MODELLING OF THERMOMECHANICAL BEHAVIOUR OF A WOUND CARBON/EPOXY COMPOSITE EXPOSED TO FIRE

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Abstract

Hydrogen is expected to be highly valuable energy carrier for the 21th century as it should participate in answering main society and economical concerns. To exploit the benefits of this energy at large scale, further research and technological developments are required to secure its storage, especially during fire exposure. Thus, studies on the thermomechanical behaviour of the composite used in the manufacture of tanks for the storage of hydrogen are important. At present, the use of epoxy/carbon fibre composites is developed widely because of its low weight and its good mechanical properties. Thus, the present study focusses on the thermal decomposition property and the influence of a fire or a heating source on the residual mechanical behaviour of such materials. To account for this point, an experimental study is introduced to improve the understanding of thermal decomposition and fire exposure mechanisms of composite using different "elementary" samples.
Firstly, to characterize the mechanical properties versus fire exposure, a thermal degradation is performed using a cone calorimeter on composite samples. These tests are led for various heat flux values and are stopped at different characteristic times. Then, the mechanical properties are characterized using tensile test on samples submitted at first to different fire time exposure. The evolution of the elastic properties and ultimate stress show that the density of energy is the main factor leading to a change of the mechanical properties and char thickness evolution. Secondly, to characterize the mechanical properties versus temperature, tensile tests are performed on samples submitted in situ to 4 homogeneous temperature conditions up to 150°C. Finally, A thermomechanical model is proposed to predict the behaviour of composite material.

1 Introduction

Carbon fibres possess very high specific mechanical properties, particularly, stiffness and strength, which make them attractive as reinforcing components in the composite materials. To take full advantage of these properties, it is necessary to combine them with a matrix material, such as polymer, that ensures the cohesion of the material, protects the fibres and transfers stress effectively to them [1] [2]. The matrix also stabilizes the fibre in compression, contributes to the resistance to damage due to impact by exhibiting plastic deformation, and provides out of- plane properties to the laminates [2-3]. Epoxy is the preferred choice as the matrix for CFs due to their good impregnation and adhesion to fibre reinforcement [4]. Carbon fibre-reinforced epoxy (EP/CF) composites have been widely used in many areas, including aerospace, automobile, marine, military, etc., due to their outstanding properties, such as high strength, high modulus and light weight [2][5-9]. It results in excellent mechanical performances, chemical and electrical resistance and low shrinkage during cure [4][10]. This type of composites can be used in winding processes to manufacture hydrogen storage cylinders. Type IV pressure vessels have demonstrated promising results: these cylinders are made of a polymeric liner (for the tightness), metallic bosses (for the connection to fuel cells, for example), and a filament wound composite shell which ensures the mechanical strength [11]. To exploit the benefits of hydrogen at large scale, further researches and technological developments are required to secure the storage. For example, Gentilleau et al. [11] studied the influence of temperature on storage tanks. Berro Ramirez et al. [12] developed a damage model to accurately simulate burst modes and pressure. Wakayama et al. [13] dealt with impact on filament wound tanks…

This present work is dealing with the thermo-mechanical properties of epoxy/carbon fibre composites. The influence of a fire or a heating source on the residual tensile mechanical behaviour is studied. The thermal aggressions are performed by using a cone calorimeter apparatus (ISO 5660), at homogeneous heat fluxes, from 20 to 60kW/m^2. During those tests, the fire exposure is stopped at different times to study the influence of the thermal energy (different heat fluxes and exposure durations) on the residual mechanical properties.

In this work, two principal issues are examined. Firstly, the thermal impact by cone calorimeter on the composite samples will be studied. The relationship between the residue thickness and energy exposure (and duration of inflammation) will be also analysed. Secondly, the influence of the thermal impact on the mechanical residual strength of composite will be presented. This work will be finished by a thermomechanical behaviour model to predict the response of composite material samples under different homogeneous temperatures.

2 Fire exposure of composite samples

The composite material is composed of T700S carbon fibres in an epoxy matrix. To study the properties of samples representative of the hydrogen storage cylinders, parallelepipedic specimens of dimensions 300×25×5mm^3 are cut from wound cylinders. Many authors show the influence of the fibre orientation on the mechanical strength [14-17]. In this work, different fibre orientations (with respect to the cylinder axis) have been studied: 90°, ± 45° and a quasi-isotropic stacking sequence (±12°/90° /±45°/90°) noted EC90, EC45 and ECiso respectively.

The evolution of the residue thickness and the residual mechanical properties vs. the thermal energy for the three types of samples is compared and detailed in [22]. The thermal aggression is performed by using a cone calorimeter apparatus (ISO 5660). The specimens have been put in a specimen holder in aluminium with a top plate in steel (only an area about 100x25mm^2 in the middle of the specimen is exposed to the cone calorimeter) and an insulating layer under the sample in refractory material (Figure 1). Then, they have been exposed to a radiant cone under a constant heat flux, possibly ranging from 20 to 60kW/m^2. During these tests, the fire exposure is stopped at different times depending on the type of the sample. For each condition (fibre orientation, flux value, exposure duration), three samples have been tested for repeatability.

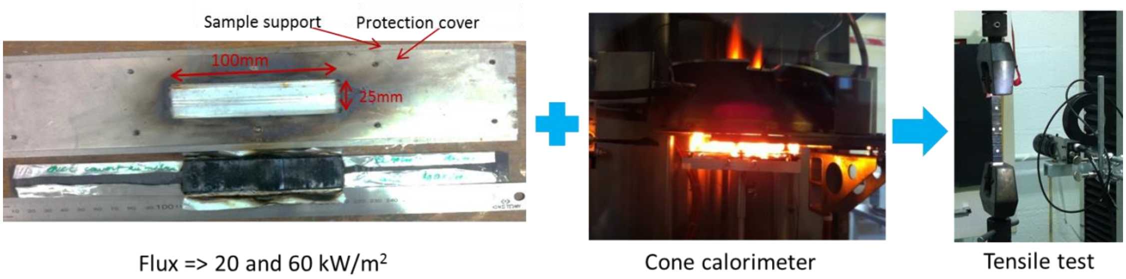

Figure 1 : Cone calorimeter and tensile machine instrumented with optical tracking

After fire exposure, if the thermal energy brought by the cone calorimeter is sufficient, a char (or residue) layer appears on the exposed sample surface.

3 Thermomechanical behaviour under homogeneous temperature

In this section we present the results of simple tensile tests on specimens at different temperatures

For all samples, the homogeneous temperature clearly affects the tensile strength as well as the stiffness: the higher the temperature, the lower the strength and the stiffness.

Figure 2-(A) compares the evolution of the stress vs. of the strain for different temperatures for the EC90 samples. The effect of temperature is significant on the material response. Between 20 °C and 80 °C, the drop of stiffness remains slight. But as soon as the glass transition temperature (108 °C) is reached and exceeded, the stiffness drops considerably. The failure mode of the specimen is a brutal

fibre / matrix debonding.

Figure 2-(B) shows a tensile test stress - strain curve for the samples EC45 and for each temperature, pointing out the influence of temperature levels on the behaviour. Compared to the evolution of stress versus strain for sample 90°, we observe a strong nonlinear evolution, which can be explained by an irreversible sliding of the plies. Note that the initial stiffness is hardly influenced by temperature in the range 20°C - 80°C. At 150°C, the curve exhibits a strong viscoelastic behaviour.

Figure 2-(C) shows the behaviour of ECiso samples versus temperature level. The rigidity decreases when the temperature level increases. The existence of a more or less long plateau after the first delamination is related to the size of the defects at the microstructure level. The curve at 150°C differs from the others: because of the viscosity of the matrix, no brutal delamination is observed but a progressive ply sliding.

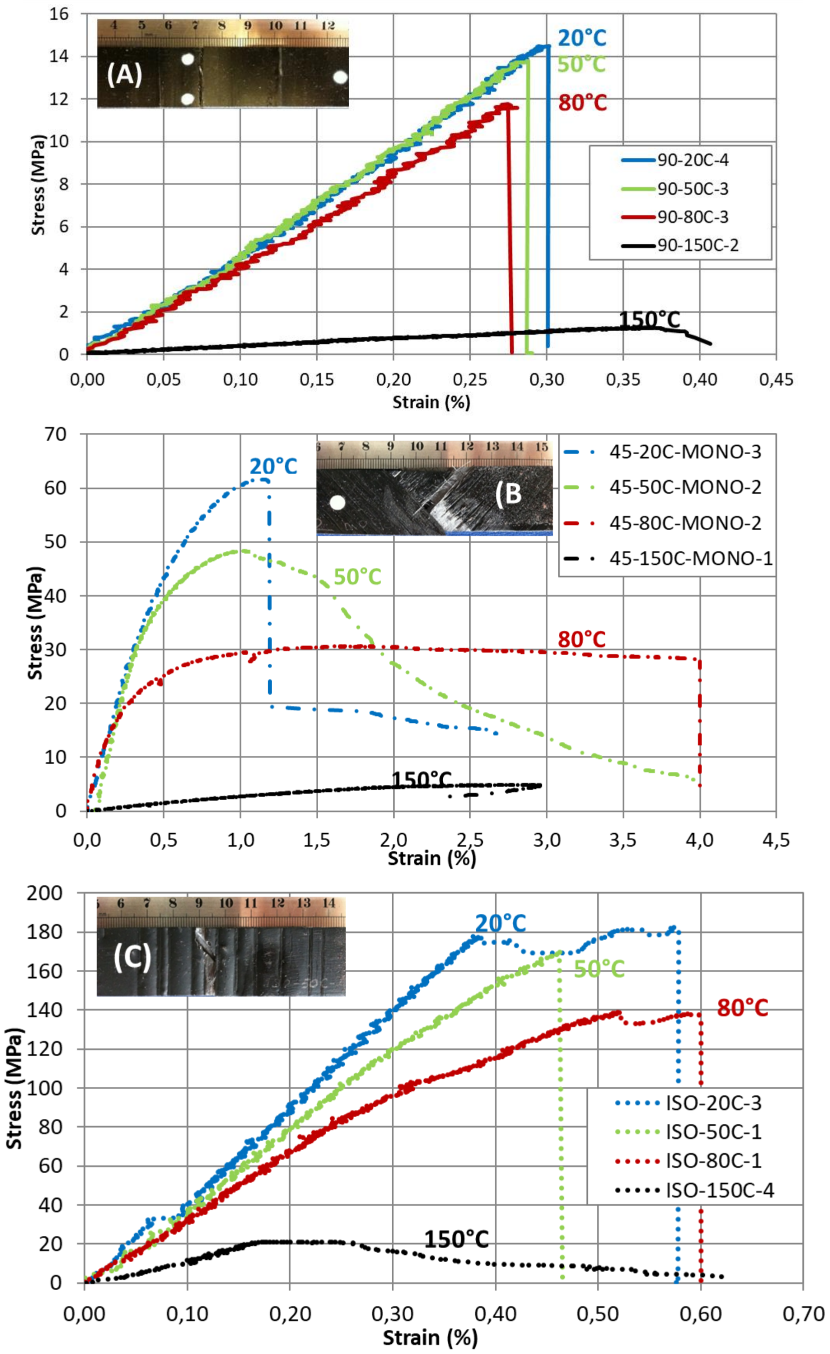

Figure 2 : Tensile stress-strain curves of samples (EC90 (A), EC45 (B) and ECiso (C)) submitted to homogeneous temperatures (20°C, 50°C, 80°C & 150°C) and mechanical loading until rupture

4 Thermo-mechanical model

To model the thermo-mechanical behaviour of wound composite material, a model based on the Hashin criteria is proposed. Hashin established the need for failure criteria that are based on failure mechanisms [19]. Hashin criteria deal with both tension and compression criteria. However, the tests described in the previous section only activate failure under positive stress. Hence, only this mode will be considered in the following. The criteria initially proposed by Hashin only consider in-plane failure mechanisms. To simulate all observed damage modes and particularly delamination, two extra criteria have been added. The first one is an out-of-plane generalization and involves the tensile stress normal to the ply and the out-of-plane shear stress components.

Fibre rupture

$$F_f = \left(\frac{\sigma_{11}}{X^T}\right)^2 + \left(\frac{\sigma_{12}}{S^L}\right)^2 = 1 \tag{1}$$

Matrix rupture

$$F_m = \left(\frac{\sigma_{22}}{Y^T}\right)^2 + \left(\frac{\sigma_{12}}{S^L}\right)^2 = 1 \tag{2}$$

Delamination failure [19][20]

$$F_d = \left(\frac{\sigma_{33}}{Z}\right)^2 + \left(\frac{\sigma_{13}}{S^H}\right)^2 + \left(\frac{\sigma_{23}}{S^H}\right)^2 = 1 \tag{3}$$

Where σ_{ij} are the components of stress tensor. X^T represents the fibre strength determined from tensile test in the direction of fibre. Y^T is the matrix strength identified from a tensile test on EC90. Z represents delamination out-of-plane strength. Because of the lack of experimental information about out-of-plane properties, this parameter is considered equal to in-plane strength (this assumption can be commonly found in the literature [21]) $S^H$and S^L represent the delamination failure by in-plane and out-of-plane shear. These parameters are identified from tensile tests performed on samples containing $+\theta/-\theta$ plies, for each temperature. These stress-based criteria are associated with brittle failure mode, observed in EC90 and ECiso samples.

The anisotropic Hill criterion is used to distinguish the brittle behaviour in the fibre direction and in the direction perpendicular to the fibres on the one hand, and the plastic-like behaviour exhibited by the EC45 samples:

$$f(\sigma) = \sqrt{\begin{matrix} F(\sigma_{22}-\sigma_{33})^2 + G(\sigma_{33}-\sigma_{11})^2 + H(\sigma_{11}-\sigma_{22})^2 \\ +2L\sigma_{23}^2 + 2M\sigma_{31}^2 + 2N\sigma_{12}^2 \end{matrix}} \tag{4}$$

An elastic behaviour is assigned to the directions 1, 2 and 3 by imposing $F, G, H = 0.$, whereas the plastic-like behaviour is controlled by the shear stress components. The plastic strain plays a dual role. It represents the irreversible deformation in the composite material and drives the progressive damage on shear modulus G_{12}, G_{13} and G_{23}.(Figure 3)

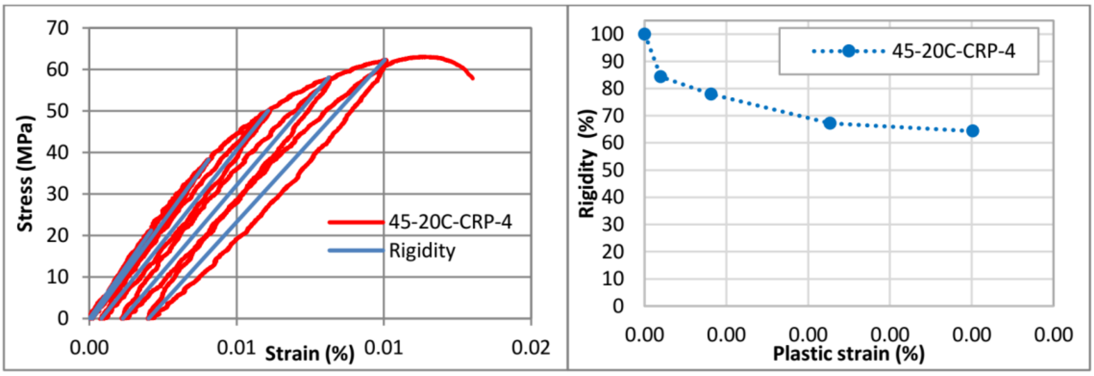

Figure 3 : Tensile test for EC45: Cyclic loading, Shear evolution vs. Plastic strain

However, the delamination criterion (3) cannot correctly predict EC45 failure, since the sample undergoes a large irreversible strain even if the maximum stress is reached. That is why an extra delamination criterion is added, based on the equivalent plastic strain ε^P.

$$\frac{\varepsilon^P}{\varepsilon^P_{max}} = 1 \tag{5}$$

Table 1 summarizes the model parameters for each temperature.

Temperature	$X_T = X_C$	$Y_T = Y_C$	Z	$S_L = S_H$	ε^P_{max}
°C	(MPa)	(MPa)	(MPa)	(MPa)	
20	2250	14,5	6,5	60	0,022
50	2250	13,8	6,2	47,3	0,03
80	2250	11,7	5,2	34,1	0,1
150	2250	1,2	0,5	3,5	0,1

Table 1 : Model parameters for each homogeneous temperature.

5 Results and discussion

As a first validation, the tensile tests on specimens EC45 are simulated in the following way. The test on parallelepipedic sample EC45 is modelled by FEA (Figure 4). The sample is partitioned into several partitions. each partition represents a fold whose fibre orientation is specified. The element type is an 8 node thermally coupled brick, trilinear displacement and temperature (C3D8T). The thermal parameters are deducted from [22]. The specific heat is 1020 J.kg^{-1}.K^{-1}, density of material is 1360 kg.m^{-3} and conductivity is 0,5 W.m^{-1}.K^{-1}. Tensile test loading is performed for each homogeneous temperature. Figure 5 shows results of the simulation of tensile tests for EC45. A fair correlation between simulation and experiment can be observed. A good prediction of the initial rigidity and the ultimate tensile strength is observed for the different temperatures. These preliminary results validate the approach.

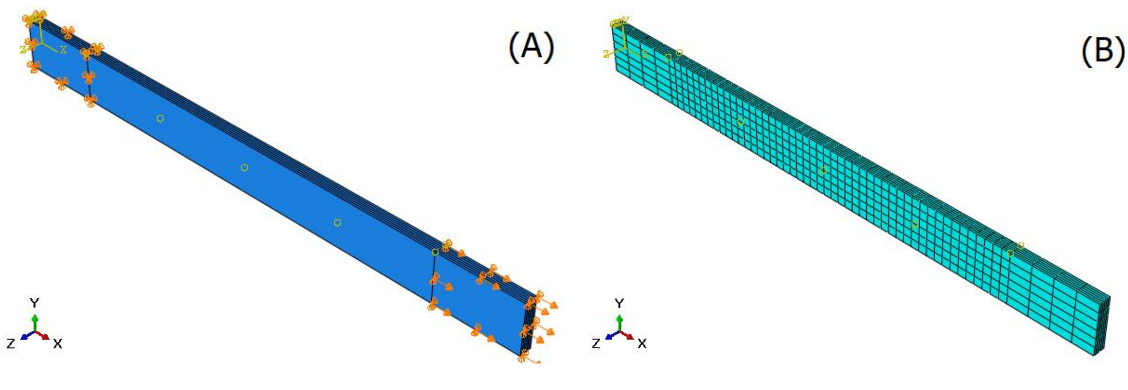

Figure 4 : Modelling of sample EC45 by FEA; (A) and Boundary condition. (B) Mesh of structure of sample EC45.

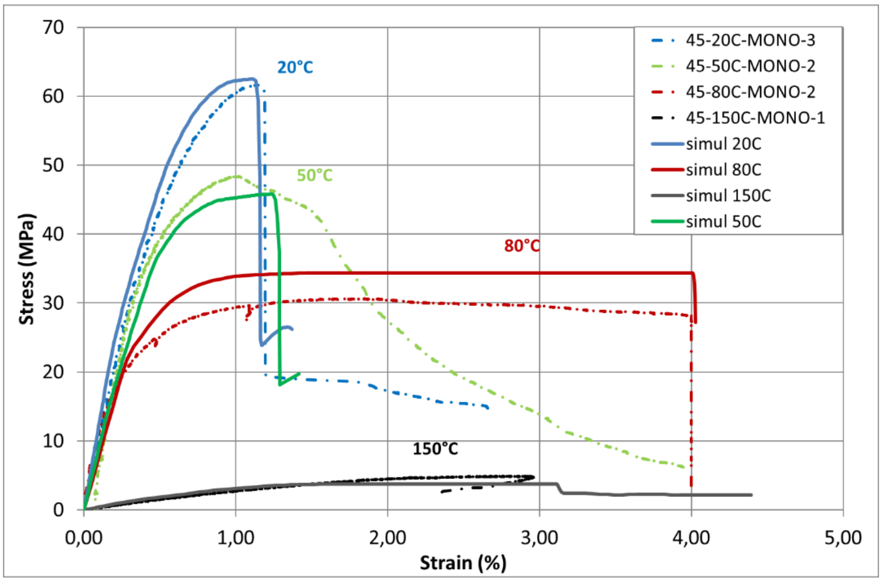

Figure 5 : Simulation (solid line) and experience (dashed line) of tensile test for EC45with time exposure (140, 180, 200, 220 (s)): Stress (MPa) vs. Strain (%)

6 Conclusion

The thermo-mechanical properties of a composite epoxy/carbon fibre have been studied. Two different types of strength reduction are observed depending on whether an ignition process occurred or not. Before ignition, the mechanical strength of exposed samples decreases very slightly and more sharply after ignition. The energy brought by the cone calorimeter is the principal parameter which leads to mechanical strength decrease.

For all samples, the homogeneous thermal exposure clearly affects the tensile strength as well as the stiffness: the higher the temperature, the lower the strength and the stiffness. Between 20 °C and 80 °C, the drop of stiffness remains slight. But as soon as the glass transition temperature (108 °C) is reached and exceeded, the stiffness drops considerably. The failure mode is depending on the sample:

– A brutal fibre/matrix debonding for EC90,
– A no brutal delamination and a progressive friction sliding for EC45
– A combination of the two modes (delamination and decohesion fibre/matrix) for ECiso.

At 150°C the viscosity of the matrix induces a nonlinear behaviour with no brutal delamination and progressive ply sliding.

To model damage of the wound composite structure two mechanisms have been validated and used in proposed model; a critical energy of initiation of decomposition to get char and the thickness of the latter to estimate the residual elastic properties.

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