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# CHARACTERIZATION AND MODELING OF COMPOSITE MATERIAL BEHAVIOUR SUBMITTED PREVIOUSLY TO FIRE EXPOSURE

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**Keywords:** Composite materials, high-pressure vessels, thermo-mechanical behaviour, modelling, delamination

## ABSTRACT

Hydrogen is expected to be highly valuable energy carrier for the 21<sup>th</sup> 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 in order to secure its storage, especially during fire exposure. Thus, studies on the mechanical and the thermal behaviours 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 degradation 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 in order to improve the understanding of thermal degradation and fire exposure mechanisms of composite using different “elementary” samples ( $\pm 12^\circ$ ,  $\pm 45^\circ$ ,  $90^\circ$  and quasi-isotropic samples).

Firstly, 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. Secondly, to characterize the mechanical properties versus fire exposure, a thermal degradation is performed using a cone calorimeter (ISO 5660) on carbon/epoxy composite samples. These tests are led for various heat flux values and are stopped at different characteristic times. The other thermal parameters under consideration are the density of energy, the presence (or not) and the duration of the inflammation. 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.

## 1 INTRODUCTION

Carbon fibres possess very high specific mechanical properties, in particular, 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 in order 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 in order to secure the storage. For example, Gentilleau *et al.* [11] studied the influence of temperature on storage. 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 work is dealing with the thermal-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, at homogeneous heat fluxes, from 20 to 60kW/m<sup>2</sup>. During those tests, the fire exposure is stopped at different times in order 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.

## 2 MATERIAL AND SAMPLES

The composite material at stake here is composed of T700S carbon fibres in an epoxy matrix. In order to study the properties of samples representative of the hydrogen storage cylinders, parallelepipedic specimens of dimensions 300×25×5mm<sup>3</sup> 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 sequence (±12°/90° /±45°/90°) noted EC90, EC45 and ECiso respectively.

## 3 FIRE EXPOSURE OF COMPOSITES SAMPLES

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 100×25mm<sup>2</sup> 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<sup>2</sup>. 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.

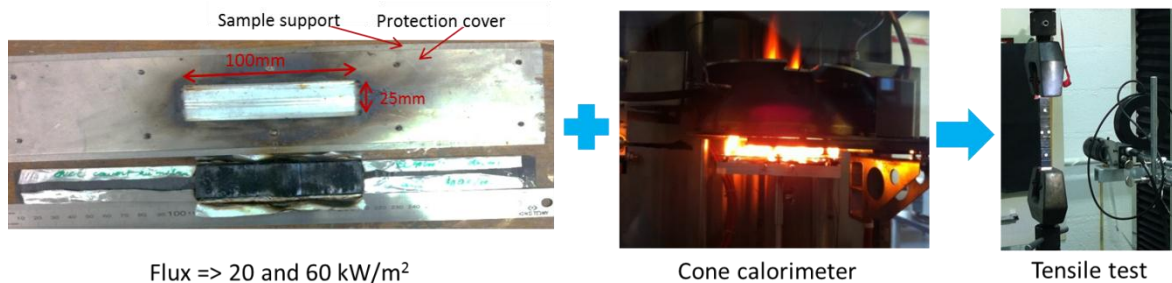


Figure 1 : Cone calorimeter and tensile machine instrumented with optical tracking

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. This char thickness is noted  $d_1$ , whereas the “virgin” composite (whose aspect is similar to that of non-exposed material) is  $d_2$ . Figure 2 shows an example of the EC90 sample after 130s exposure under a heat flux of 35kW/m<sup>2</sup>. The residue thickness is observed and measured for different exposure energy level by optical microscopy apparatus (LAICA MZ95), only for the ignited samples.

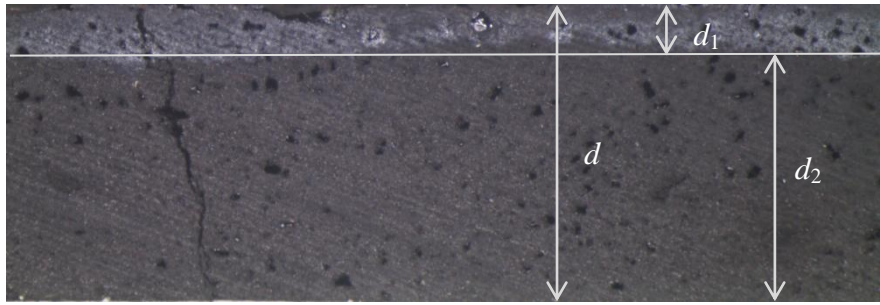


Figure 2 : EC90 sample after 130s exposure under a heat flux of  $35\text{kW/m}^2$  ( $d$ : sample thickness,  $d_1$ : char thickness,  $d_2$ : “virgin” composite thickness).

The evolution of the residue thickness vs. the thermal energy for the three types of samples is compared in Figure 3. Each test is performed at a given value of heat flux ( $35\text{kW/m}^2$ ). A general linear trend can be observed. The following conclusions can be drawn:

1. There is a threshold (in terms of energy) below which no char appears ( $3.5\text{ MJ}$  for the EC90 sample and  $5\text{ MJ}$  for the samples EC45 and ECiso).
2. This threshold difference between EC90 and EC45/iso samples is due to the heterogeneity of the resin distribution in EC90 sample (the resin density is higher near the surface) and to its degradation by oxidation even when there is no char. This influence of resin oxidation is less marked in EC45 and ECiso samples (the fibres play a more important role in these samples).
3. Once the char appeared, its propagation kinetics is almost identical for all orientations.

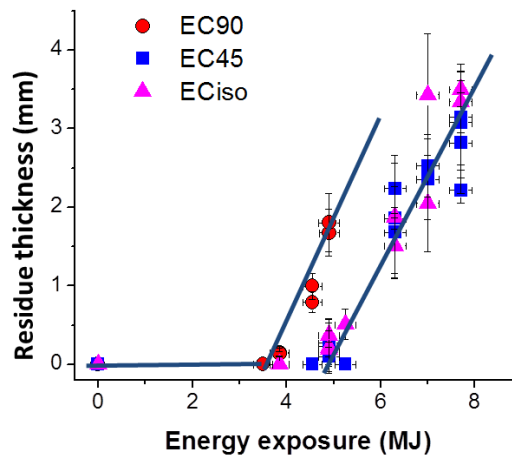


Figure 3: Residue thickness vs. thermal energy exposure for the EC90, EC45, ECiso samples (all tests are performed with the same heat flux of  $35\text{kW/m}^2$ ).

#### 4 MECHANICAL BEHAVIOUR: STRESS-STRAIN CURVE AND FRACTURE MODE

To determine the mechanical properties (in particular stiffness and strength) of the exposed samples, tensile tests up to fracture have been performed using an INSTRON 4505 machine with self-locking grips. The maximum capacity of this device is  $100\text{kN}$ . The crosshead speed is  $1\text{mm/mn}$  for all tests. Each test is instrumented with optical tracking to measure strain without contact (Fig. 1).

For all exposed samples, the exposure duration (or, equivalently, the energy input) clearly affects the tensile strength as well as the stiffness: the longer the exposure duration, the lower the strength and the stiffness. Fig. 4-(A) represents the tensile stress-strain curve for the samples EC90. The fracture mode of this sample is a complete fibre/matrix debonding as shown in Fig. 5-A. Strength undergoes a sharp drop when the sample has been subjected to the cone calorimeter, whereas the stiffness remains relatively low and less affected by the fire exposure.

Fig. 4-(B) shows the tensile stress - strain curve for the samples EC45. A strong nonlinearity is observed which is due to more progressive damage mechanisms (matrix microcracking, irreversible

sliding between plies...). This type of damage evolves until rupture by delamination due to microcracking between the plies as showed in Fig. 5-B.

Fig. 4-(C) displays the tensile stress - strain curve for the samples ECiso. Three stages can be observed: first, an elastic linear evolution, then a slightly nonlinear part probably due to microcracking and finally a complete delamination as showed in Fig. 5-C.

For all tensile tests (except EC90), the stiffness decrease appears only when inflammation occurs. The case of EC90 samples is less speaking because of the very low stiffness of the resin.

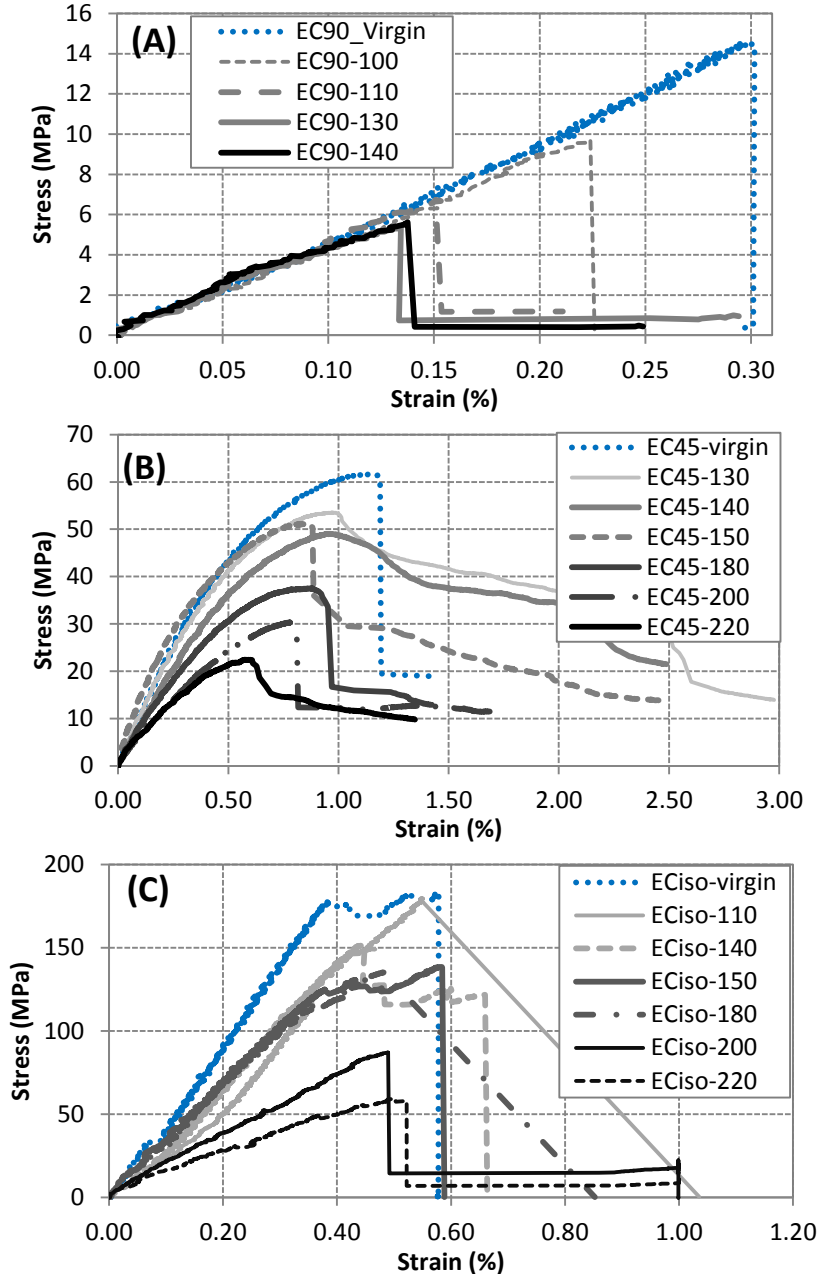


Figure 4 : Tensile stress-strain curves of samples (EC90 (A), EC45 (B) and ECiso (C)) first exposed to a given heat flux of  $35\text{kW/m}^2$ . The numbers (100, 110,...) represent the fire exposure duration in seconds and “virgin” the non-exposed sample.

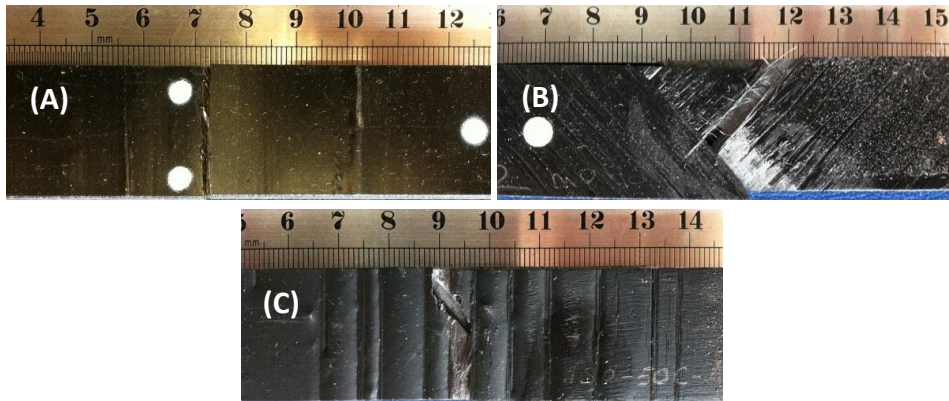


Figure 5 : Damage modes: fibre-matrix debonding for EC90 (A), sliding between plies for EC45 (B) and delamination for ECiso (C).

To compare more easily the residual mechanical properties for the different types of samples, Figure 6 plots the normalized mechanical strength, defined as the ratio ‘maximum stress of the exposed sample / maximum stress of the virgin sample’ as a function of the incident energy. Three conclusions can be drawn:

1. The thermal energy is the main driving force of the degradation for all exposed sample.
2. For all samples (EC90, EC45, ECiso) two slopes are observed; the first one before inflammation and the second one, more significant, after ignition. This shows the effect of inflammation on the evolution of the maximum stress.
3. Two sets of curves are observed: whereas the EC45 and ECiso curves are similar (very little effect of thermal energy before inflammation), the EC90 curve highlights the rapid degradation of the resin even before inflammation.

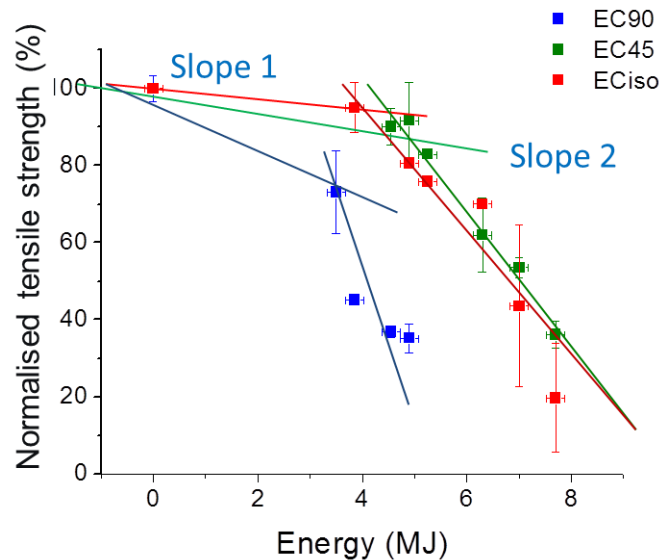


Figure 6 : Normalised tensile stress (%) vs. energy exposure (incident flux of 35kW/m<sup>2</sup>) for the EC90, EC45, and ECiso; slope 1 before inflammation, Slope2 after inflammation

## 5 THERMOMECHANICAL MODEL

In order 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 take into account in-plane failure mechanisms. In order to simulate all observed damage modes and in particular

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

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

Matrix rupture

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

Delamination failure [19][20]

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

where  $\sigma_{ij}$  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. 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:

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

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 also drives the progressive damage on shear modulus  $G_{12}$ ,  $G_{13}$  and  $G_{23}$  (Fig. 7). 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  $\varepsilon^P$ .

$$\frac{\varepsilon^P}{\varepsilon_{max}^P} = 1 \quad (5)$$

Table 1 summarizes the model parameter.

$X_T = X_C$	$Y_T = Y_C$	$Z$	$S_L = S_H$	$\varepsilon_{max}^P$
(MPa)	(MPa)	(MPa)	(MPa)	
2250	14.5	6.5	60	0.022

Table 1 : Parameter values for a temperature of 20 °C.

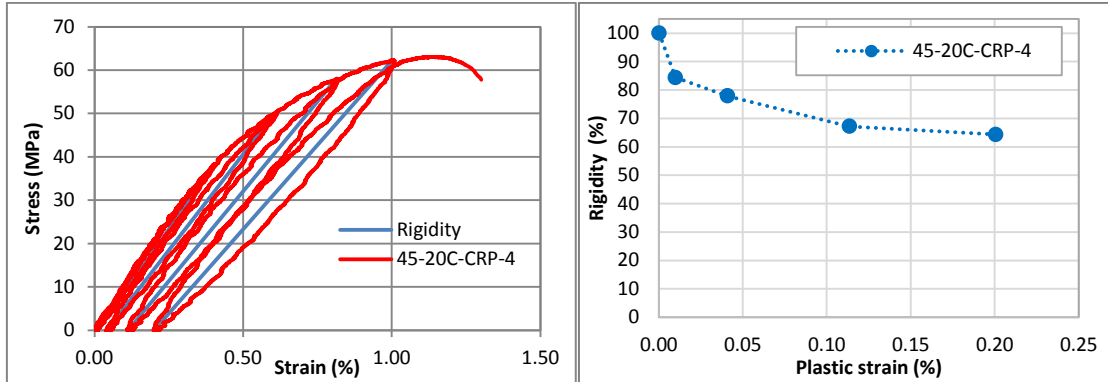


Figure 7: Tensile test for EC45: Cyclic loading, Shear evolution vs. Plastic strain

## 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 8). For given exposure conditions (incident flux, exposure duration), Figure 3 provides the char thickness. A partition of the sample is performed: the mechanical properties of char are assumed to be very low, whereas the stiffness and the strength of the rest of the sample remain equal to their initial values (Figure 8). This hypothesis is justified by the fact that resin decomposition also affects load transfer to the fibres. Therefore the mechanical load is only carried by the fibres and the resin not affected by thermal degradation. Figure 9 shows results of the simulation of tensile tests for EC45. A fair correlation between simulation and experiment can be observed. These preliminary results validate the approach.

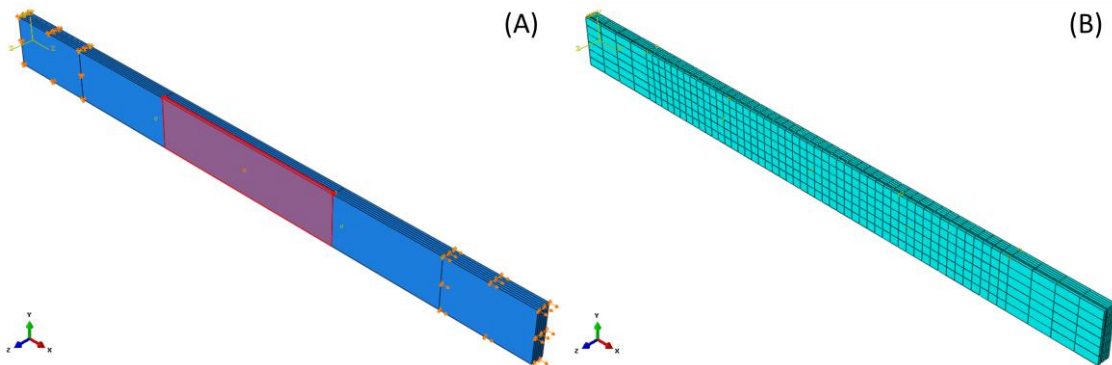


Figure 8 : Modelling of sample EC45 by FEA; (A) Partition composite (blue) and char (red) and Boundary condition. (B) Mesh of structure of sample EC45.

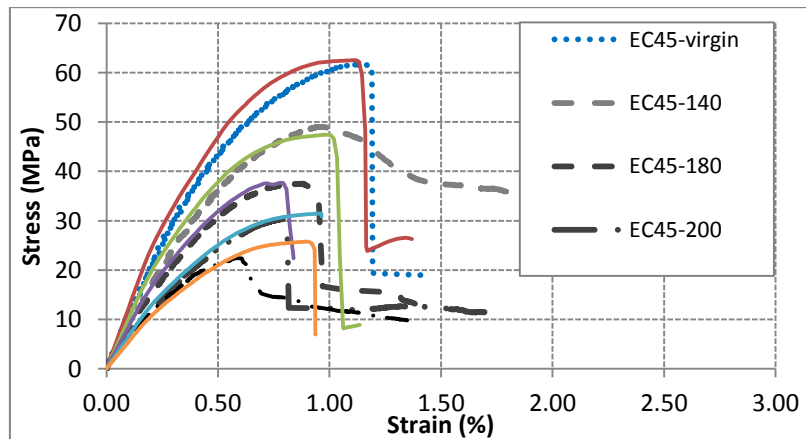


Figure 9 : Simulation (solid line) and experience (dashed line) of tensile test for EC45 with time exposure (140, 180, 200, 220 (s)): Stress (MPa) vs. Strain (%)



Figure 10 shows the normalised tensile stress versus exposure energy. The simulation response is in accordance with the experimental curve. This confirms, for EC45 samples, that the thickness of char is a key parameter in modelling the decomposition kinetics of the wound composite structures.

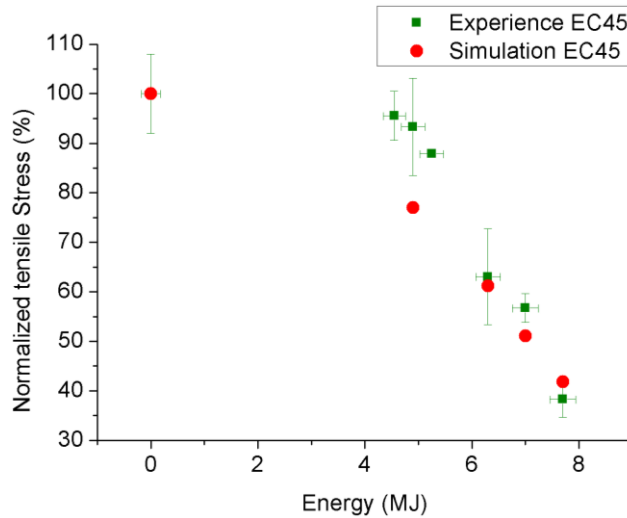


Figure 10 : Simulation and experience: Normalized tensile stress vs. energy

## 9 CONCLUSIONS

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 a given energy level, two groups are observed. A very little influence of the heat flux on the mechanical strength is observed as long as ignition does not occur, whereas mechanical strength drops after inflammation.

For the EC90 samples, the distribution of the resin and its oxidation has been found to have an influence on the degradation kinetics.

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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