Damage analysis based on the correlation between acoustic emission and E modulus degradation in flax/epoxy quasi unidirectional woven laminates

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Abstract

In the present work damage was monitored in flax/epoxy quasi unidirectional woven laminates. Several plates with different lay-up configurations: $[0°]_8$,$[0°,90°]_{2s}$,$[0°,90°,+45°,-45°]_s$ and $[+45,-45]_{2s}$ were prepared in autoclave, then the damage was monitored during the tensile test using acoustic emission technique. The tensile tests show that these composites offer good mechanical properties. The acoustic emission diagrams allowed us to follow the evolution of damage and to identify several parameters: Energy, damage threshold and the number of events, however the correlation between the stress-strain curves and AE results don't show a direct relationship between the two damage indicators (E&AE): this suggests that the shape of the strain-strain curves is due predominately to another factor than AE events.

1.Introduction

Natural fibres are being extensively explored by automotive, aviation, marine, civil and packaging industries as an environmental-friendly alternative to synthetic fibre composites [1]. Flax is probably the most commonly used bast-type fibre today. Due to its properties and availability, flax fibre has potential to substitute glass in polymer composites, even with the strength performance lower than for glass. A flax fibres can be classified into elementary fibres (sometimes referred to as fibre ultimates), which are grouped into so-called technical fibres consisting of 2-5 elementary fibres. The technical fibres are composed of elementary fibres kept together mainly by pectins, meaning that the technical fibres themselves are actually composite structures. Technical fibres have diameter of 30–300 μm, while elementary fibres have diameter of 15–35 μm [2]. The elementary fibres are the single plant cells made of concentric cell walls. Morphologically, an elementary fibre consists of a primary thin cell wall, a secondary thick cell wall and a lumen (of variable dimensions). This lumen is an open channel in the centre of the fibre. Primary and secondary cell walls differ in composition as well as in thickness. The primary walls are principally made out of pectin, while the secondary walls are made from cellulose and hemicelluloses. The latter contains numerous helically wound cellular microfibrils, which are often laid down in layers having defined orientations (microfibrillar angle) with the axis of the fibre [3]. A difference in physical, chemical, and mechanical properties results from these morphological and constitutional differences between the two walls. The elementary flax fibre contains 65-75% of crystalline cellulose (responsible for stiffness), approximately 15% of amorphous hemicellulose (responsible for strength), and 10-15% of pectin [4]. Prior studies [3,5] determined mechanical properties of a single flax fibre. It has been shown that the tensile strain-stress curves exhibit a nonlinear behaviour at the early stage of loading, with fibre stiffness decreased after the applied strain reaches ca 0.3%. This is a result of the fibre´s complex morphology and is therefore an intrinsic characteristic of the flax. Due to this complex morphology and to the anisotropic nature, flax 'mechanical properties show a large scattering. However, we can cite here the most frequently published data of the mean longitudinal values: Young's Modulus, ultimate strength and ultimate strain, which are about 70 GPa, 700 MPa and 0.3%, respectively [6]. Transverse modulus is estimated to be 8 GPa [7]. Damage behaviour studies [2, 8, 9] were performed on a unidirectional flax fibres reinforced composites, where the materials were tested under tensile loading. At the microscopic level and at low stress, microcracks arise within the material and by growing they may lead to other forms of damage such as delamination, fibre breakage, interfacial debonding…etc. In order to better understand the damage phenomena and to better control the parameters which lead to the failure, several methods and techniques have been developed in this field. Among these techniques, the AE occupies an important place in damage in-situ monitoring [10,11,12,13], including application to natural fibre reinforced composites [8,9,14]. The present paper continues this streamline of research, enriching the experimental evidence and analysis of the damage processes in flax/epoxy composites.

2. MATERIALS AND TEST METHODS

The studied material is a quasi-unidirectional woven prepregs, flax/epoxy, with areal density of the reinforcement 170 g/m2, provided by LINEO (Belgium). The woven fabric is pre-impregnated with epoxy resin (Araldite LY5150). The laminates were made in the different configurations: $[0°]_8$, $[0°, 90°]_{2s}$, $[-45°, 45°]_{2s}$ and $[0°, 90°,+45°, -45°]_s$, they were then processed using autoclave. A total fibre volume fraction of all the laminates is of 47 ± 2%. The temperature and the pressure during autoclaving reached 130°C and 4 bars, respectively. In order to analyze the plates cross sections, specimens of about 1x1 cm2 were cut from the plates' edges; these cross sections were then observed under an optical microscope. The micrographs highlight the different sequences of the laminates and reveal also a good quality of impregnation: no voids were observed in all of the prepared laminates. Rectangular samples, 250 mm long and 25 mm wide, were cut out from the 2 mm thick plates, using a diamond wheel saw, to be further subjected to tensile tests in the 0° direction. To avoid failure due to the stress concentration of the machine clamps, a 5 mm x 25 mm x 40 mm glass/epoxy composite tabs were glued at the ends of each specimen. The length of each specimen between the grips was 170 mm.

2.1 TENSILE TETS

Tensile tests were performed using an INSTRON 4505 machine with a loading capacity of 100 kN according to ASTM D3039 standard. The crosshead speed, temperature and humidity during the tests were 5 mm/min, 20°C and 50%, respectively. For each configuration three tests were carried out.

2.2 ACOUSTIC EMISSION AND STRAIN MEASUREMENT

To monitor in situ the damage development in the laminates, the traction machine was connected to the AE VALLEN equipment. The preamplification was 34dB and the noise was filtered using a threshold of 40 dB. In order to provide a good acoustic coupling, the sensors surfaces were covered with silicon grease and then attached at the two extremities of the specimen. The nominal distance between the sensors was 10 cm. Three tensile tests with AE recording were performed for each configuration. The strains were measured using a Vic2D software (the software LIMESS Maßtechnik und Software GmbH, 2006) and a camera which captured the images of the sample's center each 0,5 s during loading.

3. RESULTS AND DISCUSSION

3.1 SRAIN-STRAIN CURVES

The typical stress-strain curves for the studied laminates are represented in fig.1. The ductile behaviour is seen for $[+45°,-45°]_{2s}$ laminate, while for other laminates the brittle behaviour is observed. It is also clearly seen that these curves exhibit a non-linear behaviour at the early stage of loading. On the basis of these curves, the tangent modulus (E) degradation, during loading, was evaluated and the variation of E versus strain for one characteristic stress-strain diagram was plotted in the same graph (fig.1). Modulus versus strain curves displays three main parts. In the first portion (1) and at the low strains (approximately up to 0.2% of strain) for the $[0°]_8$, $[0°,90°]_{2s}$ and the $[0°, 90°, +45°, -45°]_s$ laminates and (up to 0,5% of strain) in the case of the $[+45°, -45°]_{2s}$ composite, the curve has a plateau. This plateau is followed by a second part (2) where the curve decreases rapidly and continuously, terminating by another plateau (3) where the slope (E) becomes constant again until the failure of the laminate (4). The Young's modulus for the different composites was then calculated by averaging the first linear part's values (Table 1). One can see the slight difference in the onset of non-linearity between the $[0°]_8$ and $[0°,90°]_{2s}$ laminates which is about 0,12% and 0,14% of strain, respectively. However, in the other laminates the non-linearity occurs at a higher strain value being 0,2% of strain in $[0°,90°,+45°,-45°]_s$ and 0,5% of strain in $[+45°,-45°]_{2s}$ laminate. Young's modulus values and the main mechanical properties: ultimate strength, ultimate strain and Poisson ratio, identified from stress-strain curves are listed in table.1. As mentioned above, the studies reported in [3,5], demonstrate the non-linearity phenomenon exhibited by a single fibre under uniaxial loading, similar to the non-linearity observed in the present work for laminates. This non-linearity was attributed by these authors to changes of the fibre morphology on micro-scale which occur during loading. In fact, the microfibrils which are embedded in a matrix of amorphous hemicellulose and which are oriented initially with a spiral angle of about 10° [3] with the axis of the fibre, exhibit a unidirectional structure. When subjected to a uniaxial loading (parallel to the fibre axis) they could undergo rearrangements [3,5] so that to be aligned as much as possible with the tensile axis. These micromechanical rearrangements are interpreted by the non-linear behaviour. Moreover, apart its complex morphology, flax is characterized by some defects, particularly the presence of "kink bands" along its length. When the fibre is under uniaxial loading these "kink bands" may undergo extensions. The non-linearity phenomenon is considered to be due to the extensions of such defects in association with the realignment of the microfibrils [3]. Further studies [9] were carried out on unidirectional flax fibre reinforced thermosetting resin composite. Under uniaxial loading the reported stress-strain curves display similar behaviour with those obtained in a single flax fibre [3,5]. The three main stages of modulus evolution were well observed. It has also been found that the stiffness, in both studies, increases with the tensile loading. The non-linearity displayed in the single flax fibre, is seen in the range 0,3-1,5% of strain [5]. Apart from similarities, there are differences in behaviour of single flax fibre or unidirectional flax fibres reinforced polyester composites and quasi-UD woven laminates studied in the present work. The range of non-linearity observed in the $[0°]_8$ is about 0.12-0.8%. The shift of this interval to lower strains in our measurements can be explained by the fact that in the quasi-UD woven fabric considerable crimp of the yarns/fibres increases the local strain in the fibres. Although the three stages of E evolution were observed in [3, 5, 9], it has been found in all of them that the non-linear region (2) (drop in the stiffness), is followed by a gradual increase of the stiffness. Whereas, in the present work as well as that reported in [15], the decrease in the stiffness is directly and lastly followed by a stable value of stiffness. The stability displayed at the last stage of E modulus let us support the hypothesis suggested by Baley et al: it may be related in part to the final stage of microfbrils re-alignment. In the $[0°,90°]_{2s}$ laminate, the curve (fig.1.b) shows an extent of non-linearity from 0,14 to 0,7% of strain, which is practically the same as displayed in $[0°]_8$ laminate, which means that the non-linearity is mainly caused by the intrinsic non-linearity of the fibres. This corresponds to the fact that in flax composites no matrix cracks in 90° yarns (which can contribute to the stiffness degradation) were observed till later stages of the loading. In the case of $[0°, 90°, +45°,-45°]_s$, this width of non-linearity extent is larger (0.2-1.4% of strain). This is not surprising, as the quasi-isotropic laminate feels also the influence of 45° plies, which manifests itself ultimately in quasi-ductile behaviour of $[+45°, -45°]_{2s}$ laminates with the wide non-linear region of the tensile diagram. For the $[+45°,-45°]_{2s}$ laminate the response of the material appears clearly to be entirely different: here the laminate behaves elastically at the beginning, followed by a quasi-plastic deformation till failure, as it is observed during bias loading of cross-ply laminate with glass or carbon fibres [16,17].The observed behaviour of flax laminates in tension differ in details from the one of glass fibre laminates. Glass fibres are perfectly linear in their tensile response, so is the [0°]n laminate. The non-linearity of the glass fibre cross-laminate stress-strain curve is explained by appearance of damage (matrix damage in 90° plies, local delaminations…etc.) [18]. In the flax reinforced laminates the non-linear behaviour of the fibres is the prevailing factor, defining the non-linearity of tension stress-strain curves for loading in the fibre directions. The effect of matrix damage is less, first because it occurs later, second, because of the intrinsic stiffening of the flax fibres. The latter feature is analogous to the behaviour of carbon textile composites [19].

3.2 ACOUSTIC EMISSION RESULTS

The damage initiation and development in the studied laminates were monitored using AE. The AE activity recorded during tensile tests is reported in term of energy and acoustic emission events. To better understand damage mechanisms which occur during loading, acoustic emission energy, cumulative energy and stress-strain curves are drawn in the same graph (fig.2). In order to avoid their damage, the two sensors were removed before the final failure of the specimen, at approximately 80 to 90% of the final failure stress. The analysis of the acoustic emission diagrams (fig.2) allowed us to identify damage thresholds in the different laminates. It is apparent that the threshold of damage is the lowest in the $[0°]_8$ laminate being 0.45% of strain. The AE diagram relative to $[+45°,-45°]_{2s}$ (figure.5.d) shows the highest damage initiation value, which reaches about 0.87 % of strain. These diagrams show also that E modulus decreases before AE events have taken place. This loss in the stiffness could be then most probably attributed to intrinsic progressive changes of flax than to the AE events. It can also be seen, in the second linear part of the stress-strain curve relative to $[0°]_8$, $[0°,90°]_{2s}$ and $[0°,90°,+45°,-45°]_s$ laminates, that damage occurs without affecting the stiffness of the material. As it can also be observable, the highest AE energy occurs in the $[0°]_8$ laminate. In case of $[+45°,-45°]_{2s}$ laminate the amount of the emitted energy is significantly lower which is approximately 10^3 times lower than the total amount of the energy emitted in the $[0°]_8$ laminate. The correlation between the overall behaviour of E curves with the distribution of AE signals in all of the studied laminates didn't show a direct relationship between these two damage indicators. The first one (E) may include mainly micromechanical changes relative to the fibre and its microfibrils. In order to highlight the AE activity occurring during loading, the total cumulative AE events versus strain for the different laminates and their mechanical behavior (stress against strain) were plotted in (figure.3). These graphs show that the number of events is substantially higher in the $[0°]_8$ laminate which is around 1000 events. The lowest number of the emitted events is observed in the $[0°, 90°,+45°,-45°]_s$ laminate (35events). This suggests that in this latter less damage occurs comparing to the other studied materials. From figure.3, one can deduce that the number of events decreases as a function of the decrease of 0° layers in $[0°]_8$, $[0°,90°]_{2s}$ and $[0°,90°,+45,-45]_s$ laminates. This dependence between the number of events and the number of 0° lamina, in addition to the fact that no matrix cracking was observed until the last stage of loading suggests that most of the events in $[0°]_8$ laminate arise mainly from the complex mechanism of fibre breakage [20]. Figure.3.a shows also that for $[0°]_8$ composite, intensive AE activity occurs at approximately 0.6% of strain. This curve exhibits an initial rise in events till 0.6% of strain, followed by a linear part. In case of [0°, 90°, +45°,-45°]s and [+45°,-45°]2s , figures.3.b and 3.c show that the curves display two main regions. In the first region the number of events increases slowly exhibiting almost a plateau shape followed by a second region where the curve approaches linearity. Intensive AE activity is seen, for both laminates, around 1% of strain. Comparing to $[0°]_8$ laminate the delay in AE activity can be explained by the fact that in the $[0°,90]_{2s}$ and the $[0°,90°,+45°,-45°]_s$ materials the interactions between the affected fibres are secured by 90° layers. The similar behaviour of these two last curves suggests that some common damage modes may occur in both laminates. Concerning the $[+45°,-45°]_{2s}$, here the events versus strain graph shows almost a linear behaviour, suggesting that one damage mode is predominant in this case which could be ascribed to matrix damage. There are also differences between the nature of cumulative events curves relative to the four laminates; for the $[0°]_8$ laminate the curve is smooth, whereas in $[0°,90°]_{2s}$ laminate the curve reveals the presence of some inhomogeneities (bursts) (figures.3.b) which indicate that some of simultaneous events occur at practically the same strain. These bursts are notably present at high strains where they became larger. In case of the $[0°,90°,+45°,-45°]_{2s}$ the curve shows that these inhomegeneities are much less pronounced (figures.6.c) and in $[+45°,-45°]_{2s}$ laminate we notice their presence notably at low strains (figures.3.d). These bursts may be considered to be characteristic of matrix deformation and/or of small cracks which in case of $[0°,90°]_{2s}$ laminate may develop when load increases, as it can be seen in figure.3.b. The correlation between figure.2 and figure.3 also reinforces the idea that there are no evidence of the relationship between the loss in the stiffness and the AE activity; contrary to expectations, no impact of the increase of AE events on the degradation of E modulus can be seen. For example, in the $[0°]_8$ laminate the loss in the stiffness which accompanies region2 reaches approximately 50% of the initial value after a considerable number of events is emitted. Whereas, in $[0°, 90°]_{2s}$ laminate the non-linear region is accompanied by the same percentage of the loss in the stiffness (≈50% of the initial value), whilst the amount of the emitted events is very low, practically negligible comparing to the total amount of the emitted events. This corroborates, the idea that the bilinearity displayed in the stress-strain curves is predominantly due to another factor rather than to the AE events.

CONCLUSION

In this study the morphology of flax and the mechanical properties of flax/epoxy composites were investigated. The microscopy observations highlighted the complex morphology of the flax fibre and the results show that the flax fibre reinforced composites offer good mechanical properties comparing to glass fibres reinforced composites. The tensile tests reveal a specific behaviour of the strain-stress curves, characterized by a bilinearity, comparing to glass fibres reinforced composites the behaviour of the stress-strain curves is predominantly linear. The acoustic emission diagrams allowed us to follow the evolution of damage and to identify several parameters relative the four studied laminates: Energy, damage threshold and the number of events, however the correlation between the stress-strain curves and AE results don't show a direct relationship between the two damage indicators: this suggests that the shape of the strain-strain curves is due predominately to another factor than AE events.

	Tensile tests results					**AE results**	
	Interval of linearity (strain interval ε%	InitialYoung's modulus (GPa)	Poisson's ratio **ν**	failure (ultimate strength)(MPa)	Strain at failure (ultimate strain) ε %	Damage threshold, strain%ε%	energy (au)
$[0°]_8$	0-0.12	27.18±0,52	0.409±0.034	296±0.5	1.65±0.055	0.45	$5.86x10^7$
$[0°,90°]_{2s}$	0.-0.14	15.69±0.15	0.126±0.014	158±2	1.62±0.5	0.54	$2.35x10^6$
$[0°,90°,+45°,45°]_s$	0-0.2	11.89±0.6	0.319±0.259	126±7.5	1.76±0.15	0.8	$1.8x10^5$
$[+4°5,-45°]_{2s}$	0-0.5	5.7±0.11	0.566±0.064	85±4	7.47±0.415	0.87	$2.9x10^4$

Table.1 *Table summing up the main tensile and AE results.* Note: the scatter gives the standard deviation in three tests.

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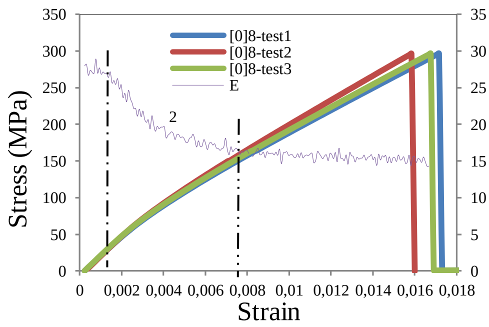

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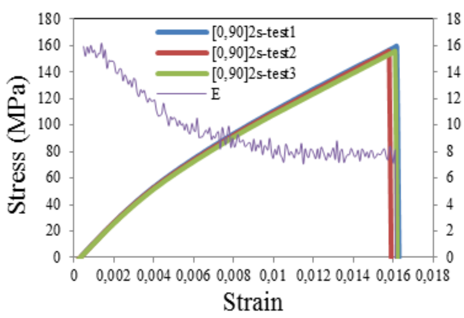

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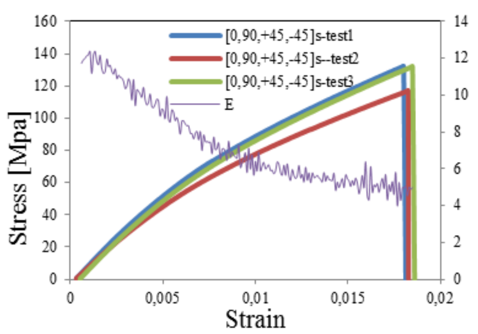

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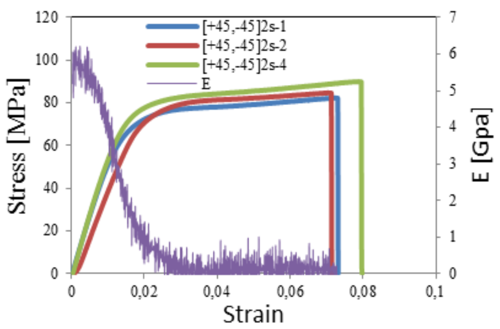

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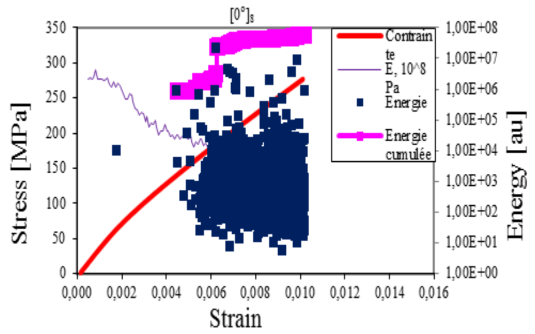
Fig.1 Strain-stress curves for the different studied laminates.



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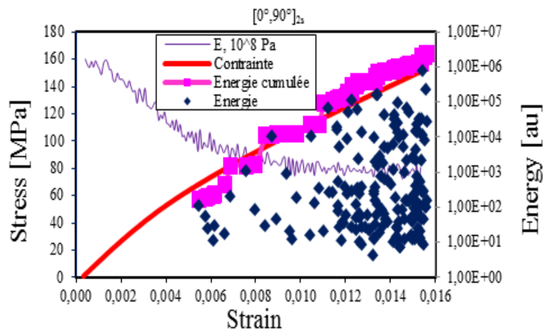

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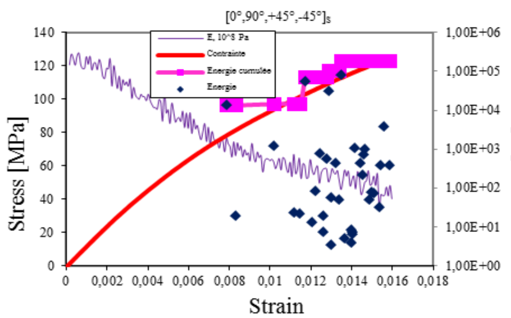

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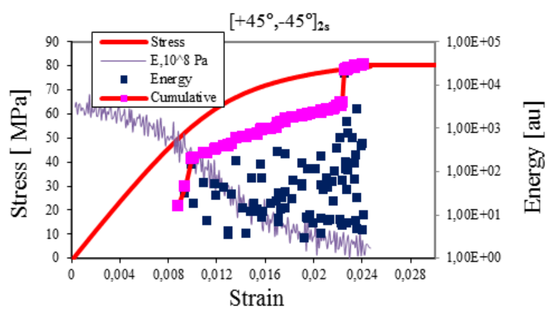

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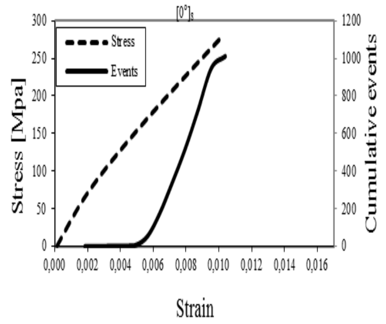
Fig.2 Acoustic emission diagrams of the different laminates.



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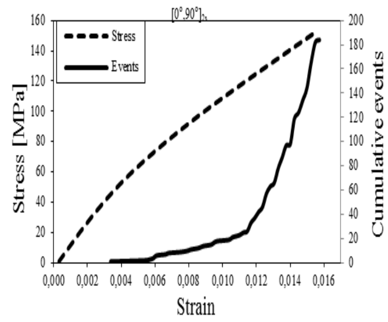

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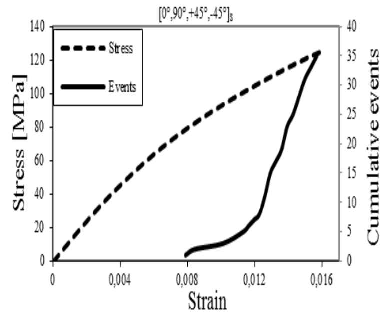

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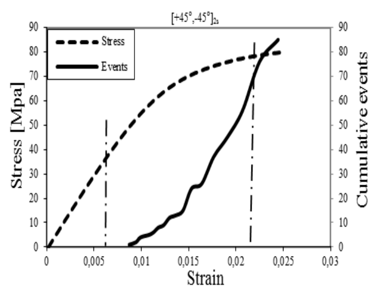

(d)

Fig.3 Cumulative AE events versus strain of the different studied laminates.

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