 and 55% of σ_{rupt} and 70 ºC between 25 and 40% of σ_{rupt}.

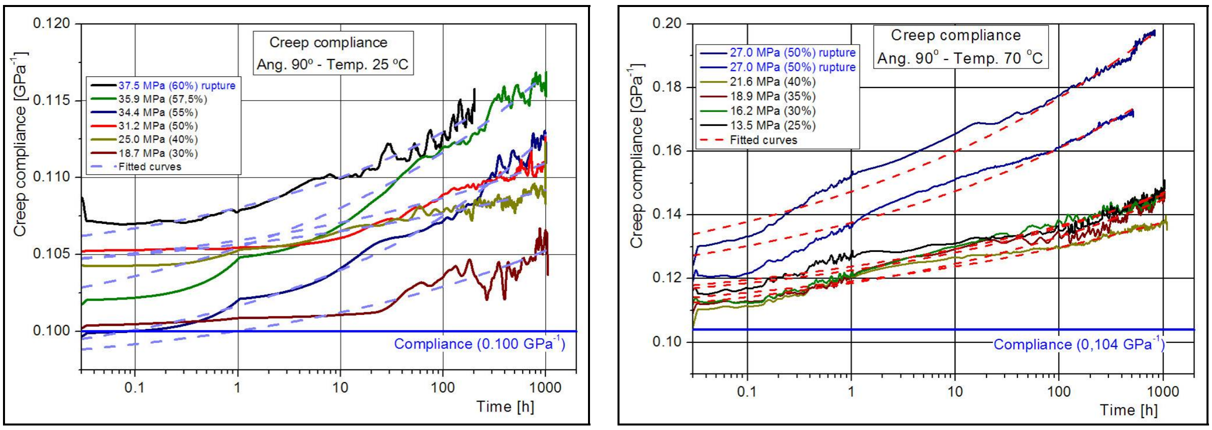


a) b)

FIGURE 6 - Creep compliance for specimens at 90°: a) 25 ºC b) 70 °C.

3.3.4. Creep behavior prediction model

Fitted curves by the method proposed by Gregory et al. and their practices compliance curves are shown in Figure 7 for all composites. One can check that there was good agreement between each pair of curves, fitted and practices, to each applied stress, specially for time over 100 h, where there are the largest number of data points, determining a closest approach between the curves. Therefore, the time elapsed in the test have influence on the values of C, n, $C(\sigma_0)$ e $n(\sigma_0)$, similarly as quoted by Zaoutsos et al. [16], who found the n term in the Schapery equation dependent of elapsed time under creep test.

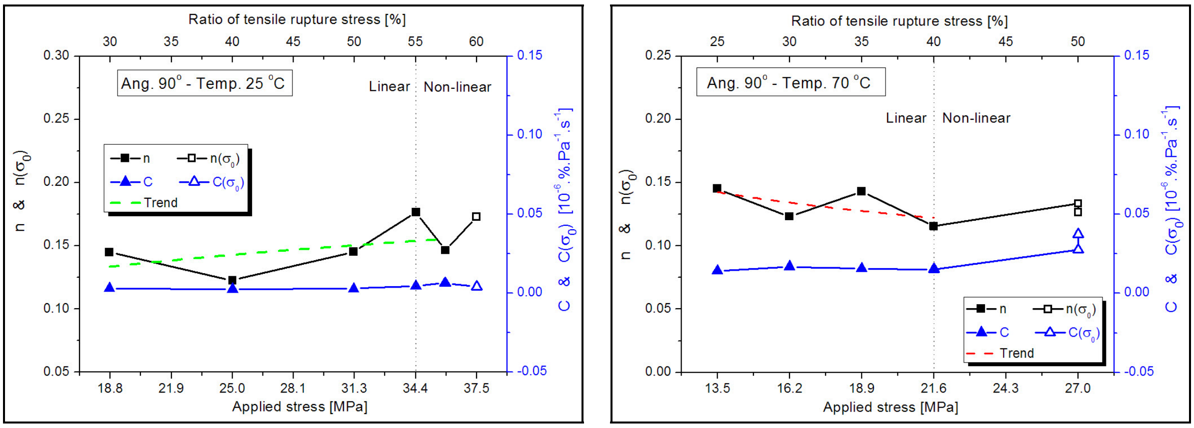


a) b)

Figure 7 - Creep compliance curves and their theoretical approaches curves to specimens at 90°: a) 25 °C; b) 70 °C.

It is worth to remind that the fitted curves shown in Figure 7, were obtained from the creep compliance curves and used to obtain the values of the terms C, n, $C(\sigma_0)$ and $n(\sigma_0)$ for each applied stress. The good agreement among each pair of curves showed that the Equations 6 and 7 were able to represent the creep behavior. However, for the use of this procedure in prediction of the creep behavior it is necessary to determine the variation trend of these values and its dependence on the stress and temperature.

In Figure 8 are shown the curves obtained from the linear terms *C* and *n* and non linear terms $C(\sigma_0)$ and $n(\sigma_0)$ from the fitted curves presented previously. It was used the same scales for the ordinates axis for all graphs in order to allow a better comparison between them. The *C* and *n* values showed a trend almost constant. This trend almost constant was expected for the linear range, in which theoretically the creep compliance curves should be coincident. Thus, it can be correct to consider the linear terms *C* and *n* as constant. The small number of data for $C(\sigma_0)$ e $n(\sigma_0)$, in both temperatures, made it impossible to determine the variation trend of these non linear terms with the applied stress, as proposed by Grégory et al., but they presented values of the same order of magnitude of the linear constant, *C* e *n*.



a) b)

Figure 8 – Behavior of the linear constants *C* and *n* and non linear terms of $C(\sigma_0)$ e $n(\sigma_0)$ for specimens at 90°: a) 25 °C; b) 70 °C.

4. Conclusion

Creep rupture tests under constant tensile loads confirmed the dependence of the composites properties related to time, showing that this parameter influenced directly the material failure, even for stresses below the static rupture limit, especially under the temperature increase. Carbon/epoxy composites at 90º showed creep rupture just above the beginning of non linear behavior. So, to predict the creep behavior, only the linear range could be considered. Creep compliance analysis showed once again the time-dependent behavior of the composite studied and its dependence with the temperature increase. In that analysis, the trend of grouping of the curves can be verified in the linear range.

The verification of time-dependent behavior was conducted by the adequacy of the method proposed by Grégory et al., based on the Schapery single integral equation. The proposed equations proved to be able to represent the creep compliance behavior, however should be taken into account determining the terms of these equations, besides the variation of the applied stress and the elapsed test time.

For the creep and creep rupture tests, the material properties was affected by time, especially when associated with working temperature above the room temperature.

5. Acknowledgement

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6. References

[1] SCHAPERY, R.A. Nonlinear viscoelastic solids, International Journal of Solids and Structures, 37, 359-366 (2000).
[2] SHAW M.T., Macknight W.J.: Introduction to polymer viscoelasticity, 3rt Ed. John Wiley & Sons, Inc (2005).
[3] Findley W.N., Lai J.S., Onaran K.: Creep and relaxation of nonlinear viscoelastic materials, with an introduction to linear viscoelasticity, 2.ed., North-Holland Publishing Company (1976).
[4] Schapery R.A.: On a thermodynamic constitutive theory and its application to various nonlinear materials. IUTAM Symposium on thermoinelasticity, East Kilbride, UK, Proceedings…, p. 259-284, 25-28 June (1968).
[5] Lou Y.C., Schapery R.A.: Viscoelastic characterization of a nonlinear fiber-reinforced plastic, J. Composite Materials, 5, 208-234 (1971).
[6] Papanicolaou G.C., Zaoutsos S.P., Cardon A.H.: Prediction of the non-linear viscoelastic response of unidirectional fiber composites, Composites Science and Technology, 59, 1311–1319 (1999).
[7] Papanicolaou G.C., Zaoutsos S.P., Cardon A.H.: Further development of a data reduction method for the nonlinear viscoelastic characterization of FRPs, Composites: Part A, 30, 839–848 (1999).
[8] Grégory A., Vogel D.; Béguelin PH., Gensler R., Kausch H.H., Jaeger M., Van Buren A.: A viscoelastic analysis of the creep behaviour of chopped strand mat e-glass fibre reinforced vinylester resin, Mechanics of Time-Dependent Materials, 3, 71-84 (1999).
[9] ASTM D3039/3039M – 95a: Standard test methods for tensile properties of polymer matrix composite materials (1995).
[10] ASTM D2990 – 95: Standard test methods for tensile, compressive, and flexural creep and creep-rupture of plastics (1995).
[11] Masuko Y., Kawai M.: Application of a phenomenological viscoplasticity model to the stress relaxation behavior of unidirectional and angle-ply CFRP laminates at high temperature, Composites: Part A, 35, 817–826, (2004).
[12] Brinson H.F.: Matrix dominated time dependent failure predictions in polymer matrix composites, Composite Structures, 47, 445-456, (1999).
[13] Raghavan J., Meshii M.: Creep rupture of polymer composites, Composites Science and Technology, 57, 375-388 (1997).
[14] Haddad Y.M.: Viscoelasticity of engineering materials, Chapman & Hall, New York (1995).
[15] Ahci, E.; Talreja, R.: Characterization of viscoelasticity and damage in high temperature polymer matrix composites, Composites Science and Technology, 66, nº 14, 2506-2519 (2006).
[16] Zaoutsos S.P., Papanicolaou G.C., Cardon A.H.: On the non-linear viscoelastic behaviour of polymer-matrix composites, Composites Science and Technology, 58, 883-889 (1998);