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Published in:
Composites Part C: Open Access

DOI:
10.1016/j.jcomc.2020.100051

Published: 01/11/2020

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Stress relaxation, creep, and recovery of carbon fiber non-crimp fabric composites

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A R T I C L E   I N F O

Keywords:
Creep
Glass transition
Structure-property relations

A B S T R A C T

Considering that structural composites are typically composed of off-axis plies, i.e. quasi-isotropic stacking sequence, their strength and stiffness are time-dependent due to the viscoelastic character of polymer matrices. This work consists of determining creep, recovery, and stress relaxation of carbon fiber-reinforced polymer (CFRP) composites. Long-term experimental analyses are conducted via dynamic mechanical analysis under several temperatures and stress levels. From the experimental observations, the changes in relaxation mechanisms are predicted using Fancéy's latch model. The rate of relaxation at different temperatures is also covered. Since at certain strain levels the viscoelastic behavior cannot be properly determined, the stress-relaxation is determined using the time-temperature superposition (TTS) principle, considering nine temperatures at three strain levels in order to cover the three main regions of the composite system (glassy, glass transition and rubbery regions). The models and experiments herein presented can be extended to any polymeric system.

1. Introduction

Lightweight components along with suitable mechanical performance make fiber-reinforced polymeric composites the best candidate for marine, aeronautical and aerospace structures [1–4]. Structural composite materials need to attend short- and long-term design requirements. Among short-term requirements, some mechanical properties of interest are flexural [5], tensile [6,7], compression [8], interlaminar shear strength [5,9] and impact [9]. Concerning long-term tests, two widely accepted test methods are stress relaxation and creep [10–14]. According to Sreekala et al. [15], the knowledge of stress relaxation behavior under different strain levels allows predicting the dimensional stability of load-bearing structures and the retention of force (by modulus) for bolts fastened to composites. In case the unrecovered strain is too large after removing the applied stress on the material, it might have decreased dimensional stability and even lead to structural failure. The recovery after some imposed stress is also important and gives important information about elastic and anelastic recovery. Depending on loading stress and temperature, total recovery can be achieved [16–18].

Changes in the mechanical properties of composite materials are related to changes in viscoelastic behavior [15,19,20] being mainly dependent of stress (creep) and strain (stress relaxation) levels and temperatures. The sensibility of microstructural changes is dependent on the test method. Creep/recovery and stress relaxation tests have more sensibility in comparison to tensile, flexural and impact, for instance. Hence, realistic physical aspects related to microscale sometimes are neglected for macrostructural determination [21]. Since macroscale mechanisms are reflections of microstructural changes, tests performed at low stress/strain levels, in spite of not represent a realistic stress/strain application, can help elucidating the microstructural behavior which can allow predicting the mechanical behavior of composite laminates at lower costs and keeping the accuracy on the achieved data [22].

Most of the studies regarding stress-relaxation studies in polymeric composites consider rubber or thermoplastic matrices. George et al. [23] studied short cellulose fiber reinforced natural rubber composites and reported that stress relaxation behavior is dependent of rubber/fiber adhesion. Stress relaxation behavior of natural rubber/polystyrene, sisal/natural rubber and pineapple/polyethylene composites is presented in Refs [23–25], and fiber orientation, fiber/matrix interaction and fiber loading influence the relaxation mechanisms. Differently than for vegetable fiber-reinforced composites [15,26], structural composites have microstructural changes related to the resin as reflection of stress/strain variation. If the imposed stress/strain exceeds maximum resistance of the composite, matrix/fiber debonding,
fiber breakage and destruction of matrix interlayers between fibers can occur. When a stress/strain is fixed, relaxation in strain (if stress is fixed) or stress (if strain is fixed) is mainly consequence of molecular-level rearrangements, fiber alignments, decreased fiber/matrix bonding etc. and depends on temperature and strain/stress levels. If it is considered that chain segments can be stress activated by different chain segments sizes which generates higher/lower deformations with different energies [16,18]. In the glassy, glass transition and rubber states it is expected that different activation segments occur, whose behavior is yet to be fully understood.

In this context, this study aims at determining stress relaxation and creep/recovery characteristics of high-performance carbon/epoxy composites in details. Changes in the relaxation mechanisms are studied using predictive models proposed by Fancye [16,17] and Cox [27]. Variations in relaxation with stress and temperature for creep/recovery and strain for stress relaxations are also examined. The rate of relaxation at different temperatures is calculated in order to explain gradual changes in the relaxation mechanism. Considering that at a given strain level the complete viscoelastic properties cannot be precisely determined, the stress-relaxation obtained data at different strain levels are superimposed by a horizontal translation along the logarithmic time-axis to overcome this limitation.

2. Experimental

Non-crimp carbon fiber (Saertex, with Hexcel carbon fibers IM7 12K) and epoxy resin (PRISM EP2400) are processed using vacuum-assisted resin transfer molding (VARTM). The fabrics are arranged consisting of stacking 16 layers of a quadraxial architecture of [0/90/±45]2s. The fiber volume fraction of the composite is ~60%, void content is ~3.4%, with a density of 1.53 g.cm−3. Before processing, the epoxy resin was vacuum degassed to remove air bubbles, and during the processing, a vacuum of 5.5 × 10−2 mPa is applied in the mold to prevent voids and to assist resin flow. The temperature of the mold and injection is of 100°C and curing is carried out at 180°C for 2 h, also in the mold. The final laminate has thickness of 3 mm [28] (Fig. 1).

Stress relaxation tests are performed in a DMA Q800 equipment from TA Instruments using a three-point bending clamp. A 10-min soak time is applied to ensure that the specimen reaches equilibrium after each temperature for all different procedures. During the test, relaxation modulus \(G(t)\) in time function data are collected, and the results are plotted against time. Specimens of average dimensions 35 × 10 × 3 mm³ are used for the experiments. For stress relaxation TTS tests, nine different temperatures (from 30°C to 190°C in steps of 20°C) at three strain levels (0.1, 1 and 5%) are carried out covering all three main regions of the composites (glassy, glass transition, and rubbery regions).

Creep/recovery tests are performed using the same clamp, at three stress levels: 1, 2.5 and 5 MPa. In each stress level, three different temperatures are applied: 50°C, 150°C and 210°C, aiming at covering the glassy, glass transition and rubbery states, respectively. A 15-min creep is followed by 30-min recovery at each stress level and temperature.

Weibull- and Eyring-based models are applied on stress relaxation and recovery (from creep/recovery) curves due to similarities in the shape of the curves.

3. Predictive viscoelastic models

Stress relaxation is investigated by using Weibull and Eyring models aiming to obtain microstructural information under different strain loads and temperatures. More precisely, the distribution of viscoelasticity elemental failures and the energies involved in the process, respectively [16,18]. Afterwards, the time-temperature-superposition principle (TTS) is applied using three different strain levels.

Just as in the experiments, creep/recovery is simulated at 50°C, 150°C and 190°C, and with 15-min creep and 30-min recovery for all conditions.

3.1. Viscoelastic modelling

The Weibull equation for stress relaxation \((\sigma_{rel}(t))\) is given by Eq. (1) while Eyring equation is given by Eq. (2):

\[
\sigma_{rel}(t) = \sigma_0 \left[ \exp \left( -\frac{t}{\eta_s} \right) \right] + \sigma_f \tag{1}
\]

\[
\sigma_{rel}(t) = A \tanh \left( B \exp \left( -\frac{t}{\tau} \right) \right) + \sigma_c \tag{2}
\]

where from Eq. (1), \(\sigma_0\) is the time-dependent stress, \(\eta_s\) is the characteristic life, and \(\beta_\sigma\) is the shape parameter. \(\sigma_f\) is the final stress as time \(t\) approaches infinity. From Eq. (2), \(A\) and \(B\) are constants and \(\tau\) is the relaxation time. The parameter \(\sigma_c\) is the final stress as \(t\) approaches infinity.

The use of both equations was first proposed by Fancye [16,17], who reported that viscoelastic changes occur due to external forces/excitations and they can be correlated with incremental jumps. Mathematically, the Weibull function is identical to Kohlrausch-Williams-Watts (KWW), i.e., an empirical stretched exponential function. On the other hand, KWW can be related to the motion of matter to molecular jumps (potential energy barrier) developed by Eyring. As stress relaxation and viscoelastic behavior of polymers are thermally activated processes acting in parallel (Maxwell represented by the spring and Voigt-Kein by the dashpot), each of the two processes is composed by a Maxwell element and an Eyring one (instead of a conventional dashpot).

Polymeric materials exhibit time-dependent behavior and hence, limited laboratory tests need to be extrapolated from shorter to longer-term times if more real conditions need to be achieved. The data obtained from short times can be treated by using a method of reduced variables which overcomes this issue. This is possible since the viscoelastic response at high temperatures is identical to the response at the low temperatures for a longer time. It is assumed that the processes involved in molecular rearrangements are temperature-dependent, i.e., it occurs at decelerated rates at lower temperatures and vice-versa and that temperature is directly equivalent with frequency of measurement (time).

The most-widely and successfully formulation used for this proposal is developed by Williams, Landel and Ferry and it is well-known as WLF
Weibull equations

\[ \epsilon_{ctot}(t) = \epsilon_i + \epsilon_c \left[ 1 - \exp \left( - \left( \frac{t}{\eta_c} \right)^{\beta_c} \right) \right] \]

\[ \epsilon_{rvis}(t) = \epsilon_r \left[ \exp \left( - \left( \frac{t}{\eta_r} \right)^{\beta_r} \right) \right] + \epsilon_f \]

Fig. 2. Scheme of creep/recovery tests based on [16,18].

\[ \log a_T = \frac{-C_1(T - T_0)}{C_2 + (T - T_0)} \]  \hspace{1cm} (3)

where \( a_T \) is the horizontal (or time) shift factor, \( C_1 \) and \( C_2 \) are constants, \( T_0 \) is the reference temperature (K) and \( T \) is the testing temperature (K).

WLF relation considers that as the free volume of the material increases, viscosity rapidly decreases. In addition, from glass transition temperature, \( T_g \), the fraction of free volume increases linearly with temperature.

Other analytical models can also be used to study stress relaxation behavior of composite materials. It is considered that underlying changes in the matrix shear modulus are responsible for the changes in stress relaxation rate induced by reinforcement in the polymeric matrix. The model was derived by incorporating the time-dependent shear modulus of the matrix in to Cox’s shear-lag Eqs. (4),(5) [27].

\[ E_{\epsilon}(t) = V_m E_m(t) + V_f E_f \left( 1 - \frac{\tanh(n(t)\beta)}{n(t)\beta} \right) \]  \hspace{1cm} (4)

\[ n(t) = \left[ \frac{4}{E_f ln \left( \frac{\rho_f}{\rho_m} \right)} \right]^{1/2} \]  \hspace{1cm} (5)

where \( E_m, E_p \) and \( V_m \) are the matrix and fiber moduli, and matrix and fiber volume fraction, respectively. According to the Equation, the relaxed modulus in any point in time is related the above parameters mentioned. The time-dependence of matrix shear modulus \( G_m(t) \) is represented by the effectiveness factor \( n(t) \), which defines the rate at which the stress is transferred between the matrix and fiber. According to Obaid et al [11], the advantage of this model is that \( n(t) \) is the underlying cause of the changes in stress relaxation. If the model is validated by experiments, then it can be used to make very practical predictions, such as dependence of stress relaxation on reinforcement architecture.

Creep/recovery curves are fitted following a Weibull distribution function, as shows Fig. 2 in a schematic manner. The terms of the equations describing the curve (red color for creep and blue color for recovery) are \( \epsilon_i \) - initial instantaneous strain from application of the load and \( \epsilon_r \) - function represents creep strain, which is determined by the characteristic life \( n(t) \) and shape \( \beta(t) \) parameters as a function of load duration. When the sample is unloaded, there is an instantaneous (elastic) strain recovery, which is then followed by time-dependent recovery strain where \( \epsilon_f \) is the remaining contribution from viscoelasticity and \( \epsilon_i \) of the viscous flow.

4. Results and discussion

Previous results related to dynamic mechanical runs with fiber volume fraction, density and void content are reported in Fig. 3 [30]. Storage modulus, \( T_{onset} \) (obtained from the beginning of the storage modulus decay) and Tan \( \delta \) are used to set up stress-relaxation test. Fiber
volume fraction was 60%, to ensure application as primary structures. Meanwhile, processing parameter enables low porosity fraction along the laminate.

The impregnation behavior can be observed in Fig. 4, in which the plain weave return means high impregnation homogeneity and lower percentage means defects. The outcome is between 20 – 40 % of weave return, which is in accordance with the literature [6,31]. This behavior ensures that the laminate has appropriate impregnation along the laminate and defects such as porosity variation, fiber misalignment, and resin-rich regions have no influence on stress-relaxation tests.

4.1. Stress relaxation

Fig. 5 a-c) shows the stress relaxation results. It is observed small differences in G(t) at different strain levels – all G(t), i.e., ~1E+10 Pa at the glassy region. All G(t) decrease with increasing temperature due to higher molecular mobility. A higher difference is observed from T_g for all strain levels, in which an abrupt decrease in modulus is seen. This change in step can be attributed to considerable changes in discrete relaxation times in which from this region relaxation spectra is not constant, having both time and temperature dependencies [29,32].

Differences in G(t) (error of ±5) can be found in typical DMA measurements [33–36]. Hence, it becomes interesting to scale relations to help rationalizing observations when different materials and/or test conditions are used [37]. Fig. 5d) shows a normalized curve at 50°C, 150°C and 190°C. The normalization follows this expression: (ε − ε0)/(ε_f − ε0), where ε is the current strain; ε0 is the instantaneous (initial) strain; and ε_f is the final strain. Small differences are observed for a strain level only at the rubbery state and it is not attributed to strain level but with intrinsic the error of measurement.

Table 1 presents the values obtained by using Weibull-based and Eyring models aiming to elucidate the results obtained for all strain levels at three selected temperature representing glassy, glass transition temperature and rubbery states. All models presented excellent fit for all temperature and stress used (Fig. 6 a-b).

Regarding Weibull parameters, σ_f and σ_0 present lower values by temperature. The relaxation processes in polymer-based materials are facilitated by temperature in which occurs molecular rearrangement of polymer chain segments aiming to decrease internal stresses. At the T_g, the values follow an anomalous behavior, showing negative values. At the glassy region, most of the segments are frozen-in while in the rubbery region most of the segments are in a viscous state. At T_g, the frozen-in segments dissipate energy as heat and the chain segments gain considerable molecular mobility (much higher proportionally in relation to other two regions). This phenomenon can contribute to this anomalous effect in the model fitting at T_g. η_g and β_g parameters also tend to decrease with increasing temperature. According to the latch model proposed by Fancy [16,18] the failure of elements in a system is modelled as mechanical latches which are time-dependent: each latch fails once it is triggered, with the recovery proceeding by one incremental step. In a molecular level, the viscoelastic deformation occurs though incremental jumps, i.e., polymer chain segments jump between positions of relative stability. As high temperature facilitates chain mobility and consequently rearrangement of polymer segments, more segments with different sizes are activated occurring ‘failure’ of the latches in a broader range. On the other hand, at the glassy region, more segments must be activated for ‘failure’ of the latches, increasing β_g values. In the current study, strain levels used does not influence the values. The relaxation time (τ) of Eyring equation is used aiming to corroborate Weibull parameters. The relaxation times decreasing are attributed to higher molecular mobility as earlier described, where molecular rearrangements occur at a higher rate and consequently the potential energy for segmental jumps further. The values decreased in function of the temperature. Strain level does not significantly change the values (values at the same order of magnitude).

Aiming to estimate the behavior of the material in function of inaccessible times, time-temperature-superposition principle (TTS curves) are constructed using Eq. (3) and presented in Fig. 7. It can be observed a similar behavior for all curves, independently of the strain level applied. For all samples, it can be noted a gap in modulus after the T_g. If the material is thermos-rheologically simple [32], the distance of the shift factor (α_T) at a determined temperature is the same for all values of time. This occurs because all retardation times at determined temperature maintain a constant ratio α_T. Hence, all data recorded at a given temperature form a segment of the total response (master curve) and a simple translation of the segment along the log time axis by the amount log at will result in superposition. For a thermos-rheological complex material [29], the shift factor becomes a function of time in addition to temperature. As an evidence of this, Guedes et al. [38] created an easy-to-use algorithm in order to construct the master creep compliance curve from dynamic viscoelastic functions from a DMA analyzer. The authors conclude that for regions where the retardation spectrum is either constant or obeys a power law behavior there are reproduced quite well by the algorithm. Here, the material becomes thermos-rheologically complex from the glass transition temperature, i.e., the discrete retardation spectrum has a drastic change in relation to all spectra so far. An interesting point to observe it that by strain level, the composite becomes thermo-rheologically simple, i.e., the gap between T_g and rubbery region reduces and shifts to similar times.

4.2. Creep/recovery

Creep/recovery tests are performed at glassy (50°C), glass transition (150°C) and rubbery (210°C) regions because the molecular response is quite different at each region. At the glassy region, there is a short-range chain segment response which is primarily dependent of chain packing and intermolecular forces [39]. Each external/internal deformation (promoted by strain or stress, for example) is stored by frozen-in segments; hence, the material response is primarily elastic. At rubbery region, the viscous component plays a major role since no elastic response can be stored and the molecules are free to move [40]. The response is primarily viscous. At T_g (usually obtained from the maximum peak of tan delta curve), all the stored energy obtained from glassy region is released as heat, giving more molecular mobility as time/temperature proceeds. In this region, the response is viscoelastic.

Fig. 8a shows a representative creep/recovery curve separated in Regions I-VI for better elucidating the results. Fig. 8 (b-d) shows experimental curves at three stress levels and temperatures. For creep, higher deformation is obtained at higher stress levels and temperatures. After 15-min creep, stress is removed, and recovery is performed for 30 min. The recovery at T_g (150°C) presents a small recovery after creep when compared to test performed at rubbery state. The samples at the glassy
region present almost full recovery. More details about each region are described below:

i) Creep - Creep behavior is described by Regions I-III according to Fig. 8a. Region I represents the instantaneous deformation; the viscoelastic response after instantaneous deformation is represented by Region II; and viscous deformation by Region III. Regarding Region I, a higher instantaneous deformation is obtained in function of both temperature and stress level. A large instantaneous deformation is an indicative that many segments are oriented to some extent along
the stress direction in shorter-time, and thereafter directional hardening made it difficult to get further orientation and rearrangement of polymer chains and entanglements [41,42]. Region II seems to be more affected by temperature than stress level (referred to curve format). In the glass transition temperature, all frozen-in segments achieve high molecular mobility and hence energy dissipation like heating occurs. The response is viscoelastic, henceforth the behavior is more complex when compared to glassy and rubbery state, where the polymer response is purely elastic or viscous. The greater chain mobility achieved leads to a sharper curve in comparison to the other two temperatures, i.e., it is occurring the change from elastic to viscous state. Region III follows the same trend of region I, that is, higher value is obtained for higher temperatures and stresses.

ii) Recovery - described by regions IV-VI in Fig. 8a. Instantaneous recovery after creep is described by Region IV; viscoelastic recovery by Region V and permanent deformation after recovery by Region VI. Independently of the stress level applied, a higher value of instantaneous recovery is obtained at 190°C (rubbery state) in comparison to 150°C. At glassy temperature (50°C), the instantaneous recovery is nearly complete, independently of the stress applied. At Tg, lower instantaneous recovery is obtained for all stress levels. These results suggest that Region IV is more temperature- than stress-dependent. When viscoelastic materials are subjected to stress they undergo deformations by molecular rearrangements and by viscoelastic flow 

[19,38,43]. If the strain is completely reversed during the creep recovery phase, no irreversible deformation occurs. This behavior is described by Voigt model and occurs when the stress during the deformation phase does not exceed the elastic limit. Maxwell model describes irreversible deformation. This behavior is typical of materials that exhibit viscous flow (chain slippage) [38]. At Tg, plastic and elastic deformations occur simultaneously, so the analysis becomes more complex. The small recovery obtained can be attributed to abrupt changes in relaxation times from heat released of frozen-in segments. In this heat-release process abrupt changes in molecular rearrangement can be contributing to a very small elastic recovery. After Region IV, viscoelastic recovery (Region V) and permanent deformation (Region VI) happen. Regions V and VI are calculated using Weibull, Eyring and Cox shear-lag models with permanent deformation after recovery.

All discussions initiate by comparing glassy and rubbery regions and, consequently, glass transition values in different strain levels. The recovery parameters for the three stress levels are shown in Table 2. The best fit for all temperatures is obtained at 5 MPa. At 1 MPa and 2.5 MPa, where the curves present a poorer fit (comparative curve in Fig. 9 at 50°C), suggesting that the models are more suitable for higher stress levels. At 1 MPa and under 50°C, the results for stretched exponential show an unsuitable physical value. The parameters εr and εg increase with temperature. It is expected that an increase in temperature leads to an increase in the number of available conformational states, hence increasing the viscous flow and the creep strain. The βg parameter also increases in function of the temperature showing a reversal trend when compared to stress relaxation analysis. As temperature increases, polymeric chains gain molecular mobility, consequently the possibility of more deformation modes increases with different chain sizes. The ηg parameter decreases with temperature due to more molecular mobility achieved in the chain segments attributed to more thermal energy decreasing the characteristic lifetime due higher creep strain.

Relaxation time from Eyring model decreases with temperature when compared with glassy and rubbery region, meaning that it becomes easier to polymer chain segments to gain different available conformational modes. If different chain segments containing different sizes separated from each other on a scale of time/temperature, a broadening in the spectrum takes place. This movement requires different sizes of free volume “holes” [44,45]. At the rubbery region, the molecules segments are in quite free to move while at glassy region only a portion of the segments are capable to undergo deformations aiming to decrease internal deformation though rearrangements, i.e., the atoms are frozen-in to fixed positions, and energy is absorbed only by increasing their vibrational amplitudes [46]. At Tg, higher values are obtained when
Fig. 8. a) Creep/recovery representative curve; b) Creep/recovery curves at 50°C, 150°C and 190°C at a) 1 MPa, b) 2.5 MPa and c) 5 MPa.

Table 2
Weibull and Eyring parameters calculated for recovery test.

<table>
<thead>
<tr>
<th>Models</th>
<th>Parameters</th>
<th>50°C</th>
<th>150°C</th>
<th>190°C</th>
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<tbody>
<tr>
<td>Weibull 1 MPa</td>
<td>(\varepsilon_f (%))</td>
<td>4.74E-3</td>
<td>2.96E-2</td>
<td>2.85E+2</td>
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<td></td>
<td>(\varepsilon_r (%))</td>
<td>439.65</td>
<td>1.56</td>
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<td></td>
<td>(\eta_r (s))</td>
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<td>4.05E-6</td>
<td>9.90E-2</td>
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<td></td>
<td>(\beta_r)</td>
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<td>0.15</td>
<td>30.67</td>
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<tr>
<td></td>
<td>SS*</td>
<td>7.53E-5</td>
<td>7.85E-5</td>
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<td>Weibull 2.5 MPa</td>
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<td>SS*</td>
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<tr>
<td>Eyring 1 MPa</td>
<td>(\tau (s))</td>
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<td>Eyring 5 MPa</td>
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\(SS^* = \text{Sum of squares}\)
compared to same stress level. The anomalous effect of the glass transition cannot be described by Fancoy’s latch model but for phenomenology of theory of relaxation as presented by Scherer [46]: at lower temperatures the time to achieve equilibrium configuration increases, making any structural changes impossible. By increasing temperature, the structure falls out of equilibrium and enter the glass transition region. This “fall-out” occurs in a wide range of temperatures (depending of the test conditions), in which the average relaxation time by some two or three orders of magnitude [47] and the structure seems to “frozen” in a laboratory scale (order de 100s) followed by an abrupt fall in some thermodynamic property as volume or enthalpy for example [37]. The energy released in the process after this abrupt fall is very high when compared to glassy and rubbery states. Therefore, this anomalous effect for all models calculated at $T_g$ can be related to this phenomenon.

5. Conclusions

This study presented a complete structure versus property relationship of a high-performance composite laminate manufactured via VARTM. Creep, recovery and stress relaxation of quasi-isotropic composites were deeply investigated by means of long-term experiments at several strain and stress levels and different temperature ranges. In addition, the time-temperature superposition principle is applied to better understand the stress relaxation of the composite at its glassy, glass transition, and rubbery regions. In parallel, Weibull and Eyring analytical models are employed to predict and simulate the experimental observations more accurately.

Both experimental results and analytical predictions were successfully developed and an excellent correlation between them were found. The higher the applied strain/stress, the higher the creep/recovery of the laminates. The long-term behavior of the composites was also found as temperature-dependent, in which they deformed more for higher temperatures. The experiments and models herein developed were vital to understand the structural versus property relationship of quasi-isotropic composite laminates. Next steps of this research include evaluating the suitability of the proposed models for high strain/stress levels.

Acknowledgments

The authors acknowledge the financial support from CNPq (process number: 153335/2018-1), FAPESP (process number: 2006/02121-6) and CAPES.

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