CHARACTERIZATION AND MODELING OF THE TIME-DEPENDENT BEHAVIOR OF THE RTM6 STRUCTURAL EPOXY INVOLVING RECOVERY, CREEP AND BACK STRESS

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Abstract

The time-dependent behavior of RTM6 structural epoxy is addressed with an emphasis on back stress upon unloading and on the strain recovery observed at zero stress after deformation. For this purpose, creep (at high stress) and recovery tests after different levels of unloading were imposed on compression cylinders. These tests have highlighted the presence of two competing deformation mechanisms, giving rise sometimes to a rate-reversal creep behavior. A first modeling approach has been developed based on the work of Khan and al. [1]. Furthermore, this behavior is assumed to originate from a deformation mechanism that is locally strongly heterogeneous. This statement is further validated experimentally through Atomic Force Microscope (AFM) testing to identify the change in local properties in deformed and undeformed, fully or partly cured, specimens.

1. Introduction

In the aeronautic industry, the use of polymer matrix high performance composites to reduce weight has been undeniably increasing over the last decades. However, if the use of such materials has reached an important part of the structure of current aircrafts, it goes with the production of a large versatility of shape, size and microstructure of composites; thus, strongly pushing for the development of predictive modeling capabilities based on the constituents properties, involving the plasticity and damage evolution of the matrix.

In earlier studies [2, 3], the mechanical behaviour of the industrial grade epoxy resin HexFlow RTM6 was studied under essentially radial, monotoneous loading paths leading to the identification of a phenomenological constitutive model including a strain rate and temperature dependent hardening law and a pressure dependent yield locus as well as a pressure-dependent principal stress based failure criterion. Moreover, it has been shown by Melchior and al. [4] that the use of a more accurate description of the non-linear large strain behavior of the matrix in the composite's RVE, leads to a better prediction of the behavior of a biaxial woven composite under 45° off-axis tensile loading.

The objective of the present study is to push even forward the understanding and modeling of the viscoplastic behavior of RTM6, focusing on the time dependent behavior under creep and recovery tests during unloading. Indeed, the rate dependence of the unloading behaviour and recovery after unloading of RTM6 shows a significant *kinematic hardening* upon unloading, as well as the competition between two deformations mechanisms : (forward) *creep* at high stress; and (backward) *recovery* at zero stress after deformation. These phenomena constitute indirect footprints of the underlying viscoplastic mechanism that control the irreversible deformation under static or cyclic loadings. Here, an attempt is made to relate the kinematic hardening and the kinetics of the creep/recovery during unloading with the heterogeneity of stress and relaxation times in the epoxy network.

2. Materials and methods

2.1. Material and processing

The material under consideration is the mono-component HexFlow RTM6 epoxy resin, certified for aeronautics where it is usually used as a matrix for CFRP composites. It is an amine-cured epoxy resin supplied by Hexcel and consists of a premixed system composed of tetraglycidylmethylenedianiline (TGMDA) and of two diamine curing agents M-DEA and M-DIPA. After curing, the RTM6 epoxy resin has a high glass transition $T_g = 220\,[°\mathrm{C}]$ determined by differential scanning calorimetry (DSC).

Two specific curing cycles were developed in order to produce two batches of specimens with different cross-linking densities : one high ($\geq$ 95% degree of conversion), and one medium ($\approx$ 80% degree of conversion). The purpose of producing a second batch with partly cured conditions (and thus non-fulfilling aeronautics standards) is to play on the crosslink density in order to possibly affect the heterogeneity of the deformation occurring in the epoxy at the micro- or nano-scales.

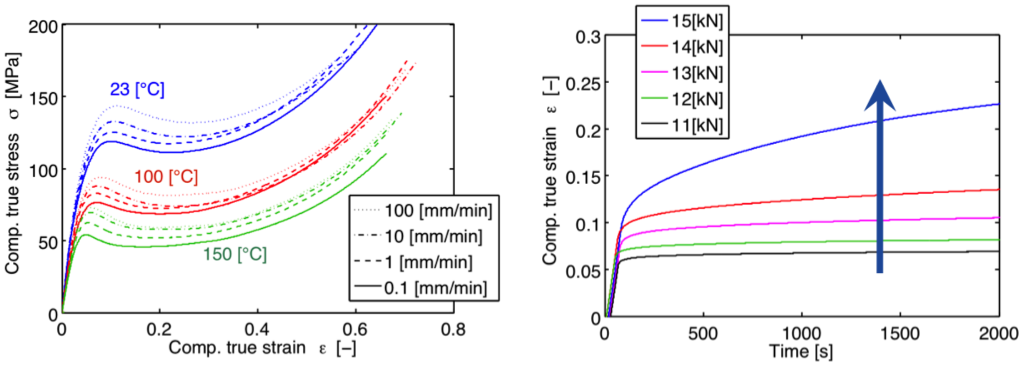
2.2. Testing methods

Macroscopic tests: Small cylindrical specimens with diameter and height both equal to 12[mm] were machined for uniaxial compression tests. The tests were carried out on a screw-driven Zwick-Roell universal testing machine with a 250 [kN] external load cell at room temperature and with solid lubrication. Correction for machine compliance was accounted for.

Local microscopic analysis: Atomic Force Microscope (AFM) mapping in HarmoniX mode was pursued on neat and deformed specimens, after microtomy surface preparation.

3. Results

Compression tests at constant crosshead speed and pre-yield creep tests had already shown in previous work [2] that RTM6 epoxy resin exhibits a strong elasto-viscoplastic behavior at large strains, as illustrated on Figure 1, even at room temperature.



(a) Compression tests at different rates and temperatures.

(b) Pre-yield creep compression tests.

Figure 1. Mechanical characterization of the *elasto-viscoplastic* behaviour of RTM6 epoxy resin.

Here, a focus was made on two specific features : the strong kinematic hardening upon unloading and the ability to partly recover the applied plastic strain after unloading. These two material behaviors can be observed in Figure 2 (a) and (b), respectively.
Therefore, a more thorough study of the rate (time) dependent behavior of RTM6 *after* deformation was pursued. For this purpose, post-yield creep tests were performed after deformation and at different levels of stress during unloading, as shown on Figure 3.

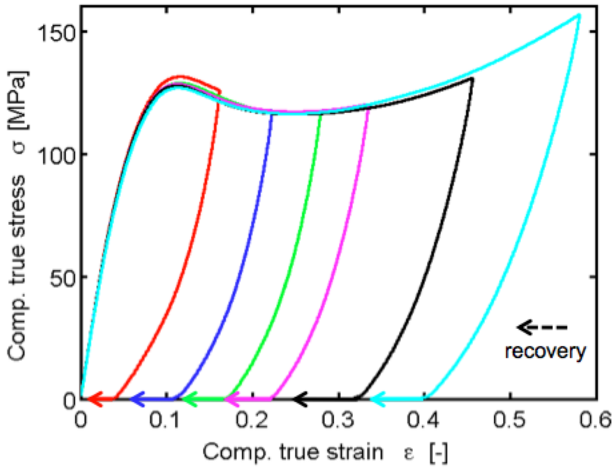
A competition between a *forward* and *backward* deformation mechanisms is observed, with the two limiting case being :

- the usual *viscoplastic flow* giving rise to creep at high stresses (already observed in earlier work);
- and the *reverse creep* at zero stress (free specimen) leading to a partial recovery of plastic deformation.

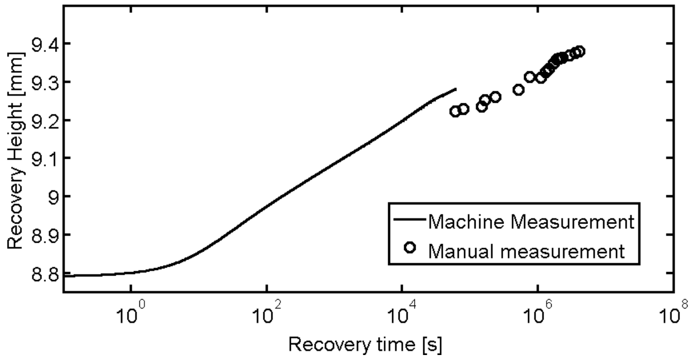
Moreover, Figure 3 shows that a phenomenon of *rate-reversal* creep at intermediate load levels can occur, meaning that the dominance zones of each mechanisms overlap. Figure 3 (b) shows indeed that for the intermediate creep loading levels of 50% and 60% small amount of recovery (backward flow) precedes forward creep showing the coexistence of two different mechanisms with two different time scales. The same phenomenon was observed for other glassy polymers (but not cross linked) by Khan and al. [1].

4. Modeling

The modeling approach proposed by Khan, will be applied to the present results. The implementation is currently being pursued and will be presented at the conference, it is based on a modified overstress (VBO) viscoplastic model with a double component configuration in order to take into account the two contributions to deformation and predict the non-monotonic creep behavior.

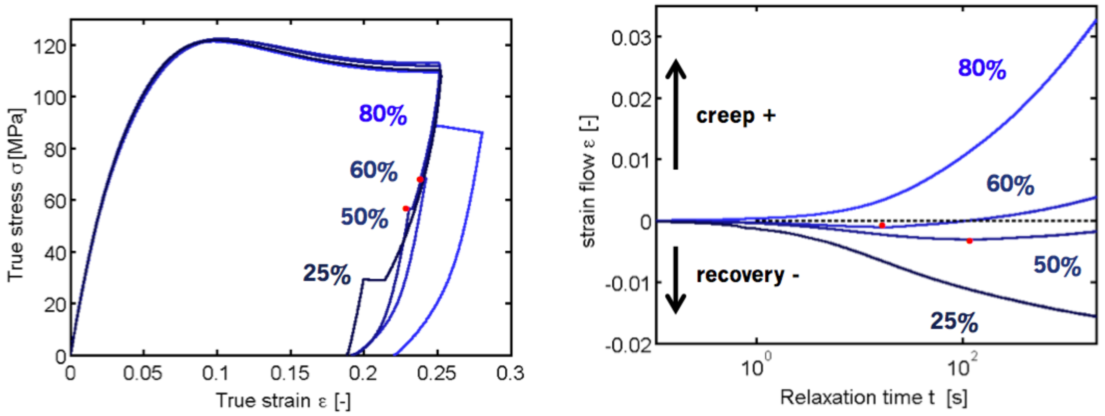


(a) Compression tests with unloading and recovery step for several applied strains.



(b) Specimen height evolution during recovery after a total applied strain of 0.45.

Figure 2. Focus on the strong kinematic hardening during unloading and subsequent recovery happening after deformation in RTM6.



(a) Compression true stress-true strain curve. (b) Creep strain increment as function of time.

Figure 3. Illustration of *rate reversal* creep during unloading (red dots indicate the reverse point).

5. Discussion and micro-analysis

The initial objective is to better understand and comprehend the presence of strong kinematic hardening upon unloading (also called *Bauschinger effect*) coupled with the presence of recovery mechanisms. The *Bauschinger effect* originates from the presence of a local microstructural heterogeneity which lead to polarized fluctuations of the internal stress field which can give rise during unloading to an earlier reverse plasticity.

Micro-shear bands have been observed by optical microscopy through polarized light (cf. Figure 4). This kind of plastic localization is expected in the case of glassy polymers exhibiting an intrinsic strain softening [5]. A nano mechanical characterization of the associated gradient of properties induced by these shear bands, was recently performed via AFM testing on neat and deformed specimens. The results of this study will be presented at the conference.

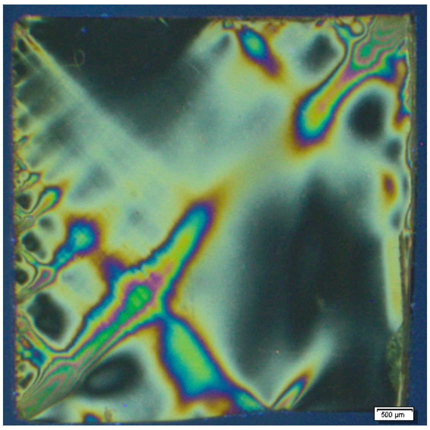


Figure 4. Observation of RTM6 microshear bands through optical microscopy with polarized light.

6. Conclusion

The present contribution has highlighted in RTM6 structural epoxy the presence of strong kinematic hardening upon unloading, coupled with important time-dependent behavior, function of the unloading stress level. Two limiting case behavior were identified with a *forward* deformation mechanism through creep at high stress; and a *backward* deformation mechanism with recovery at zero applied stress. Intermediate stress level can give rise to a complex phenomenon of rate-reversal creep. Moreover, the physical origin of the observed *Bauschinger effect* was investigated; and local micro-mechanical analysis tends to show the presence of important stress heterogeneity.

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