EFFECTS OF MANUFACTURING PROCESS AND WATER AGEING ON THE MECHANICAL BEHAVIOUR OF TWO REINFORCED COMPOSITES: FLAX-FIBRES AND GLASS-FIBRES

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Abstract

This paper aims at investigating on the effects of manufacturing process and water ageing on the mechanical behaviour of quasi-unidirectional Flaxï Fibre Reinforced Epoxy (FFRE) and quasi-unidirectional Glassï Fibre Reinforced Epoxy (GFRE) composites. Two processes have been used to manufacture the various samples of these materials: hand lay-up process and platen press process. Tensile specimens were subjected to water ageing at room temperature. The manufacturing process type, water uptake and their effects on tensile properties of FFRE and GFRE composites will be investigated. Analysing the different types of damage mechanism of these composites by using acoustic emission (AE) enabled us to obtain several informations about their degradation process, during tensile tests. The obtained results showed that the implementation technique that was considered significantly affects the tensile properties of both composites and thereby modifies their behaviour in a moist environment.

1. Introduction

Under growing pressure from ecological concern and environmental regulations to reduce CO2 emission, the market demands more materials derived from renewable and biodegradable resources, which are recyclable at life end. The use of natural fibres as reinforcement in composite materials can help satisfy these new requirements. The specific properties of these natural fibres were in some cases better than those of glass fibres [1]. In addition, they have more advantages due to their great performance, low cost, and low relative density and environmentally superior to glass fibres in general [2]. This suggests that natural fibres, as flax and hemp fibres, have a potential of being used in many applications, in particular, for automotive domain [3]. Unfortunately, the main drawbacks with using these green materials remain their inherent susceptibility to moisture expansion, which has the effect of inducing a decrease in mechanical properties, and the dispersion in their physical and mechanical properties [4]. The understanding of their behaviour with environmental conditions can contribute to the development of bio-composites. In this context, study of the effect of moisture absorption on the mechanical properties of these materials is very important. Indeed, several research works have been realised to investigate the ageing effect on the mechanical properties and the damage mechanisms of reinforced vegetal fibre composites [5-8]. In the study by Arbelaiz et al. [5], the influence of water uptake on the sorption characteristics of flax fibre bundle/polypropylene composites was investigated. After immersion in distilled water at room temperature, results showed that mechanical properties decreased because of the change of structure and properties of ýbres, matrix and interface fibres-matrix caused by water molecules With the aim of analysing the different types of damage mechanism of vegetal fibre composites, several authors have used the Acoustic Emission (AE) technique combined with Scanning Electron Microscopy (SEM) [7-10]. Assarar et al. [8] used Acoustic Emission to describe the damage due to the ageing by water immersion for Flaxï Fibre Reinforced Epoxy (FFRE) and Glassï Fibre Reinforced Epoxy (GFRE) composites. The collected parameters by AE were used as input descriptors in the proposed classification method. They suggested that fibre/matrix interface weakening is the main damage mechanism induced by water ageing for both composites. De Rosa et al. [9] evaluated the level of impact loading and the residual strength of jute-glass hybrid laminates by correlating the AE activity to the applied cyclic stress on impacted specimens at various energies.

The main objective of this work is to assess the effects of manufacturing process and water ageing on the mechanical behaviour of quasi-unidirectional (FFRE) and quasi-unidirectional (GFRE) composites, which are subjected to water ageing at room temperature. The manufacturing process type, water ageing and their effects on tensile properties of FFRE and GFRE composites have been investigated. Next, the amplitude distribution and the cumulative number of hits of AE events were used in order to identify the damage mechanisms according to the manufacturing process and water ageing.

2. Experimental methods

2.1 Materials and manufacturing processes

Unidirectional Flax-fibre and E-glass-fibre fabrics acquired form DEPESTELE Group were used in this study. The weights of unidirectional fabrics were 200 and 300 g/m^2 for Flax-fibre and E-glass-fibre, respectively. The weft and warp ratio was 9/91 for both fabrics. The polymer matrix used to produce the composites was an epoxy system (E_m = 3.35 GPa) consisting of an epoxy resin SR1500 and a SD2503 hardener (at a mixing ratio of 100:33 by weight). The composites materials were manufactured by two manufacturing processes at room temperature:

- Hand lay-up process: The fabrics were manually impregnated with the resin system using a roller. The mold was closed and placed inside a pocket. Then, plates were cured at room temperature with a pressure of 30 kPa using vacuum moulding process.
- Platen press process: The fabrics were impregnated with the resin system by hand using a roller and they were placed between two steel trays covered with Teflon paper. Plates were then cured in a press under pressure of 5 bars at 25°C for 12 h.

For both manufacturing processes, plates of 450 mm by 300 mm were manufactured from four ply unidirectional laminates, with a plate thickness of approximately 2 mm. The fibre volume fractions V_f for FFRE and GFRE composites are given in table 1.

Materials	**Fibre volume fraction (V_f)**	
	Hand lay-up process	***Platen press process***
FFRE	0.25	0.35
GFRE	0.50	0.54

Table 1. Fibre volume fraction V_f for FFRE and GFRE

2.2 Water ageing

After being dried in an oven at 60°C during 75h, tensile specimens were immersed in water at room temperature (20°C) for 52 days. At regular intervals of immersion test, the specimens were removed from water, wiped with a dry cloth and weighed with a balance of ±1 mg precision. The specimens were put back in water to enable the continuation of the absorption process until an equilibrium plateau was reached. The relative of water uptake (M_t) was determined by the following equation:

$$M_t = \frac{W_t - W_0}{W_0} * 100(\%) \tag{1}$$

Where W_0 is the weight of dry specimen and W_t is the weight of wet specimen at time t. Generally, in vegetal fibre reinforced plastics, moisture uptake follows a Fickian behaviour. In literature, several models have been developed in order to describe the moisture absorption parameters of composites materials. In the case of one-dimensional approach, Fickôs laws show that the water uptake increases linearly with the square root of time, and then gradually slows until an equilibrium plateau is reached.

$$H = \frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} \sum_{i=0}^{\infty} \frac{1}{(2i+1)^2} \exp\left(\frac{-(2i+1)^2 \pi^2 D t}{h^2} \right) \tag{2}$$

Where D is the diffusion coefficient and h is the thickness of the specimen.

With the aim of determining the diffusion coefficient D, an analytical modelling by using Matlab software was developped. From experimental data, an optimization program was made to determine this coefficient. This optimization was based on the minimisation of the quadratic error (Eq. 3) between the mesured $H_M = M_t/M_\infty$ from experimental curves and the predicted H_F calculated from Eq 2, as described in [11]:

$$q_I = \sum_{I}^{n} (H_M - H_F)^2 \quad (I = 1, 2, \ldots .n) \tag{3}$$

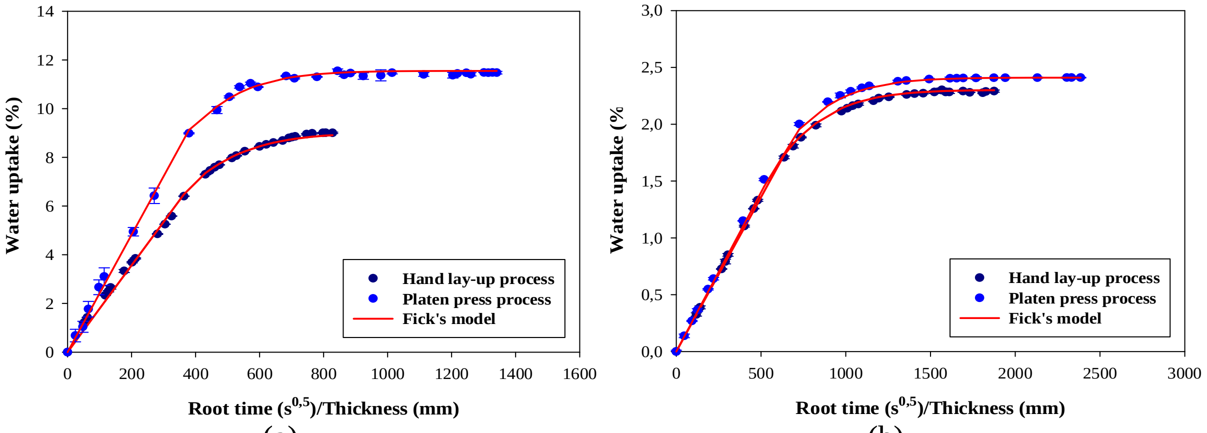
2.3 Testing procedures

The tensile tests for all the samples were conducted with an universal mechanical testing machine Instron (model 3382) at room temperature, with a cross-head speed of 2 mm.min^{-1}. All the tensile test specimens are in rectangular form (20 mm x 247 mm) according to ASTM D3039-76 standard. A clip-on extensometer with 50 mm gauge length was used to measure the longitudinal strain. Tensile tests were coupled with Acoustic Emission (EA) measurements achieved by using two piezoelectric sensors with a frequency range 100 kHzï1 MHz, as described in [7]. The AE technique enabled to analyse the different types of damage mechanism of these composites and to obtain several informations about their degradation process.

3. Results and discussion

3.1 Water absorption

Figure 1 shows the percentage of water absorbed for FFRE and GFRE composites, according to the square root of ageing time divided by thickness h for both manufacturing processes. The solid curves show the theoretical water uptake behaviour calculated from Eq. (2). The curves in figure 1 present two different parts. In the first part, which is linear, the water uptake increases proportionally with increasing the square root of ageing time divided by thickness. The second part is non-linear until the saturation point, that is to say the weight gain increases gradually until an equilibrium plateau is reached. We also remark that the percentage of water uptake is different for both composites and for each manufacturing process. For the hand lay-up process, the weight saturation of FFRE composite is four times more important that of GFRE composite (around 9% for the FFRE composite and 2% for the GFRE composite). In the case of the platen press process, the weights saturation were 11% and 2.5% for FFRE and GFRE composites, respectively. These results show that FFRE composite is more sensitive to the water absorption than GFRE composite. This can be explained by flax-fibre morphology. Indeed, at the centre of the elementary flax-fibre, the hollow part in the middle called lumen, contributes to the water absorption process [12]. The diffusion coefficients, calculated from Eq (2) also depend on the nature of the reinforcement (flax-fibre or glass-fibre) and the manufacturing process for FFRE composite. For GFRE composite, the diffusion coefficients are almost equal for both manufacturing processes. For example, in the case of FFRE, the diffusion coefficient *D* is about 7 x 10^{-7} $mm^2.s^{-1}$ for hand lay-up process and 8 x 10^{-7} $mm^2.s^{-1}$ for platen press process. This is directly related to the flax content that is different for both manufacturing processes (table 1).



(a) (b)

Figure 1. Evolution of water uptake of samples after water ageing for both manufacturing process: a) FFRE, b) GFRE

3.2 Mechanical properties

In order to study the influence of manufacturing process and water ageing on the mechanical behaviour of FFRE and GFRE composites, we illustrate in figure 2 typical stressï strain curves obtained for aged and unaged FFRE and GFRE composites for both manufacturing process. The behaviour of aged and unaged GFRE is linear until the failure (figures 2c and 2d), whereas the flaxï fibre composite presents two linear zones (figures 2a and 2b). The first one, which is purely elastic and enables measuring the modulus of elasticity in tensile test. The second zone is also linear until the failure. These two different parts have been recently described on the works of Poilâne et al [13]. For aged FFRE composite, we observe a moderate increase of the curve slope until failure. This could be attributed to the sliding of the primary and secondary walls of flax fibre caused by the interface weakening between these walls (due to humidity), which leads to the reorientation of the microfibrils [7]. In order to assess the influence of manufacturing process and water ageing on the mechanical properties of FFRE and GFRE composites, figure 3 illustrate the evolution of the Youngĝs modulus and maximal stress properties of unaged and aged materials after water ageing. These results show that the manufacturing process affect the mechanical properties of FFRE and GFRE. It is observed that the platen press process increases the Young's modulus and maximal stress of both composites. This increase is related to the fibres volume fraction which is different for both manufacturing process. For example, in the case of FFRE composite, the fibres volume fractions are respectively 0.25 and 0.35 for hand lay-up and platen press process.

In order to assess the contribution to the fibres volume fraction to the effective composite modulus, the longitudinal elastic modules of the composite were calculated using the rules of mixture:

$$E_c = E_f V_f + E_m (1 - V_f) \tag{4}$$

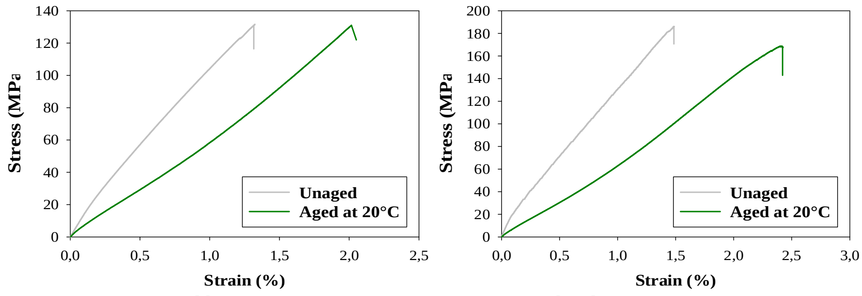
Where E_f is the longitudinal modulus of the fibres, E_m is the modulus of elasticity of the matrix and V_f is the fibres volume fraction.

For the flax fibre, the elastic modulus parallel to its axis was assumed to be equal to 52.5 GPa, average value proposed by Baley et al. [14]. The calculated stiffness of the FFRE composite, parallel to the fibre axis, is equal to 15 GPa for $V_f = 0.25$ (hand lay-up process) and 20 GPa for $V_f = 0.35$ (platen press process). We remark that, by using the rules of mixture, the increasing of the fibres volume fraction leads to a theoretical increase in the longitudinal modulus of 5 GPa. Experimental results show that the modulus increases more, from 14 GPa to 22 GPa, i.e. from 8 GPa. The technique of platen press process only improves the stiffness, which could be explained by a flattening of the fabric and a better fibre-matrix cohesion.

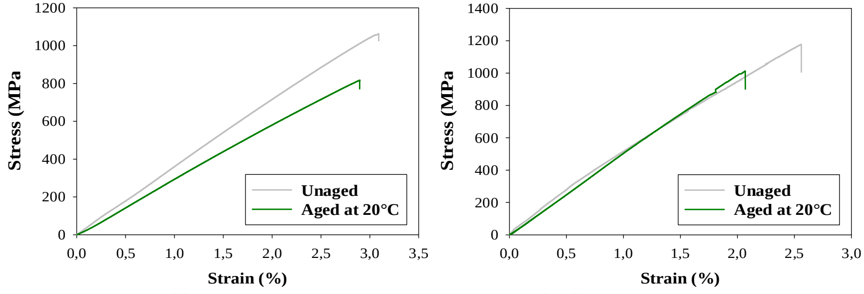
Similar considerations may apply to GFRE composites, but are more severe. The rules of mixture indicate an increase of 3 GPa with the increasing of V_f from 0.50 to 0.54, whereas we observe a much higher increase of 14 GPa with experimental tests.

These results show, besides the effect of the fibres volume fractions, an improvement of the longitudinal modules caused by the manufacturing processes for both composites.

These results also show that the water ageing leads to significant variations in the mechanical properties. Indeed, after 52 days of ageing, figure 4 shows that Young's modulus is affected by water ageing. For aged FFRE composite, Young's modulus decreases by 54% for hand lay-up process and by 58% for platen press process (figure 3a). The evolution is different for the tensile strength because the ageing leads to a slight decrease of this property by 1% and by 6%, respectively for hand lay-up and platen press process (figure 3b). This could be related to the reorientation of the microfibrils and the swelling of flax fibres caused by the moisture.

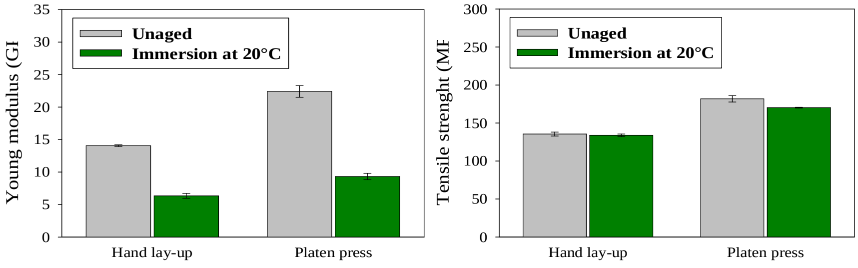


a) Hand lay-up process FFRE b) Platen press process FFRE

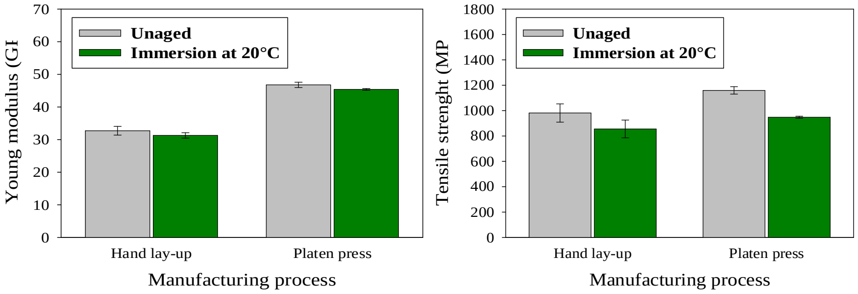


c) Hand lay-up process GFRE d) Platen press process GFRE

Figure 2. Stress-strain response of unaged and aged FFRE and GFRE composites under tensile loading



a) Youngôs modulus FFRE b) Tensile strength FFRE



c) Youngôs modulus GFRE d) Tensile strength GFRE

Figure 3. Evolution of the mechanical properties of FFRE and GFRE composites according to the manufacturing process

3.3 Damage mechanisms analysis

During the ageing process of composites materials, several damage mechanisms at the microscopic scale can occur according the composite type and the loading direction relative to the fibres direction. In order to get information about these mechanisms, the different signals recorded during tensile tests were analysed by considering the tensile stress, the hits number and the amplitude of AE events according the loading time, as described in Figure 4. The obtained results of AE signals show that the matrix cracking and fibre matrix debonding for amplitudes between 40 and 60 dB [8] presents more than 90 % of damage. This mainly govern the damage within the studied materials. The acoustic activities start from the beginning of tensile test for FFRE and GFRE composites. This can be explained by the degradation of the matrix as well as that of the matrix/fibre interface caused by the water ageing [8]. Figure 4 also shows that the damage mechanisms evolution is different for unaged and aged composites. Indeed, we observe a decrease of the number of acoustic events with the water ageing. The acoustic activity remains very low during the first seconds of the loading test. Then, the number of hits increases gradually until the failure. These results are in agreement with the decrease in the mechanical properties for both composites.

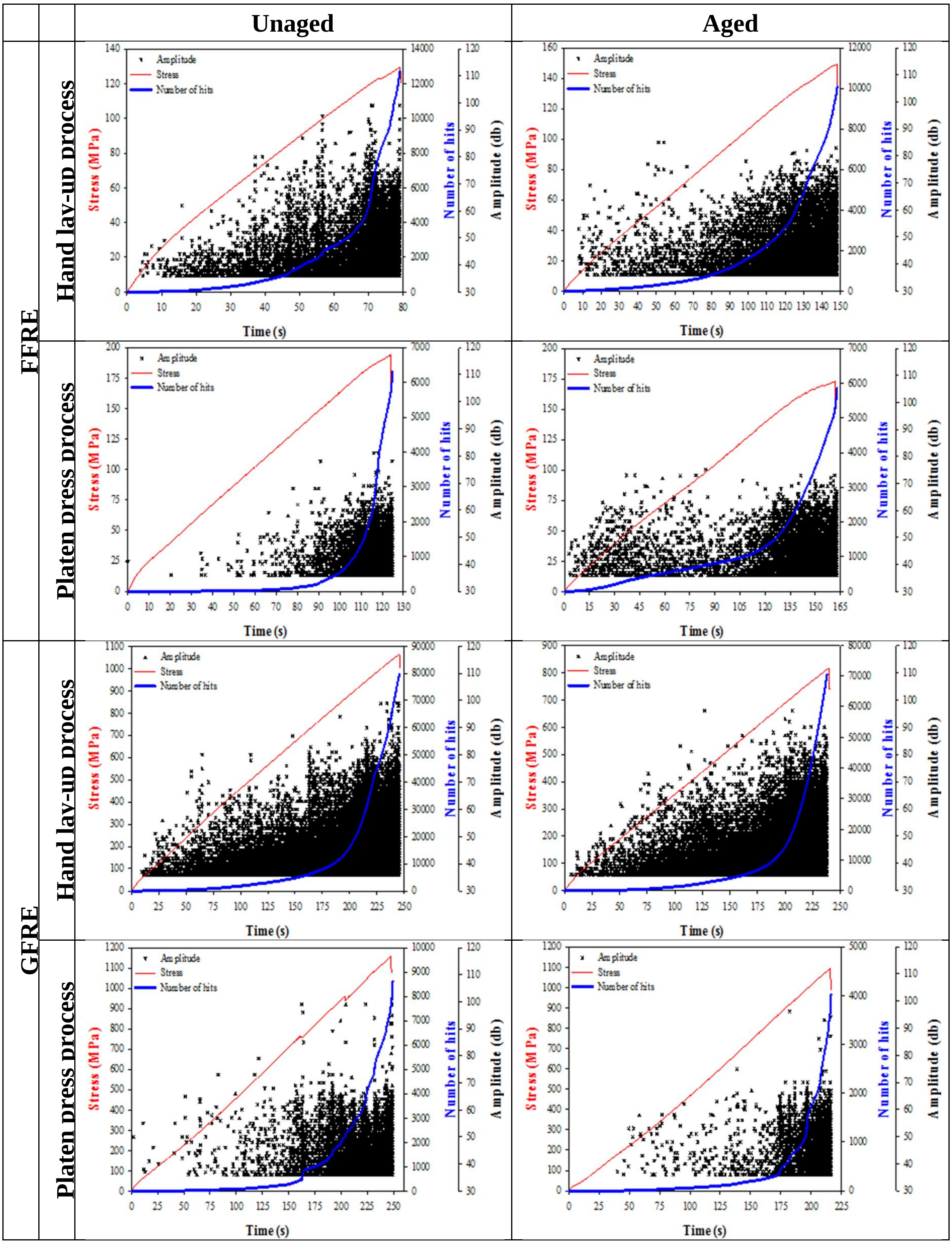


Figure 4. Tensile stress, hits number and amplitudes of AE signals versus tensile tests time for unaged and aged FFRE and GFRE composites

4. Conclusion

This study present the effect of manufacturing process and water ageing on the mechanical properties of quasi-unidirectional Flaxï Fibre Reinforced Epoxy (FFRE) and quasi-unidirectional Glassï Fibre Reinforced Epoxy (GFRE), which are subjected to water ageing at room temperature. The obtained results show that the water absorption behaviour of both composites is found to follow Fickian behaviour. The mechanical properties of both composites are clearly affected by the water ageing; a significant reduction in the Youngôs modulus was observed. The obtained results from the classification of AE signals enabled describing quantitatively the different damage mechanisms.

Finally, the implementation technique that was considered for the creation of the various samples of these materials (hand lay-up process and platen press process at room temperature) significantly affects the tensile properties of FFRE and GFRE composites and thereby modifies their behaviour in a moist environment.

For the future work, it would be interesting to study the damage mechanisms by AE technique combined with scanning electron microscopy examination in order to have a better understanding of our experimental data.

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