INVESTIGATIONS ON THE CREEP/RECOVERY BEHAVIOR OF WOVEN-PLY CARBON FIBER REINFORCED PPS OVER GLASS TRANSITION TEMPERATURE

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Abstract :
This study aims at investigating the viscous behavior of carbon woven-ply PPS laminates. Its specificity consists in evaluating the contribution of PPS matrix to the creep-recovery response of C/PPS laminates at temperature higher than T_g of PPS (95 °C), where matrix viscoelasticity and viscoplasticity are prominent. The reinforcement is a balanced 5-harness satin weave and the studied stacking sequence is an angle-ply lay-up $[(+45, -45)]_7$*, in order to observe the most critical viscous behavior. Several creep-recovery tensile tests were carried out at 120°C during 24 hours of loading and 48 hours of recovery to gather pieces of information about angle-plied C/PPS long term behavior. In addition to experimental investigations, a numerical modelling has been developed in order to apprehend the time-dependent behavior. For this purpose, a linear spectral viscoelastic model and a generalized Norton-type viscoplastic model have been implemented in the FE code Cast3m.*

1 Introduction/State of art

In high-technology industry such as aeronautics, composite materials offer a large number of perspectives with their high strength/weight ratio or their forming abilities. Over the last four decades, thermosetting (TS) matrix composites have been widely used in despite of some manufacturing issues such as difficult raw material storage, irreversible and long curing processes. In such context high performance thermoplastic (TP) matrix composites are presenting promising alternatives to TS drawbacks (recycling abilities, short time processes, …). PPS is one of the most widely studied high-performance TP. This work follows an experimental study [1] on the mechanical properties of TP based composites reinforced with carbon fibers under severe conditions (120°C and hydrothermally aged). Further expansion of TP-based composites relies on the knowledge of their long-time behavior (fatigue, creep or relaxation). That's the reason of this work on the investigation of the viscous behavior of carbon woven-ply PPS laminates.

In the literature, creep behaviour of TP composite has almost exclusively been investigated on UD laminates. Several parameters have been identified to have a large effect on the viscous behaviour of TP matrix composites such as the stacking sequence [2; 3], the reinforcement architecture [4], the solicitation type [5; 6], the matrix crystallinity [7], or the temperature [2].

2 Experimental investigations

2.1 Materials

The composite material under study is a carbon fabric reinforced PPS prepreg laminate, supplied by Porcher. Each ply consists of a balanced fabric with 0 and 90° oriented fibers. The reinforcement is a 5-harness satin weave of T300 3K carbon fibers supplied by Soficar and the matrix is a high performance TP (PPS) supplied by Ticona company, referenced as Fortron 0214. The matrix has a glass transition temperature of 95°C and a crystallinity of 30 % (DMC and DMTA tests were performed at the Crismat/CNRT laboratory in Caen). The volume and mass fraction of carbon fibers are respectively of 50% and 57%. Prepreg C/PPS plates were hot-pressed with a stacking sequence $[(0,90)]_7$. Dog-bone shape specimens were cut out from 600x600 mm plates by waterjet in order to obtain specimens with a $[(+45, -45)]_7$ sequence. Such a sequence is chosen to study the viscous behavior of angle-ply laminates at temperature higher than Tg, in order to understand their contribution in a QI laminates for other types of loading.

2.2 Experimental procedure

All the tests were performed using a 100 kN capacity load cell on a MTS 810 servo-hydraulic testing machine with a temperature control system (oven and temperature controller). Creep tests were performed at 120°C at room moisture, during 24 hours of loading followed by 48 hours of recovery. All tests were performed under the damage threshold in order to obtain a purely viscoelastic-viscoplastic behavior. This threshold was determined by a load-unload test at 120°C on the same sequence. Loop after loop, the maximum loading level was increased by 5% of the ultimate tension stress (σ_u=160 MPa). Because the linear zone is extremely reduced at temperature higher than Tg, the elastic modulus was measured by several methods. A correlation parameter evaluation method was considered to be the most accurate. The damage threshold is assumed to be at 5% with $d = 1 - \frac{E}{E_0}$. For this material, it is measured at 80 MPa ($\cong$ 50% of σ_u). According to this result, 6 creep/recovery tests have been imagined to study the viscoelastic/viscoplastic behavior of angle-ply C/PPS laminates. The loading rate was set to 50 MPa/s and the unloading time at 100 seconds to avoid any compression over-shoots and buckling problems.

2.3 Results

In order to study the viscous behavior of C/PPS laminates, a $[(+45, -45)]_7$ lay-up is relevant because the laminates response is conditioned by the PPS matrix behavior. Under such conditions, it is crucial to know that at 120°C, the matrix is about 25°C over its Tg. Therefore, according to previous mechanical tests [1], C/PPS angle-ply laminates inherit of the highly ductile behavior of PPS matrix, and loose about 60% of its shear modulus (1.35 GPa) and 30% in term of ultimate stress (159 MPa). Considering this, creep tests results (Figure 1) show a highly viscous long-term behaviour for $[(+45, -45)]_7$ laminates. Total creep strain reaches about 9 % for the maximum loading step (80 MPa), with about 3.5 % from instantaneous effect and 3% from viscoplastic origin (residual strain). For each creep stress, this residual strain represents about the third of the total creep strain with a slight decrease to

20 % for the lowest loading steps. In addition to that, the 10 MPa creep test is almost purely viscoelastic, confirming the viscoplastic flow threshold around this stress level.

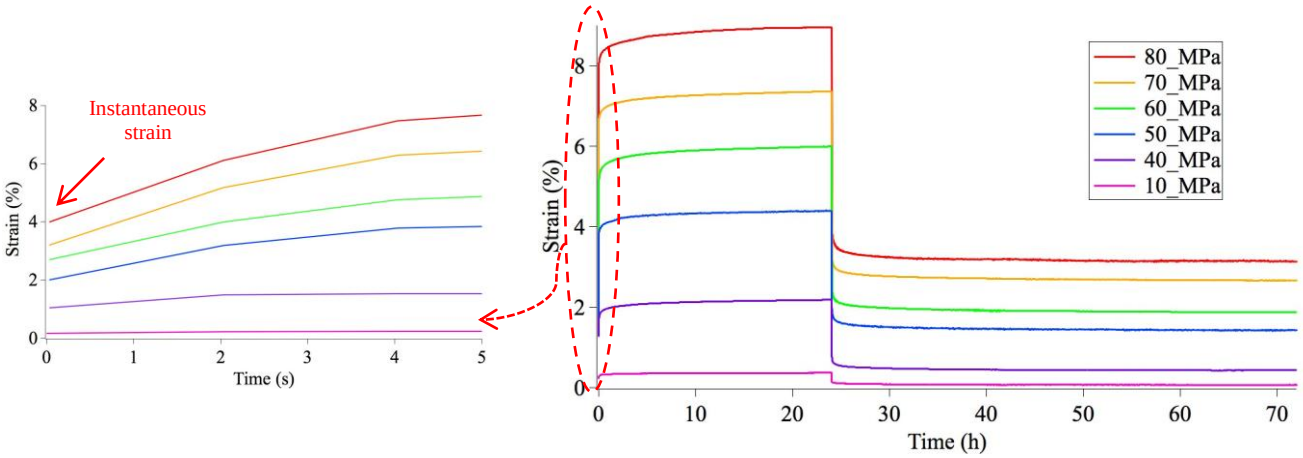

Figure 1. Creep/recovery test of six different loading levels

3 Numerical modelling

3.1 Literature review

In order to apprehend more precisely the viscous behavior of the high performance thermoplastic matrix PPS at high temperature, there is a need to develop modelling tools of this viscoelastic viscoplastic behavior. Over the past decades, several authors attempted to describe the time-dependent behavior of PMCs. For relatively small loading levels, the viscous behavior is described as linear or nonlinear viscoelastic. As explained by Findley [8], when the strain exceeds 1 or 2%, this viscoelasticity becomes nonlinear in most of the viscous materials. Beyond these loading levels, viscoelastic models loose their accuracy to predict the material viscous behavior. Indeed, when the load increases, the viscoplastic contribution become predominant. In the same manner, most of the viscoelastic viscoplastic modelling loose their accuracy in the vicinity of Tg [9].

From the simple rheological formulation to the more complex approach of Schapery's model [10], most of the viscoelastic models comes from polymers and was later adapted and extended to composite materials. One of the first formulation was developed by Findley [8], it's a simple empirical method (power law) to predict linear viscoelastic behaviour. However, this formulation shows its limits when the stress level increases. To avoid this problem, most of the authors adopt a nonlinear viscoelastic formulation, developed by Schapery [10] and based on fundamental thermodynamical laws. This formulation was widely used to predict viscous behavior of composite materials. For example, Dillard et al. [11] and Papanicolaou et al.[12] studied the viscous behaviour of C/Epoxy laminates by using this formulation. In addition to that, many authors tried to improve Schapery's model to enlarge its application field. That's why, Haj-Ali et al. [13] presented a numerical FE formulation and validated it with creep and relaxation tests on a TP matrix (PMMA) and Pasricha et al.[14] incorporated the effect of physical aging with a scaling time coefficient according to Struik's effective time theory. This model can, also be easily associated with a simple viscoplastic formulation [15] to be able to predict the behaviour of highly viscous materials. Nevertheless, most of these studies are limited to UD reinforcement and off-axis loadings. It remains also that one of the main drawbacks is the use of this formulation for multiaxial loading, which can be complex.

3.2 Numerical formulation

3.2.1 Viscoelastic formulation

Under the assumption of small strains, the total strain is usually divided in three different parts :

$$\underline{\varepsilon} = \underline{\varepsilon}^{e} + \underline{\varepsilon}^{ve} + \underline{\varepsilon}^{vp} \quad (1)$$

In order to predict the viscoelastic behavior of PMCs, a viscoelastic spectral linear was proposed for polymer materials [16], then extended and validated on PMCs [17]. The two main advantages of this formulation is the limited number of parameters (three) and an adapted expression for multiaxial loadings in comparison with the Schapery's formulation for example. All expressions are formulated in the ply orthotropy frame. This spectral model lays on a decomposition of the viscoelastic strain rate $\underline{\dot{\varepsilon}}^{ve}$ in elementary mechanisms $\underline{\xi}_i$ associated to a relaxation time spectrum :

$$\underline{\dot{\varepsilon}}^{ve} = \sum_{i=1}^{n_b} \underline{\dot{\xi}}_i \quad (2)$$

The viscoelastic formulation assumes a distribution of the respective relaxation mechanisms weights according to $\tau_i = \exp(n(i))$, relaxation time of the ith mechanism. The total number of mechanisms n_b is set at 30 to have an overall view of the mechanisms [17]. The mechanism distribution is a Gaussian (Figure 2) according to the initial formulation [17]. This distribution is characterized by two parameters : n_0 its standard deviation and n_c its average. From a physical point of view, n_0 gives an enhanced effect on late mechanisms and increase of n_c tends to homogenize the weights.

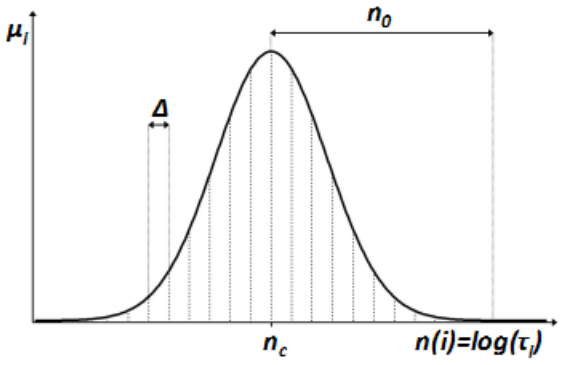

Figure 2. Gaussian spectral lay-out of the viscous relaxation mechanisms

On this diagram (Figure 2), Δ is the interval between two relaxation times. To obtain the relaxation times $\tau_i = \exp(n(i))$, $n(i)$ can be defined as :

$$\mathrm{n(i)} = n_c - n_0 + (i-1)\Delta \quad \text{with } \Delta = \frac{2n_0}{n_b - 1} \quad (3)$$

From this definition, the viscous mechanism weights can be calculated by :

$$\mu_i = \frac{1}{n_0\sqrt{\pi}} \times \exp\left(-\left(\frac{n(i)-n_c}{n_0}\right)^2\right) \quad (4)$$

Once each mechanism is identified on the spectrum, it must comply with the following differential equation deriving from a thermodynamical potential [18] :

$$\dot{\underline{\xi}}_i = \frac{1}{\tau_i}\left(\mu_i \underline{\underline{S}}^{ve}\underline{\sigma} - \underline{\xi}_i\right) \tag{5}$$

where $\underline{\sigma}$ is the Cauchy stress tensor and $\underline{\underline{S}}^{ve}$ is the viscoelastic compliances tensor associated to the viscous anisotropy tensor $\underline{\underline{A}}^{ve}$ by $\underline{\underline{S}}^{ve} = \underline{\underline{A}}^{ve-1}$, for an unidirectional ply :

$$\underline{\underline{S}}^{ve} = \begin{vmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \beta_{22}/E_2 & (-\beta_{23}.\nu_{23})/E_2 & 0 & 0 & 0 \\ 0 & (-\beta_{23}.\nu_{23})/E_2 & \beta_{22}/E_2 & 0 & 0 & 0 \\ 0 & 0 & 0 & \beta_{44}/G_{12} & 0 & 0 \\ 0 & 0 & 0 & 0 & \beta_{44}/G_{12} & 0 \\ 0 & 0 & 0 & 0 & 0 & \beta_{66}/G_{23} \end{vmatrix}$$

with $\beta_{44}, \beta_{22}, \beta_{23}, \beta_{66} = 2\beta_{22}G_{23}/E_2$ material viscosity parameter in the different direction and (G_{12}, G_{23}) the shear moduli. According to the hypothesis of an elastic behavior in the fiber directions (i.e. direction 1 and 2 for a woven composite) and a plane stress state (case of a thin laminated plate), $\beta_{22} = \beta_{23} = \beta_{66} = 0$.
The three parameters n_0, n_c, β_{44} (Table 1) are obtained from a purely viscoelastic creep test (Figure 3.a).

3.2.2 Viscoplastic formulation
In order to predict the viscous behavior of C/PPS material, the viscoelastic spectral formulation is completed witch a viscoplastic model. A generalized Norton-type model has been adopted. The viscoplastic behavior is activated only when the stress exceeds the yield stress (temperature dependent), which is the same for plasticity and viscoplasticity in PMCs. In addition, the elastic and viscoplastic behaviors are supposed to be independent for the considered loading rates, and the restoring phenomena is negligible.
The elastic domain is defined by the following viscoplastic yield function :

$$f_{vp}(\underline{\sigma}, \underline{X}) = \overline{(\underline{\sigma} - \underline{X})} - \tau_y(T) \text{ with } \overline{(\underline{\sigma} - \underline{X})} = \sqrt{{}^{T}(\underline{\sigma} - \underline{X})\underline{\underline{M}}(\underline{\sigma} - \underline{X})} \tag{6}$$

with a contracted product on two index $(\underline{\sigma} - \underline{X}) : \underline{\underline{M}} : (\underline{\sigma} - \underline{X}) = (\sigma_j - X_j)M_{ij}(\sigma_i - X_i) = {}^{T}(\underline{\sigma} - \underline{X})\underline{\underline{M}}(\underline{\sigma} - \underline{X})$. In the case of a linear kinematics hardening, thermodynamic force $\underline{X}$ associated with $\underline{\alpha}$ is defined by $\underline{X} = \delta\underline{\alpha}$. $\underline{\underline{M}}$ is a fourth order tensor describing the anisotropy of viscoplastic flow in shear loading associated with the PPS matrix [18] :

$$\underline{\underline{M}} = \begin{vmatrix} 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{vmatrix}$$

The evolution laws derived from a thermodynamical potential can be written for $\dot{\underline{\varepsilon}}^{vp}$ the viscoplastic strain rate and $\dot{\underline{\alpha}}$ the kinematic hardening rate.

$$\dot{\underline{\varepsilon}}^{vp} = K\langle f_{vp}\rangle^N \frac{\partial f_{vp}}{\partial \underline{\sigma}} \text{ and } \dot{\underline{\alpha}} = -K\langle f_{vp}\rangle^N \frac{\partial f_{vp}}{\partial \underline{X}} \tag{7}$$

with N a parameter representing the material rate sensitivity and K a parameter seen as a penalty coefficient. When K → ∞, the classical time-independent plasticity expression is recovered. With regard to the viscoplastic yield function $f_{vp}(\underline{\sigma}, \underline{X})$, those laws can be written as :

$$\underline{\dot{\varepsilon}}^{vp} = \dot{\lambda}_{vp} \frac{\underline{\underline{M}}(\underline{\sigma}-\underline{X})}{(\underline{\sigma}-\underline{X})} \text{ and } \underline{\dot{\alpha}} = \dot{\lambda}_{vp} \frac{\underline{\underline{M}}(\underline{\sigma}-\underline{X})}{(\underline{\sigma}-\underline{X})} = \underline{\dot{\varepsilon}}^{vp} \tag{8}$$

where $\dot{\lambda}_{vp} = \sqrt{{}^{T}\underline{\dot{\varepsilon}}^{vp}\underline{\underline{M}}^{-1}\underline{\dot{\varepsilon}}^{vp}}$ is the viscoplastic multiplier homogenous to a strain rate.

In order to generalize the classical approach of time-independent plasticity, the viscoplastic "dynamic" yield function can be introduced :

$$f_{vp}^{dyn}(\underline{\sigma}, \underline{X}, \dot{\lambda}_{vp}) = f_{vp}(\underline{\sigma}, \underline{X}) - \left(\dot{\lambda}_{vp}/K\right)^{\frac{1}{N}} \tag{9}$$

The two viscoplastic parameters K and N can be identified from monotonic tensile tests (Figure 3.b) at different strain rate - 50, 5 and 0.5 mm/min (Table 1).

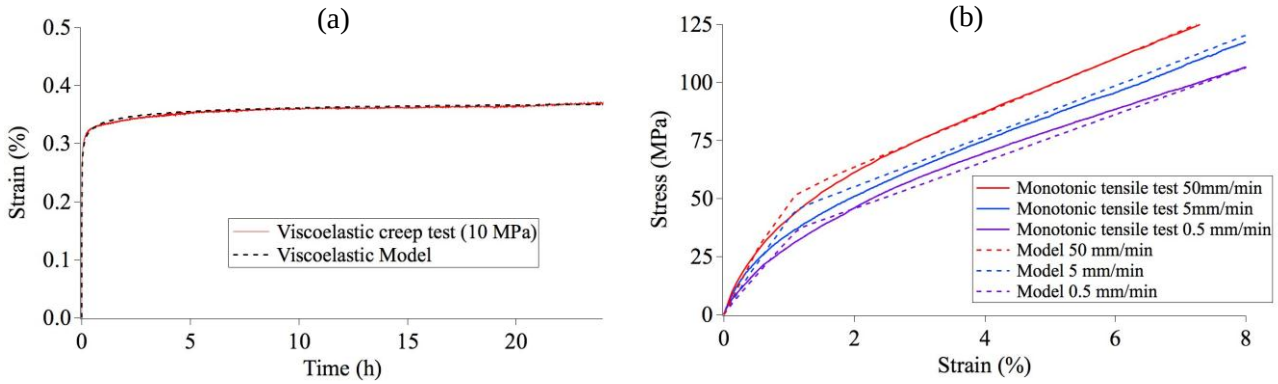

Figure 3. Viscoelastic parameters identification on a purely viscoelastic creep test (a) and viscoplastic parameters identification on tensile tests at different strain rates (b)

E_1 (Gpa)	E_2 (Gpa)	$G_{12}(T)$ (Gpa)	ν_{12}	$\tau_y(T)$ (Mpa)	n_0	n_c	β_{44}	δ (MPa)	K	N
56.5	56.5	1,35	0.04	10	4.05	6.9	0.6	400	$8.4e^{-12}$	9,5

Table 1. Mechanical properties of the ply and identified parameters

3.2.3 Time Discretization

The previous constitutive laws have been time-discretized (backward Euler) and implemented into a FE code (Cast3m). The stability analysis of this method was done by Boubakar et al. [18]. For an incremental method associated with a Newton iterative scheme, material state has to be calculated on a time interval $[t_n, t_{n+1}]$ from a strain increment $\Delta\underline{\varepsilon}$, knowing the previous converging state : $\left({}_n\underline{\sigma}, {}_n\underline{\xi}_i, {}_n\underline{X}\right) + \Delta\underline{\varepsilon} \rightarrow ({}_{n+1}\underline{\sigma}, {}_{n+1}\underline{\xi}_i, {}_{n+1}\underline{X})$

with ${}_n(.)$ and ${}_{n+1}(.)$ respectively the previous convergent quantities and the current ones

At the end of each increment, the state of the material is calculated by a classical return-map algorithm. It consists in predicting an elastic state, which can be followed by an inelastic correction to comply with the yield function. ${}_{n+1}\dot{\lambda}_{vp}$ at each increment is calculated by forcing $f_{vp}^{d}({}_{n+1}\underline{\sigma}, {}_{n+1}\underline{X}, {}_{n+1}\dot{\lambda}_{vp}) = 0$.

3.3 Validation

After the identification of the parameters (Table 1), the model has to be tested on several validation tests. The first validation test is a multi-steps creep tests at 120°C. The specimen was loaded during five steps of 24 hours with stress levels from 40 to 80 MPa, followed by a recovery. This test validates the modeling for a series of constant loading steps (Figure 4.a). The numerical modeling permits to predict adequately the material's behavior for such a loading. It can be noticed that a multi-step test involves different viscous mechanisms in comparison with the single-steps creep tests (Figure 1). Indeed, maximum creep strain reached at 80 MPa for a single-step test is about 3% under the maximum for a multi-step test. Moreover, the creep kinematic at each step seems to increase with the number of steps and the loading levels.

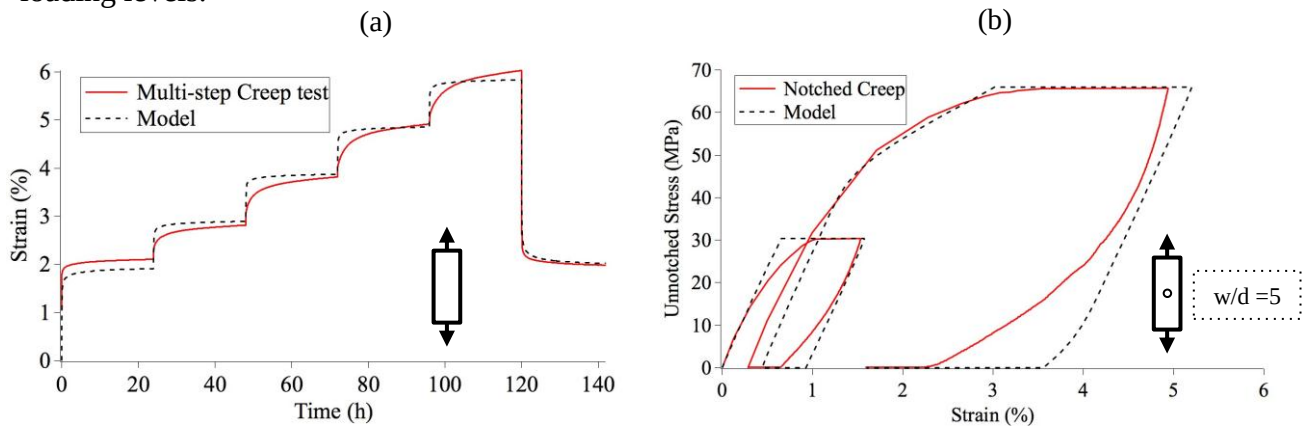

Figure 4. Validation of the model for five-steps creep test (a) and notched creep test (b)

Furthermore, the numerical model allows us to simulate the behavior for structural loadings. The response of a notched $[(+45, -45)]_7$ C/PPS subjected to a creep-recovery test at 120°C has been simulated. Two successive creep-recovery tests were performed at unnotched stresses equal to 30 and 66 MPa, respectively corresponding to notched stresses equal to 40 and 80 MPa (effective section) during 24 hours. The results (Figure 4.b) are presented in a stress-strain diagram in order to show the ability of the model to predict the loading and unloading phases. This result shows that the model gives a pretty close response, slightly overestimated in the second unloading phase. We can explain the difference by the inhomogenous stress state in the hole area, which creates prematurate damage. Nevertheless, this result on a structural test is quite satisfying for the viscoelastic viscoplastic formulation.

Conclusion

This work was aimed at investigating the long-term behavior of carbon woven-ply PPS laminates. A $[(+45, -45)]_7$ lay-up was investigated in order to understand the role of 45 ° plies in Quasi-isotropic sequences. The originality of this work consisted in studying the behavior of woven-ply TP-based laminates at temperature higher than Tg where the viscous behavior is predominant. Multiple creep tests showed that over 50 % of the total strain has its origin in time-dependent effects : viscoelasticity and viscoplasticity. To investigate more precisely this viscous behavior of PPS matrix at high temperature, a numerical modeling consisting of an association of a viscoelastic spectral and a generalized Norton-type viscoplastic formulation has been adopted. The resulting simulations predictions were compared to the experimental results in order to examine the ability of the model to predict the response of CPPS laminates to complex loadings at a temperature higher than Tg. From this comparison, it appears that the numerical model gives a good representation of the viscous behavior of the C/PPS cross-ply

laminate, even for a structural test. In a future work, the long-term behavior investigation on woven-ply PPS laminates will be compared to the one of C/Epoxy laminates. A formulation taking damage into account will be adopted to make the numerical model able to predict the long-term behavior beyond the damage threshold.

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