Mechanical Characterization of Untreated Waste Office Paper/Woven Jute Fabric Hybrid Reinforced Epoxy Composites

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ABSTRACT: The aim of this work is to assess the opportunity to use untreated waste office paper, alone and in combination with jute fabric, as a reinforcement in epoxy composites. Five different stacking sequences were manufactured and tested. Adding untreated waste office paper sheets has been revealed to increase both flexural and tensile strength of the neat resin and of the untreated jute fabric reinforced composites. The effect of the hybridization on tensile and flexural behavior has been evaluated through scanning electron microscopy observations and acoustic emission. The results confirm that waste office paper sheets can be used as a reinforcement for an epoxy resin, thus representing a viable alternative to paper recycling. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 119: 1366–1373, 2011

Key words: biofibers; composites; mechanical properties; acoustic emission; waste paper

INTRODUCTION

In recent years, the continuous and increasing demand for environmentally friendly materials has been proved to focus on biocomposites from plant-derived fibres and from recycled fibre based products. Natural fibres like flax, jute, hemp, banana, and sisal are emerging as realistic alternatives to glass fibres in various industrial sectors owing to their low cost, low specific weight which result in higher specific strength and stiffness, nonabrasiveness, abundant availability, biodegradability and problem-free disposal. Moreover, biocomposites are also claimed to offer environmental advantages such as reduced dependence on nonrenewable energy/material sources, lower pollutant emissions, lower greenhouse gas emissions, enhanced energy recovery, and end life biodegradability of components.[1–3] In particular, the use of recycled lignocellulosic fibres such as waste paper, waste wood, and agricultural residues in the development of composite materials, is attaining increased importance both in scientific and the industrial world where they offer a valid answer to maintaining a sustainable development of economical and ecological attractiveness, especially to the enormous quantity of waste paper/wood generated daily.[4,5]

According to the Green Press Initiative,[6] it has been estimated that, in the 2006 the United States newspaper industry consumed about 8.7 million metric tons of paper and 95 million trees, whilst each year ~ 30 million trees are used to make books marketed in the United States alone. As a consequence, a considerable amount of paper, mainly coming from newspapers, magazines, cardboards and office paper (about 32% of total municipal solid waste by volume) ends up in landfills. Hence, it is increasingly recognized to recycle newspapers and used papers. In particular, the United States is one of the world leaders in the recovery and recycling of newspaper, recycling 71.2% of the newsprint consumed in 2002.[7]

It is worth noting that the conventional recycling process of waste paper into new paper requires special treatments for the removal of inks, dye, size agents and binders, cleaning, refinement and reforming is more expensive and releases more carbon dioxide than manufacturing new paper.[8–10] In contrast, when the waste paper is used for manufacture of composites no extensive cleaning and refinement is required. The recycled paper reinforced composites consist of lignocellulosic fibres and other inorganic fillers, which contain printing inks and other process aid material. Furthermore, such composites can be processed similarly to wood-based composites.[11] Therefore, use of recycled paper as reinforcement for

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composites allows reducing notably the potential manufacturing cost, thus improving thermal and mechanical properties too.[12]

Such composites involve a large area of applications such as load bearing roof systems, subflooring, doors, windows, furniture, automotive, and interior parts.[5,12,13]

Several authors have investigated the properties and processing of recycled newspaper fibres as a possible reinforcement for thermoset[14,15] and thermoplastic polymer matrix[16–20] and the need of using various coupling agents to improve the interaction and the interface fibre-matrix owing to the incompatibility between the hydrophilic fibres and the hydrophobic polymers.[21–29]

In contrast, the literature on exploitation of waste untreated paper sheet forms is rather limited.[15,30]

In fact, whilst Yadav[30] et al. investigated the mechanical and water uptake properties of waste newspaper reinforced phenolic-epoxy matrix composites using waste newspaper sheets (at various weight fractions from 0.30 to 0.65) directly in the matrix without any pretreatment, Prud'homme[15] studied laminates of oriented and unoriented holocellulose paper, and of Whatman filter paper.

The aim of this article is to investigate the application of untreated waste office paper sheets (UWOPS) as a renewable natural reinforcement for epoxy composites. No treatments on paper sheets were applied. Moreover, UWOPS/untreated plain woven jute fabric (UPWJF) hybrid laminates have been investigated to characterize the effects of the UWOPS on mechanical properties (tensile and flexural strength) of the UPWJF. Different stacking sequences of UWOPS and UPWJF laminates were tested. The results presented in this article show that the UWOPS significantly improves the mechanical properties of the neat resin and of the UPWJF.

In addition, these results have been supported by scanning electron microscopy (SEM), which allowed characterizing fibre-matrix interface, and by acoustic emission (AE) analysis, which enabled investigating failure mechanisms.

MATERIALS AND METHODS

All the laminates were manufactured using the Hand Lay Up Process in an aluminium mould (250 mm × 250 mm). The composites were left to cure for 24 h at room temperature followed by 24 h-post curing at 60°C. The fibre weight fraction was controlled by using measured weights of fibre and matrix. The final weight fraction for all the configurations was 0.36 ± 0.02 whilst the final thickness was 3 (±0.1) mm.

UPWJF (hessian cloth: 300 g/m^2) and UWOPS (Fabriano: 80 g/m^2) were used as reinforcement in this work. In particular, no treatments on the paper sheets were applied. The resin used was a low viscosity epoxy system (SP Systems Ampreg 26) with a slow hardener (mixing ratio 100 : 33.3 by weight).

TABLE I
Summary of Laminate Configurations: (a) Jute Only (J); (b) Paper Only (P); (c) Sandwich Structure: Paper Core—Jute Skin (J/P/J); (d) Sandwich Structure: Jute Core—Paper Skin (P/J/P); (e) Paper Intercalated with Jute (P/J/P/J/P)

Sample series	Stacking sequence	Jute layers number	Paper layers number	Overall fibre content (wt%)
J	4 Jute	4	–	35.0 ± 2
P	18 Paper	–	18	37.5 ± 2
J/P/J	1J/8P/1J	2	8	36.6 ± 2
P/J/P	4P/2J/4P	2	8	37.2 ± 2
P/J/P/J/P	3P/1J/2P/1J/3P	2	8	35.5 ± 2

Five different stacking sequences were tested. The first configuration (J) contained 4 layers of jute fabrics. The second configuration (P) has 18 layers of recycled paper sheets. The third (J/P/J) and fourth (P/J/P) configurations were created as sandwich structures with paper layers as core and jute layers as skins and jute layers as core and paper layers as skins, respectively. At last, the fifth configuration (P/J/P/J/P) included jute layer intercalated with paper sheets. Furthermore, samples of neat resin were manufactured to allow a comparison with the aforementioned samples. The number of layers for the hybrid composites were chosen to maintain the same weight of the two types of reinforcement, namely UPWJF and UWOPS.

The five different stacking sequences of the laminates are summarized in Table I and Figure 1.

It was estimated a low content of voids (void content of 2% ±0.5) by digital image analysis. From the laminates were cut the tensile and three-point bend specimens having a length of 140 mm, a width of 20 mm and a thickness of 3 (±0.1) mm. The mechanical characterization was carried out by longitudinal tension test (ASTM D-3039) and three-point bending test (ASTM D-790) using an Instron 5584. Crosshead speeds of 1 mm/min and 2.5 mm/min were used for tension and bending tests, respectively. The span-to-depth ratio was 32 : 1. Five specimens for each type of stacking sequence were tested.

An AMSY-5 AE system by Vallen Systeme GmbH was used to detect AE signals during the mechanical tests. The threshold setting was 35 dB and the total gain was set at 34 dB. Four PZT sensors resonant at 150 kHz (Deci, type SE150-M) were used. Two sensors were placed on the surface of the specimens at a distance of 120 mm and 90 mm (for flexural and tensile tests, respectively) to allow linear localization and other two sensors were used as guard sensors. A scanning electron microscope Hitachi S-2500 was used to investigate the fracture surfaces of composites.

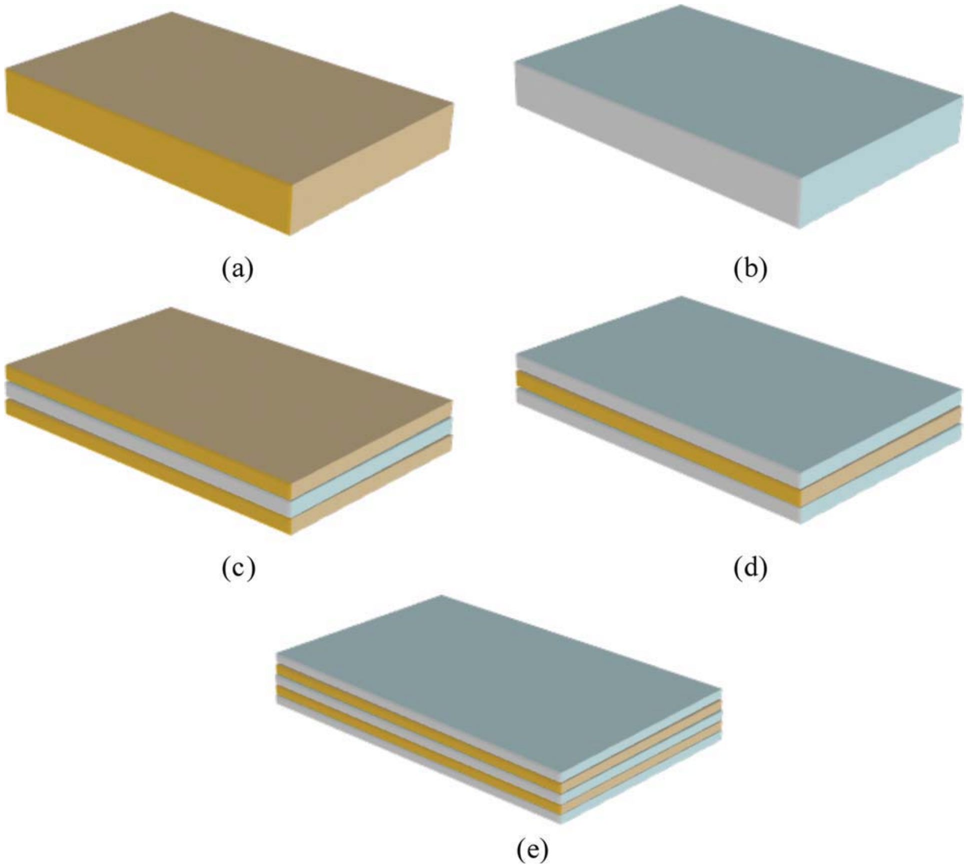

Figure 1 Sketch of the five configurations tested. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

RESULTS AND DISCUSSION

The results of the mechanical characterization are summarized in Figures 2–5 which show the strength and modulus for each type of test and laminate configuration tested (NE stands for neat epoxy). As a general result, the use of both reinforcements, namely jute fabric and paper sheets, resulted in a better flexural and tensile behavior compared with those of neat epoxy. In particular, as regards both tensile and flexural strength, the best performance is exhibited by the paper reinforced composite. In contrast, the J laminate configuration showed the lowest values for both strength and modulus in flexural and tensile test. It is to be noted the positive role played by the addition of paper sheets in jute reinforced composites: this is apparent when comparing

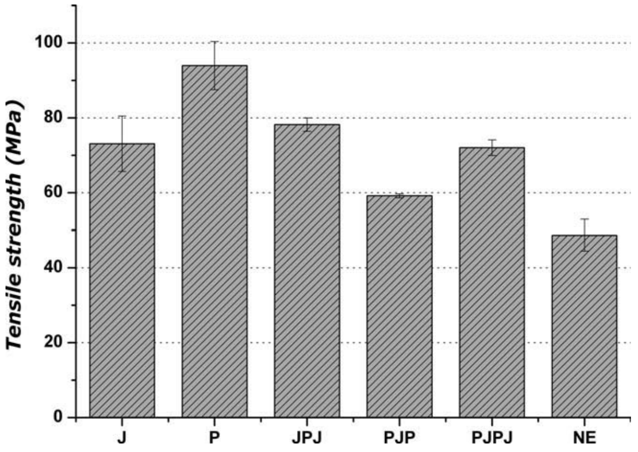

Figure 2 Average tensile strength for each laminate configuration tested.

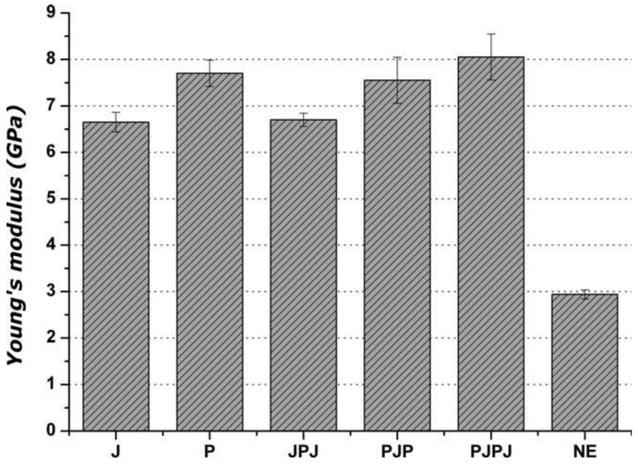

Figure 3 Average Young's modulus for each laminate configuration tested.

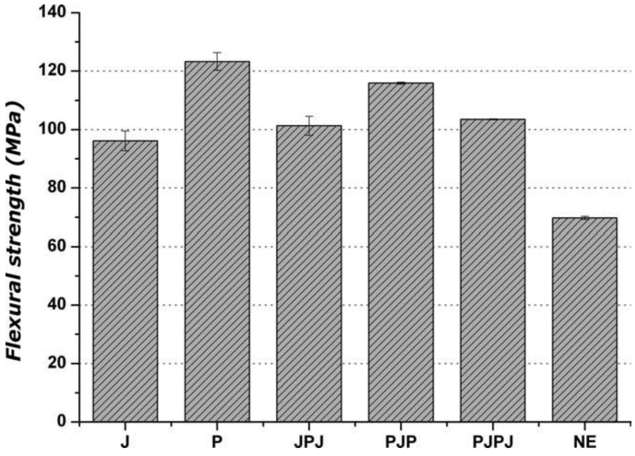

Figure 4 Average flexural strength for each laminate configuration tested.

the performances of the hybrid composites, which showed the highest flexural and tensile moduli. An increase in mechanical properties due to the addition of paper sheets was also found by Prud'-Homme,[15] resulting in a significant reinforcement of a PMMA matrix. As regards the flexural behavior, it can be noted that better performances are obtained when the paper sheets are placed as skins in the hybrid composites. This can be explained by the fact that in flexural testing, the outer layers are those subjected to higher stresses (compressive and tensile) and the presence of paper sheets well impregnated with resin can result in a resistance higher than that offered by jute layers. In fact, the composites belonging to the configuration JPJ are those having the lowest flexural strength. This is not surprising, because the jute fibres are untreated and it is well known the lack of compatibility between the lignocellulosic fibres and polymer matrices. In contrast, the paper sheets seem to be well impregnated by the resin and this improves the fibre-to-fibre bond strength and thus their strength.[15] Paper is an extremely complex biological material. A paper sheet consists of a two-dimensional array of fibres bound

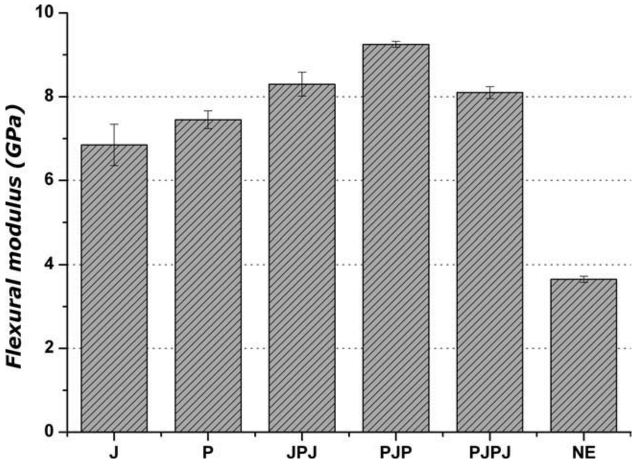

Figure 5 Average flexural modulus for each laminate configuration tested.

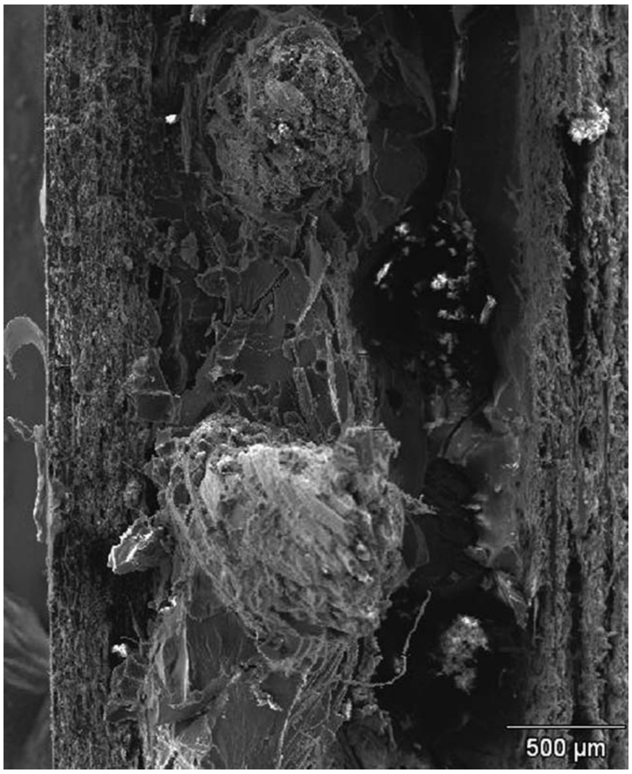

Figure 6 SEM micrograph showing the fracture surface of a PJP specimen failed in flexure.

Figure 7 SEM micrograph showing the fracture surface of a JPJ specimen failed in flexure.

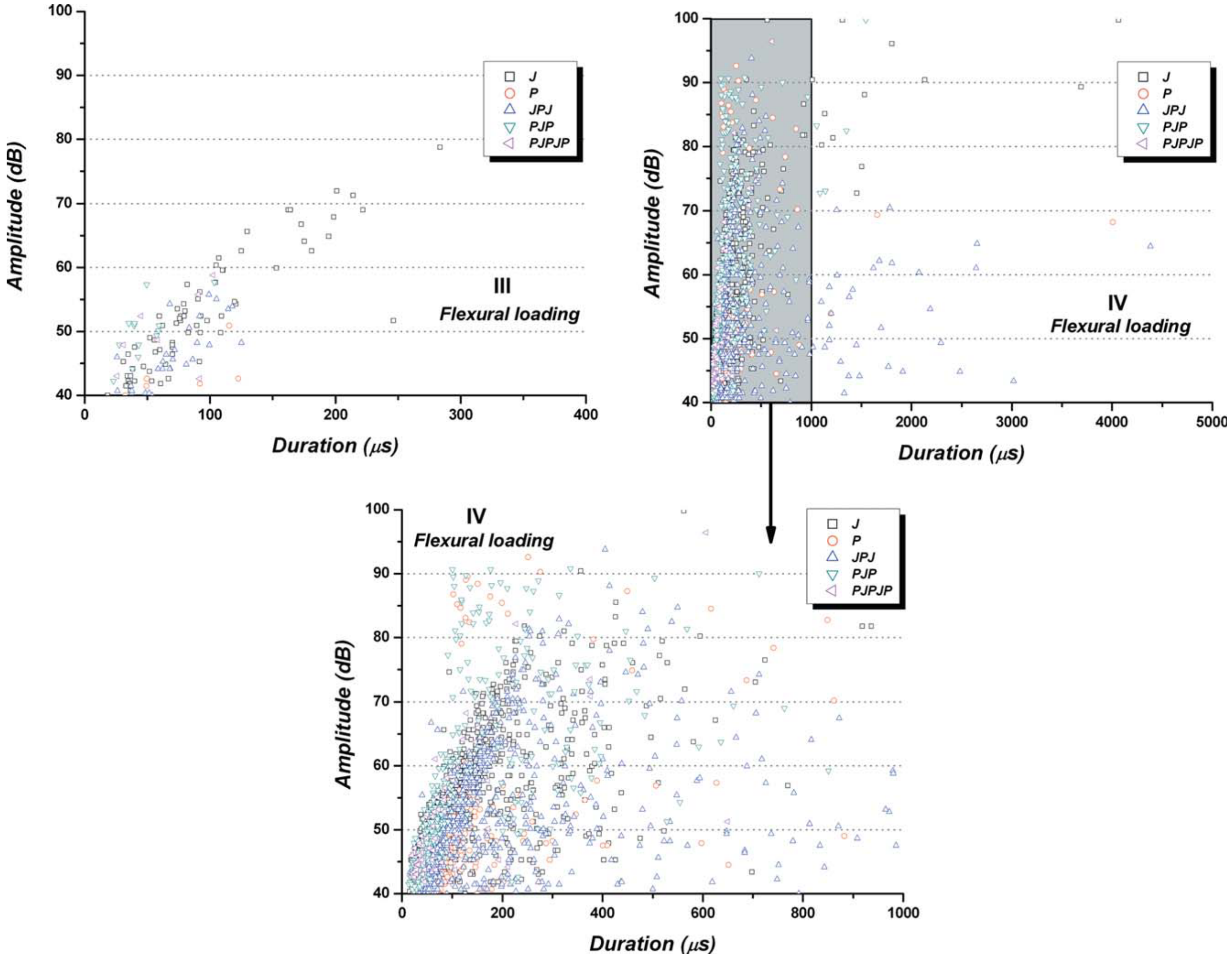

Figure 8 Acoustic emission signal amplitude vs duration during flexural loading. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

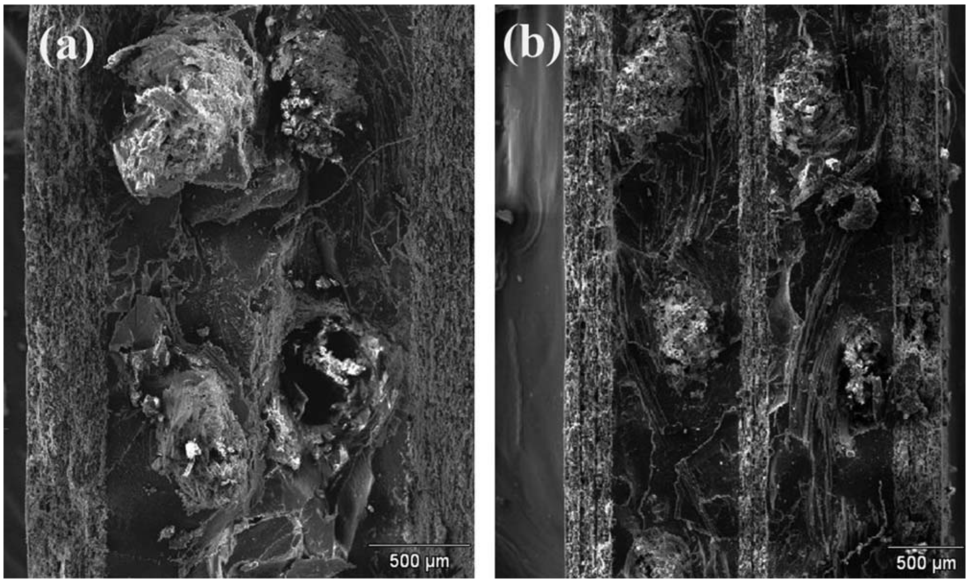

Figure 9 SEM micrographs showing the fracture surfaces of (a) PJP and (b) PJPJP specimens failed in tension.

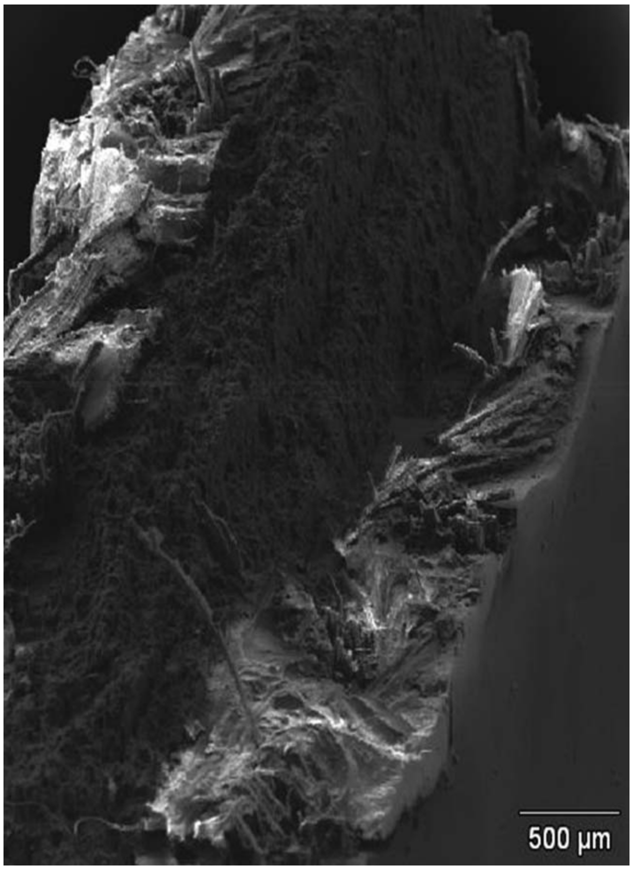

Figure 10 SEM micrograph showing the fracture surface of a JPJ specimen failed in tension.

together by hydrogen bonds at crossovers. The strength of a paper sheet derives from the strength of fibres and the number and strength of the fibre-fibre bonds. In addition, each fibre has well defined lamellar microstructure. Paper is, therefore, among the most complex engineering materials. The most important constituents of the final product are cellulosic fibres and the pore space formed by and in between the fibres.[31–33] This behavior is confirmed by the SEM images of fracture surfaces. In Figure 6 is shown the fracture surface of a PJP specimen failed in flexure. It can be seen that along with jute fibres pull-out, the paper sheets subjected to tensile stresses show some damage thus confirming the role played by the outer layers in determining the strength of the composite. In contrast, Figure 7 shows the fracture surface of a JPJ specimen failed in flexure: here, the failure is shown to occur at the jute/paper interface on the tensile side. In this case, the jute layers were not able to withstand the tensile load: this caused the failure at the interface and prevented the composite from attaining a high strength.

The use of AE to monitor the mechanical behavior of both paper and natural fibre composites had some coverage in literature already[34–36] In paper, two possible mechanisms causing AE have been identified: these are fibre failure and fibre bond failure, or a combination of these, the first being more energetic. In this work, a typical parametric analysis of AE signals has been performed, which is quite common in literature.[37,38] In particular, the correlation between signal amplitude and duration proved useful and its evolution during the loading was studied. Four load levels were identified, namely I = 0–25% $F_{\max}$, II = 25–50% $F_{\max}$, III = 50–75% $F_{\max}$, IV = 75–100% $F_{\max}$. The results are shown in Figure 8. During the first two intervals no significant AE signals were detected for all the configurations, whilst few signals for paper reinforced composites were detected during the whole loading. This is not surprising because even during AE monitoring of paper sheets most signals occur just before the load reached the maximum and signal attenuation is of concern.[36,39] In particular, stress–strain curves for paper can be typically divided in two parts: prefailure, which is almost elastic up to the maximum stress, and a tail, which arises due to the cohesive properties of paper.[35] For small strain rate, most of AE originates from tail. In this case, an enhanced limited mobility of lignocellulosic fibres due to the polymer could even prevent frictional pullouts of fibres from the network which usually account for signals at low loads. In the third load interval, most of signals are characterized by medium amplitude (45–60 dB) and low duration (up to 150 μs) which can be ascribed to matrix cracking and interface failures.[37,38] Only J laminates show signals characterized by higher amplitudes which is a sign of early damage. The occurrence of signals of high duration (more than 1000 μs) and low-medium amplitude (50–70 dB) due to delaminations is particularly evident for the JPJ specimens (also confirmed by SEM observations, Fig. 7). Signals of high amplitude and low duration, especially for the two configurations with more paper sheets arranged as outer layers (P and PJP), are to be ascribed to lignocellulosic fibre breakages of paper, which is a sign of the reinforcement effect provided by the paper sheets. For PJPJP configuration, most of AE signals originates from interface failures which limited the strength of the composite, likely due to presence of more jute/paper interfaces.

The explanation of the variation of tensile strength as a function of stacking sequence is somewhat complicated due to the lack of a clear trend. In fact, SEM images of the fracture surfaces (Fig. 9) show features which are quite common to all the configurations tested, particularly extensive jute fibres pull-out. An exception was represented by the JPJ configuration, where the fracture occurred in the more resistant paper core (Fig. 10) thus confirming the high strength shown in tension (Fig. 2). In this regard, the AE analysis can give an insight into the failure modes which occurred during testing. The results of the analysis are summarized in Figure 11. In the first

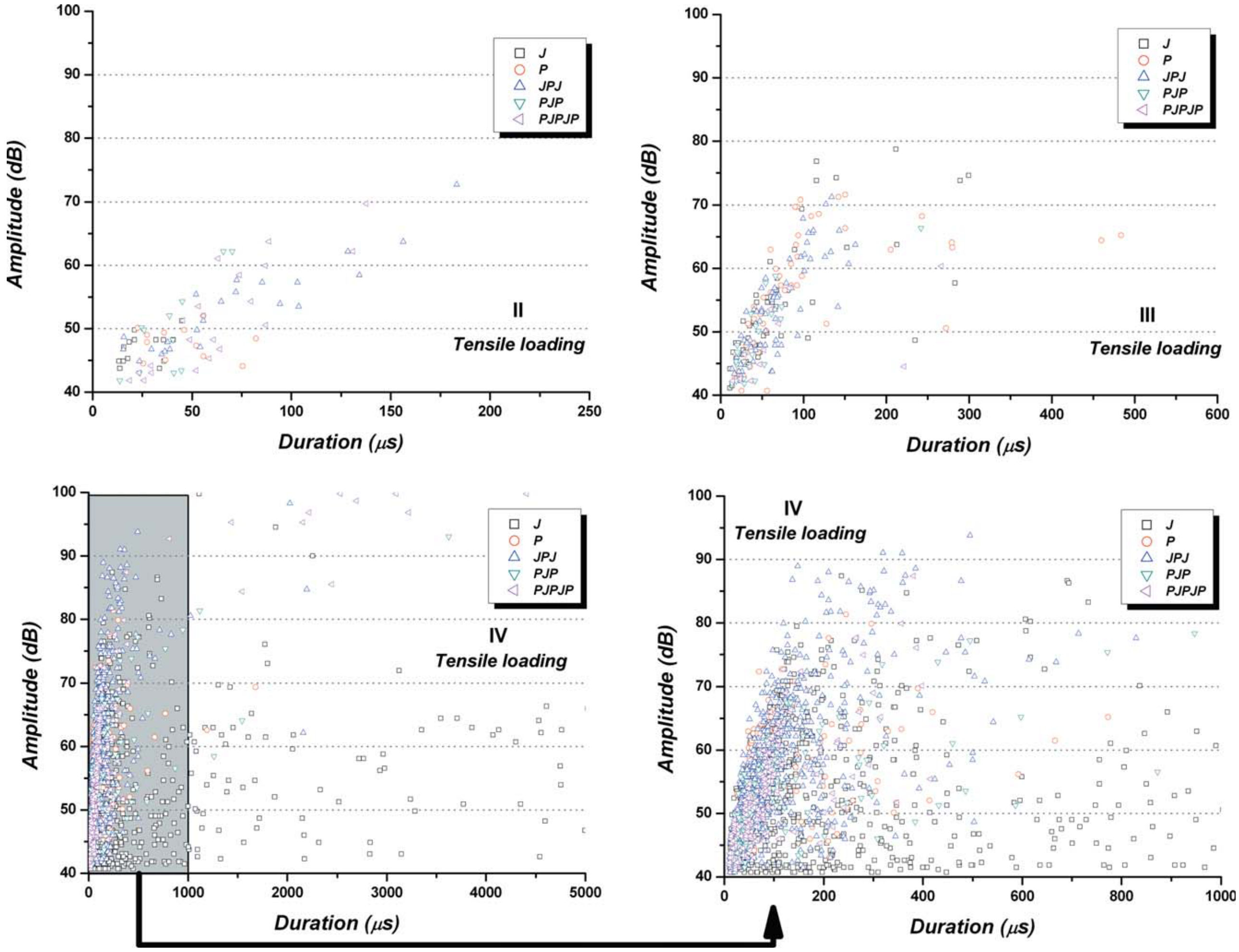

Figure 11 Acoustic emission signal amplitude vs. duration during tensile loading. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

loading interval (0–25%) no AE signals were detected. During the second interval (25–50%) few signals were detected for all the configurations tested. In particular, J specimens show signals characterized by low amplitudes (40–50 dB) and durations (15–100 μs) which are usually related to matrix cracking[37,38] which often leads to debonding. The same behavior is also shown by PJP and P specimens. The other two configurations, namely JPJ and PJPJP, are also characterized by signals (higher amplitudes) which are more suited to interface damage. However, interface failure is never isolated as a damage mechanism, but very often is linked to pull-out mechanisms. In the third loading interval, it is to be noted that, along with matrix cracking, an increase of the signal amplitudes occurred (which is particularly true for J and JPJ specimens). Higher durations are reported for P specimens which could underline the presence of some damage mechanism active at the paper/paper interface, which seems to be confirmed by the fracture surface (Fig. 12). It has been reported in literature that events characterized by high duration and low/intermediate amplitude are, in fact, often associated with delamination and debonding.[37] These are processes with both a longer physical duration and a longer acoustic duration. The fourth interval is marked by an extensive interface damage, which is clearly represented by the increase in duration of signals (particularly for J specimens). Signals having high duration (>1000 μs) and amplitude (80–95 dB) are due to fibre breakages which occurred close to final failure, especially for jute fibres which are bundles of many single fibres. For PJP specimens few signals of fibre breakage are present, thus pointing out that no full exploitation of reinforcement has occurred but mainly interface damage particularly due to the poor jute/epoxy interface. The reinforcement effect played by the paper core in JPJ specimens can be highlighted by signals characterized by high amplitudes but medium durations (250–400 μs) due to lignocellulosic fibre breakages.[37,38] It seems that paper sheets offer higher resistance when they are all grouped (P and PJP). In contrast, when there is a succession of resistant (paper) and less resistant (jute) layers, a decrease in strength is observed.

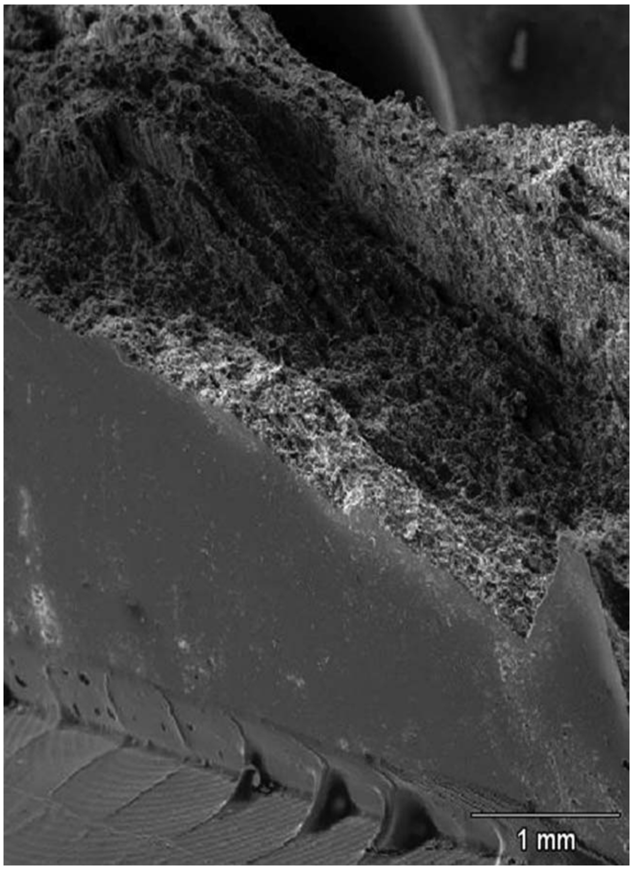

Figure 12 SEM micrograph showing the fracture surface of a P specimen failed in tension.

CONCLUSIONS

In this work, the effect of adding untreated waste office paper, alone and in combination with jute fabric, as a reinforcement in epoxy composites, has been evaluated. In particular, five different stacking sequences have been tested. The paper reinforced composites showed the best tensile and flexural strength, whilst the jute reinforced composites did not perform as well as paper ones. Better results were obtained through hybridization of jute with paper sheets. An attempt to clarify the role of the stacking sequences on the mechanical behavior has been performed through the use of AE. AE analysis, along with SEM observations, allowed gaining a better insight into the failure modes of the composites tested. The results confirm that the use of waste paper sheets as reinforcement in polymer composites shows potential and could offer an alternative opportunity to paper recycling.

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