Contribution of Acoustic Emission to Evaluate the Influence of Hygrothermal Aging on Mechanical Behavior of Hemp Reinforced Polypropylene Composites

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ABSTRACT

In recent years high interests of scientific and industrial worlds were concentrated on natural fibers composites. Natural fibers as hemp, flax and sisal have become suitable alternatives to glass fibers as NFs present several advantages as lightness, strength, recyclability and are relatively cheap and abundant. However, their high hygroscopic nature and their sensitivity to temperature must be taken into account. In this paper, the influence of hygrothermal aging on mechanical behaviour of Hemp/ isotactic polypropylene composites were studied using flexural tests associated to acoustic emission (AE).

INTRODUCTION

Natural fibres (NF) such as jute, coir, sisal, bamboo and pineapple are known to have high specific strength and can be effectively used in composites in various applications [1]. However, unlike traditional synthetic fibers (carbon, glass, Kevlar,…etc.), the behavior of NF depends strongly on temperature and humidity [2] . Exposure to certain levels of temperature can lead to a loss of integrity and in particular to thermal and oxidative degradation of fibers [3]. Also, their high hygroscopic nature must be taken into account. Indeed, water and humidity can act as a plasticizer and a degradation agent. In this paper, the influence of hygrothermal aging on mechanical behaviour of Hemp/ isotactic polypropylene composites were studied using flexural tests associated to acoustic emission (AE).

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MATERIALS AND METHODS

Materials

The constituent materials of composites used in this investigation are made from non woven hemp fiber mat (400g/m^2) randomly oriented in the plane and isotactic polypropylene (iPP) (Moplen HP500V) in powder form The micronized powder average diameter is in the range 50–200 µm. Fibers were used in as received state. The humidity content of the fibres is about 4%. The dry impregnation was made in two steps: the iPP powder was first scattered on the top of the substrates using manual sieve and then the use of high voltage electric field allows the distribution of powder throughout the porous structure (Fibroline method). The impregnated Hemp/polypropylene mats were then by thermo-compressed in a steel mold at 200°C and 55 bars pressure. The fiber average weight fraction is 40%.

Hygrothermal aging

Hygrothermal behavior of hemp/PP composites was achieved by exposure to 80% relative humidity at three temperatures. The relative humidity was controlled using supersaturated salt solutions (NaCl and KCl). The maximum considered aging time in this study was equal to 90 days. Hemp/PP samples were cut in 4 mm thick composite sheets and then polished to eliminate surface irregularities. Sample dimensions were about 75 mm long, 10 mm wide and 4 mm thick

For water absorption measurements, the specimens were withdrawn from the water, wiped dry to remove the surface moisture and then weighed using an electronic balance accurate to 10^{-5} g to monitor the mass change during the water absorption process. The moisture content M(t) absorbed by each specimen was calculated from its initial weight (w_0) and its weight after absorption (w_t) as follows:

$$M(t) = 100.\left(\frac{w_t - w_0}{w_0}\right) \qquad (1)$$

Three point bending test

Three point bending tests were carried out using an INSTRON-5582 test machine at 2mm/min crosshead speed in a conditioned room (T = 23 ± 2ºC; RH = 50 ± 5 %). The sample dimensions were about 80 mm long, 10 mm wide and 4 mm thick. The span to depth ratio (L/h) is chosen to be 15, to minimize shear stress.

Acoustic emission

Acoustic emissions (AE) signals during bending tests were acquired and processed with the AE acquisition and analyzing system Mistras 2001 from Physical Acoustics Corporation (PAC). The AE signals were detected by a piezoelectric sensor (nano 30) attached to the sample by a silicon grease and a mechanical specific device on the tensile side. The signals were amplified by a pre and a post-amplifier with a total gain of 70dB. The detection threshold of AE was fixed at 35dB.

RESULTS AND DISCUSSION

Hygrothermal aging

Figure 1 shows the moisture absorption (M(t)) variations according to the square root of time (t½) of the neat matrix (PP) and the hemp/PP composites exposed to 80% relative humidity at 23, 70 and 90°C. The neat PP matrix is almost inert to moisture. Indeed, it showed very little moisture uptake after moisture aging after three months whatever the temperature (about 0.02%) while moisture absorption levels for composites are in the range 1-2,5 %, depending on temperatures.

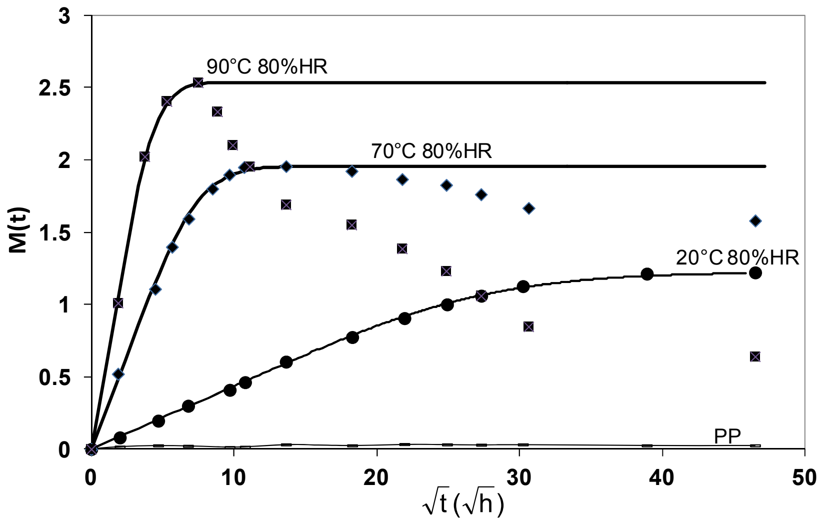

Figure 1. Water uptake variations as a function of the square rout of time of the neat matrix and hemp/PP composites exposed to humid atmosphere (80% RH) at various temperatures. Solid lines are fits based on Fick's law.

For ambient temperature the aging behavior of the composite can be considered as Fickian. For the highest temperatures, (70°C and 90°C), the composite curves showed similar profiles and one can distinguish the presence of three zones. The first one is linear corresponding to a rapid increase of moisture content. The second region corresponds to a plateau at which the aged material reaches saturation. This last region is partly overlapped by the third zone where a large decrease in moisture uptake is observed. This deviation from the Fickian profile which probably characterizes the start of material degradation of composites occurred after 14 days (70°C) and 3.5 days (90°C). As the moisture absorption for iPP is negligible, it can be assumed that the most of the water moisture is absorbed by the hemp fiber component in the composite [4]. In the cases of inorganic fiber (glass or carbon) reinforced polymer composites the weight gain depends mainly on matrix properties and volume content, at least in the early stages [5]. For natural fiber reinforced polymers the fibers must be taken into account. Indeed, the natural fibers are hydrophilic in nature. The hemp fiber cell wall is mainly composed of cellulose (≈ 72 %), hemicelluloses (≈ 10 %) and lignin (≈ 3 %) [6]. The high hydrophilic property of natural fibers is associated to the presence of hydroxyl groups in the carbohydrate fraction of the cell wall which have a strong possibility to interact with water molecules, forming hydrogen bonding to the detriment of those between the macromolecules. If the fiber is exposed to humid environmental conditions, water penetrates through the cell walls of the fiber and the fiber swells until the cell walls are fully saturated with water (bound water). The moisture content at which this occurs is called the fiber saturation point (FSP) [7] and is about 35% for free Hemp fiber [8]. Beyond this FSP, the water is present in the cell lumen and voids as free water. The maximum moisture levels absorbed by the hemp fibers in the composites can be then approximated considering the average fiber weight fraction (40 w%) and the maximum water content in the composites. The fibers show ≈ 3.1, 4.6 and 6.3% water absorption at 23, 70 and 90°C, respectively. Then, the hemp fiber maximum uptake is still well below the fiber saturation point.

Flexural tests

Flexural moduli decreased after exposure to humid atmospheres (80% RH) during three months. The mean decrease of composite moduli was 5 at 23°C, 9% at 70°C and 30% at 90°C according to as received composites. The mean decrease of the stress limit was about 1% at 23°C, 20% at 70°C and 60% at 90°C. As the neat polypropylene is not sensitive to water aging, the decrease in flexural properties can be therefore attributed to modifications of both fiber and interfaces properties. As earlier mentioned, when water came in contact with hemp fibers it penetrates into the cell wall acting thus as a plasticizer. The fiber cell wall has hence the tendency to undergo both reversible softening and swelling [9] which increase with immersion time until an equilibrium condition is attained. This swelling involves initiation of micro-cracks in the surrounding interfacial zones [10] causing the weakening of fiber/matrix interfaces. This phenomenon reduces the stress-transfer efficiency between fiber and matrix and facilitates further moisture penetration into the fibers and their degradation due to hydrolysis. This leads to deteriorated mechanical properties.

Acoustic emission

These trends are well highlighted by both SEM analysis and acoustic emission (AE) analysis. AE is an efficient technique for in-situ health monitoring of composite materials. This is achieved by monitoring the cumulative number of events and the analysis of AE parameters of the signal emitted by a failure event signature waveform (hit) like the amplitude, the energy or the duration of the event. The amplitude of the signal is often used because it is independent on the detection threshold. Several authors [11-12] have shown on different inorganic (glass or carbon) fibers reinforced composites that events with low amplitude (between 35–50 dB) are correlated with matrix microcracking, intermediate amplitude with delamination and microcracking coalescence (50 db to 60dB) and matrix/fiber debonding (60 dB-70 dB), high amplitude with fiber/matrix friction associated to pull-out (70 dB-85 db) and fiber breakages (>85 dB). However, it is more critical to achieve a relationship between amplitude and fiber failure mechanisms in the case of natural fiber reinforced polymer due to their complex architecture [13]. Different authors have shown that amplitude distributions of untreated hemp fiber failures [14-15] are about between 50and 90 dB, the amplitude of AE events due to high strength fiber breakages being significantly higher for than those of low strength fiber failures.

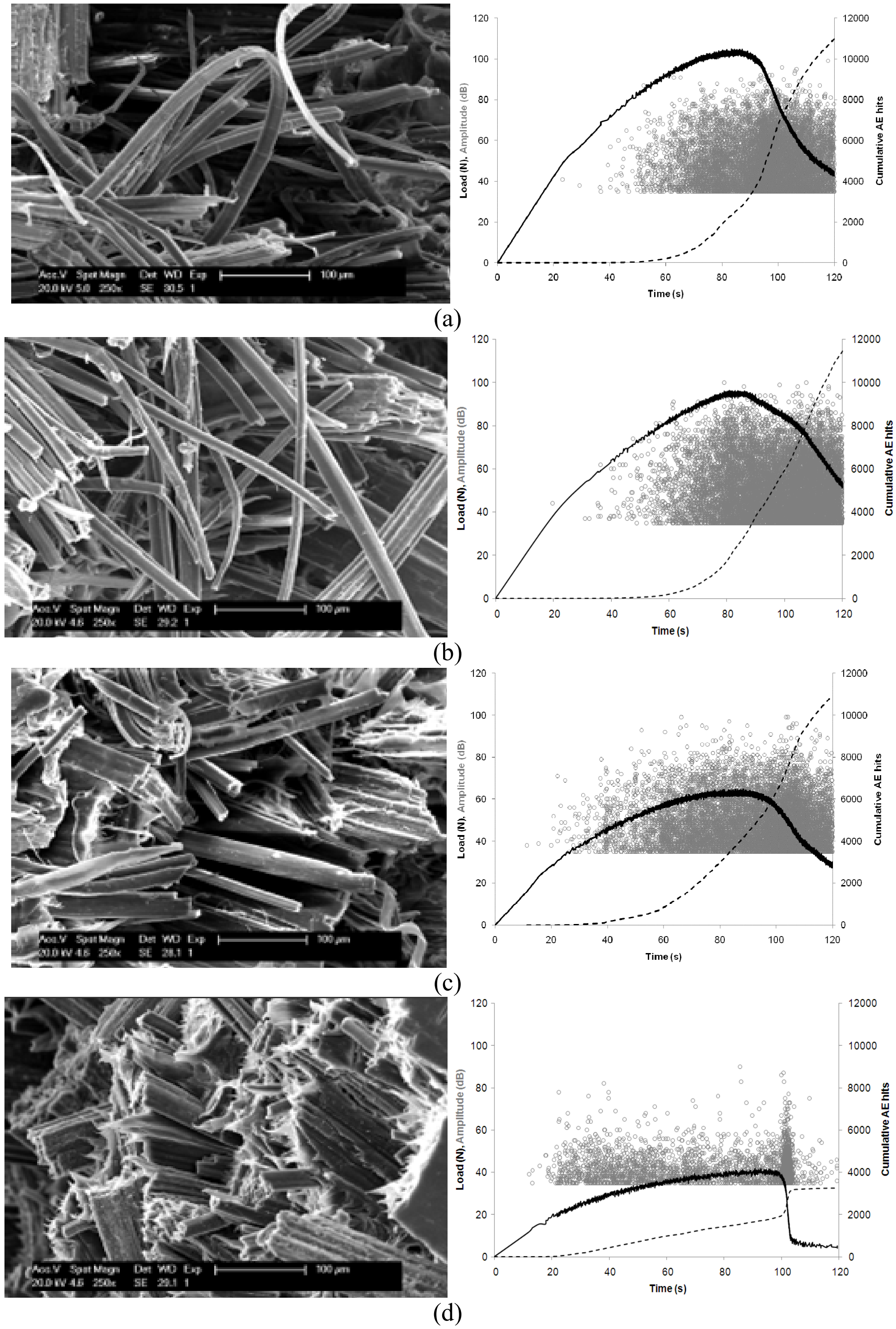

Figure 2. SEM fracture feature and EA analysis (Each individual event is indicated by a circle) during flexural test after three point bending test of hemp/PP composites, (a): as received, (b): Aged at 23°C 80%RH, (c): Aged at 70°C 80%RH, (d): Aged at 90°C 80%RH.

The SEM typical failure features observed on the tensile side of hemp/PP composites are presented in Figure 2 with the load and the cumulative total number of AE events, which are plotted according to time (deflection) and the amplitude of the AE signals for as received sample (a) and after aging in humid atmosphere (80%RH) at 23°C (b), 70°C (c) and 90°C (d). Figure 3 shows the acoustic emission event amplitude distribution for all the cases in order to discriminate fracture mechanisms. The pull-out phenomenon can be observed for as received samples (Figure 2.a). It consists of long extracted fiber almost clean with only slight matrix debris on the fiber surface, thus indicating that the stress transfer was well governed by mechanical interlocking and friction. The load-time curve shows a linear part corresponding to the elastic behavior. The maximum of the load corresponds to the occurrence of a macro-crack on the tensile side resulting of the coalescence of microcracks. Then the damage gradually progresses through the thickness. The deviation from linearity of the load-time (deflection) coincides with the onset of acoustic emission signals (>5 events) indicating that failures begin to occur (t = 40s). The plot of acoustic events cumulative according to time displays an exponential evolution indicating a strong acoustic emission. The amplitude of the acoustic emission events shows a quasi triangular-shaped appearance over a range between 35 dB (average amplitude of 400 events) and about 85 dB indicating different damage mechanisms as matrix cracking, fiber/matrix debondings and pull-out (delamination is unlikely to occur in the case of non woven mat composite) and probably some fiber breakages of low and intermediate strength fibers. Moreover, some AE events of high amplitude (85-90 dB) are also revealed from Figure 3 suggesting fiber breakages of high strength fibers. The interface fiber/matrix is strong enough to transfer stresses from matrix to fiber.

After exposure to 23°C and 80%RH, few changes (Figure 2.b) can be noticed concerning both SEM and AE analysis (cumulative and distribution), which are consistent with the analysis of moisture absorption kinetics for this condition and with the mechanical properties evaluated by three point bending test. Indeed, the amount of water absorbed is low and acts only as a fiber plasticizer. Indeed, the stress at maximum, the modulus and the time at which the first acoustic signals appears (t=35s) were only slightly reduced.

At 70°C and 80% RH (Figure 2.c.), the aging conditions were more severe compared to that at 23°C. The acoustic signals occurred over 35 and 90 dB as in the previous cases. At a first approximation, it is possible to share the AE amplitudes into two distinct populations, the first one for the amplitudes ranging between 35 dB and 55 dB, and the second one for the amplitudes above 65 dB. The total number of events corresponding to the lower amplitudes is much greater than for the two previous cases while the number of acoustic signals having higher amplitude is smaller. Indeed, the amount of absorbed water is higher and fibers are then both plasticized and damaged and their strength greatly lowered. Premature fiber breakages are frequent increasing therefore low amplitude events. The fibers were easily extracted as fiber/matrix interfaces are becoming weaker. The threshold time (deflection) at which the first acoustic signal appears considerably lowered (22 s) and a high decrease in stress at break is observed. The SEM failure surface micrographs displays pull-out lengths smaller (Figure 2.c) than those observed on non-aged composites.

The very severely aged composite shows a different behavior during flexural tests than the other composites: reaching the maximum load, an abrupt load drop is observed accompanied by a strong acoustic emission (Figure 2.d), while failure appears in a progressive way in the case of unaged and less severe aged composite damages. Moreover AE amplitudes are mainly distributed between 35 dB and 60 dB (Figure 3) and the number of events greatly decreases with aging: This may suggest fiber/matrix interface weakening and serious degradation of the fibers by the water absorption finally causing the composite to fail catastrophically from the tensile side without any pull-out. The high level of fiber degradation is well highlighted by the presence of numerous debris on elementary and technical (set of pectin or lignin-bonded fibers) fibers (Figure 2.d).

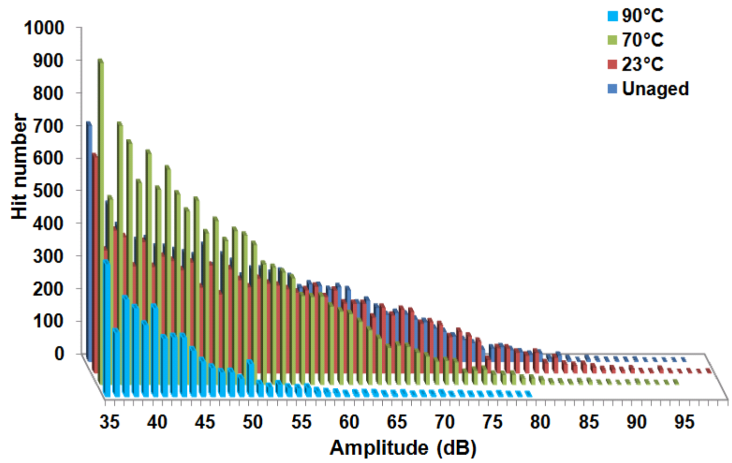

Figure 3. Acoustic emission amplitudes distributions for as received and aged NDF/PP composites.

CONCLUSIONN

AE is an efficient technique for in-situ health monitoring of natural fiber reinforced composite materials (NFC) subjected to serious environment. The acoustic analysis of hygrothermal aged NFC under flexural loading pointed out fiber/matrix interface weakening and degradation of the fibers by the water absorption finally causing the composite to fail catastrophically for the most severe aging conditions. These results are confirmed by SEM post-mortem analysis of failure features.

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