Abstract

Cost-effective and reliable damage detection is crucial for the use of composite materials due to the relatively large number of failure mechanisms they can experience in service. Of the methods available, acoustic emission (AE) is considered as one of the most effective for on-line and in situ evaluation of structural health of composites. Thus, AE generated during three-point bending of aramid-reinforced epoxy composites was monitored by embedded and surface-mounted polyvinylidene-fluoride (PVDF) thin-film sensors instead of traditional piezoceramic ones. The parametric analysis of the AE signals detected and scanning electron microscopy observations enabled to verify the suitability of these sensors to be embedded as well as to characterize the failure mechanisms of the specimens tested. This work confirms the use of AE as an advanced, cost-effective, sensitive technique for detecting and locating potential damage mechanisms in composite materials.

Keywords: PVDF sensors, aramid-epoxy composites, three-point bending, structural health monitoring.

Introduction

Woven fabric reinforced composites are among the most important and widely used forms of textile structural composites due to their cost-effectiveness, high processability and enhanced out of plane mechanical properties. The increased use of such composites in the industry, from aerospace to construction, has required suitable and reliable testing procedures. Of primary importance in developing such procedures is the detection of damage initiation, progression and failure modes during loading of composites in real time. A host of nondestructive test methods have been developed to detect defects and damage in composite materials but AE monitoring is the primary technique used for the identification of different types of failure in composites for in service monitoring because it is a passive method, which only requires a network of receivers (Hamstad and Sendeckyj, 1993). In this regard, AE monitoring has some unique advantages over other NDT techniques: can detect the dynamic processes associated with the degradation of structural integrity, it is nondirectional and can be used in situ to monitor a structure while it remains in service. This feature makes AE a suitable sensing technique for structural health monitoring (SHM) (Speckmann and Henrich, 2004). Other unique capabilities include: high sensitivity, location of damage regions and sensitivity to any process that generates stress waves.

The solution analyzed in this work involved the use of polyvinylidene fluoride as sensor’s material and the AE as sensing technique. In particular, to fulfill the task concerning the in situ damage detection, the AE sensors have been embedded in the composite laminates during their manufacturing stage.
PVDF is a semicrystalline polymer, which owes its strong piezoelectricity to the strength of the electric dipole (between carbon and fluorine atoms) and to the large number of dipoles per unit volume, but most importantly because it crystallizes in a polar crystal phase characterized by a cooperative dipole alignment (Davis, 1987). PVDF differs in many respects from traditional piezoceramic materials. Ceramic transducers are in general brittle, stiff, easily damaged from mechanical shock or vibration and are difficult to machine in complex shapes. In contrast, polymeric transducers can be easily tailored to different shapes, are tough, flexible when machined into thin films thus not affecting the mechanical motion of the structures they are mounted on. Nevertheless, polymeric transducers can undergo mechanical relaxation phenomena and are prone to thermal depolarization and electromagnetic interference. At present, some of the most important applications of PVDF can be considered to involve pyroelectric, electromechanical and electroacoustic transduction (Sessler, 1981; Marcus, 1982; Bar-Cohen et al., 1996; Riande and Diaz-Calleja, 2004; Measurement Specialties, 2006).

Several papers are available in literature dealing with the use of PVDF as AE sensor (Stiffler and Henneke, 1983; Narisawa and Oba, 1984, 1985; Hamstad, 1995; Or et al., 2000; Kim et al., 2005; Park et al., 2005; Bar et al., 2004, 2005) even though as surface mounted AE sensors.

The aim of this work is to verify the suitability of PVDF sensors to be embedded and their influence on the flexural behavior and integrity of the resulting composite laminates. Furthermore, the AE monitoring of composites during loading and the characterization of AE signals acquired were addressed to identify the failure mechanisms involved.

Materials

The specimens tested in this work were obtained from square aramid/epoxy panels (250 x 250 mm) made using RA175H4 fabric and a SP106 resin, both from SP Systems. The areal weight of the fabric, a 4-harness satin weave containing Kevlar49 fibers, was 170 g/m². The samples were manufactured by hand lay-up in a closed mould using ten layers of fabric. This manufacturing technique made the embedment of PVDF sensors easier. In fact, during the layer stacking process, PVDF sensors were arranged in the middle of the stacking sequence of the laminate. The thickness of the cured laminates was 3 ± 0.2 mm and the fiber volume fraction was 0.38 ± 0.02 (as calculated from the perform weight per unit area). From the panels, rectangular specimens having a length of 200 ± 5 mm and a width of 30 ± 0.5 mm were removed and subjected to three-point bending tests. Two different types of specimens were produced: the first type (labeled as type A) had no embedded sensors (served as reference material) whilst the second one (labeled as type B) had two embedded sensors.

PVDF sensors used in this work were the DT1-052K manufactured by Measurement Specialties Inc. These sensors are rectangular element piezo-film with silver ink screen printed electrodes and are supplied with a thin urethane coating over the active sensor area; the lead attachment legs are free of this coating (Measurement Specialties, 2006). In Table 1 are reported some typical properties of the piezo-film and the geometrical dimensions of the sensors used in this work (Measurement Specialties, 2006). The PVDF sensors for type A specimens were surface mounted using a double-side tape. A reliable interconnection to piezo-film was made through the use of a conductive silver epoxy resin manufactured by ITW Chemtronics (CW 2400), which provided not only excellent electrical conductivity but also high-strength bonding.
Table 1. Typical properties of piezo film and dimensions of PVDF sensors used.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Parameter</th>
<th>PVDF</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_{31}$</td>
<td>Piezo Strain Constant</td>
<td>23</td>
<td>$10^{-12} \text{(m/m)/(V/m)}$</td>
</tr>
<tr>
<td>$d_{33}$</td>
<td>Piezo Strain Constant</td>
<td>-33</td>
<td>$10^{-12} \text{(m/m)/(V/m)}$</td>
</tr>
<tr>
<td>$g_{31}$</td>
<td>Piezo Stress Constant</td>
<td>216</td>
<td>$10^{-3} \text{(V/m)/(N/m^2)}$</td>
</tr>
<tr>
<td>$g_{33}$</td>
<td>Piezo Stress Constant</td>
<td>-330</td>
<td>$10^{-3} \text{(V/m)/(N/m^2)}$</td>
</tr>
<tr>
<td>$k_{31}$</td>
<td>Electromechanical Coupling Factor</td>
<td>12%</td>
<td></td>
</tr>
<tr>
<td>$k_{0}$</td>
<td>Electromechanical Coupling Factor</td>
<td>14%</td>
<td></td>
</tr>
<tr>
<td>$Y$</td>
<td>Young’s Modulus</td>
<td>2-4</td>
<td>$10^{9} \text{N/m^2}$</td>
</tr>
<tr>
<td>$p$</td>
<td>Pyroelectric coefficient</td>
<td>30</td>
<td>$10^{-6} \text{C/m^2 K}$</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>Permittivity</td>
<td>106-113</td>
<td>$10^{-12} \text{F/m}$</td>
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<tr>
<td>$\varepsilon/\varepsilon_0$</td>
<td>Relative Permittivity</td>
<td>12-13</td>
<td></td>
</tr>
<tr>
<td>$\rho_m$</td>
<td>Mass Density</td>
<td>1.78</td>
<td>$10^{3} \text{kg/m^3}$</td>
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<tr>
<td>$T$</td>
<td>Temperature Range</td>
<td>-40 to 80...100</td>
<td>°C</td>
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<table>
<thead>
<tr>
<th>PVDF Sensor</th>
<th>Length (mm)</th>
<th>Width (mm)</th>
<th>Thickness (μm)</th>
</tr>
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<tbody>
<tr>
<td>DT1-052K</td>
<td>41</td>
<td>16</td>
<td>70</td>
</tr>
</tbody>
</table>

Methods

Before performing flexural tests, the wave velocity of AE signals in the laminates was measured in order to make possible the localization of AE events during the tests. The procedure used is described elsewhere (Caneva et al., 2007). The value obtained was 4600 m/s, which should be considered as an approximate value due to the highly anisotropic nature of composite materials. Its accuracy was checked by means of calibration tests through the Hsu-Nielsen source method (Caneva et al., 2007) providing satisfactory results.

Fig. 1. SEM micrograph showing the PVDF sensor embedded in aramid reinforced epoxy.

Fig. 2. SEM micrograph showing details of the sensor/matrix interface of an aramid reinforced laminate.
After these preliminary tests, the specimens were subjected to three-point bending tests in accordance with ASTM D-790 (three-point loading). These tests were performed in an Instron 5584 at a constant crosshead speed of 2.5 mm/min with a span-to-thickness ratio of 20:1. The strain at the mid-span was determined by means of strain gauges. In order to embed PVDF sensors, width and length ratios were selected to limit edge and large inclusion effects. The PVDF sensors were located within the span length with a center-to-center distance of about 60 mm. The position of sensors was selected in order to have them close to the stressed area during bending with the aim of enabling the detection of AE signals as well as verifying their influence on the mechanical response of the composite laminates. Ten specimens were tested for each composite system (types A and B). The tests were monitored by two PVDF sensors and the signals were acquired by an AMSY-5 AE system manufactured by Vallen Systeme GmbH. The AE acquisition settings were as follows: threshold = 40 dB (ref. 0 dB = 1 μV), rearm time = 0.4 ms, duration discrimination time = 0.2 ms and total gain = 34 dB. Attenuation of AE waves was measured using the Hsu-Nielsen source procedure. It was small enough to prevent from doing any correction of the measured amplitudes owing to the small distance between the sensors. In linear location calculations, the position of the sensor is assumed coincident with the center of the sensor itself. Only AE signals localized between the sensors have been considered in the off-line parametric analysis.

The failure modes and microstructures in the composites were investigated with scanning electron microscopy (SEM).

**Results and Discussion**

A major concern during the laminate cure cycle was with the health of the embedded sensors. The maximum temperature of 75°C during fabrication of laminates was very close to the Curie transition temperature for PVDF sensors, which could result in depoling associated with a sharp decrease of piezoelectric properties. In addition, residual stresses at the matrix/sensor interface could affect the ability of the sensor to detect AE events. To investigate these aspects, SEM analysis was performed on composite laminates. The results of this investigation are summarized in the following micrographs (Figs. 1-3), which refer to the cross section of the specimens. The cross sections were graphite coated before SEM observations.

As for embedded PVDF sensors, as it can be seen in Figs. 1-3, the interface between the sensor and the surrounding matrix represents a region of continuity between the different materials and does not seem to be present any kind of remarkable damage able to affect in a negative way the piezoelectric behavior of the sensor itself. In fact, especially from Fig. 3, there are no defects such as microcracks due to different thermal expansion coefficients of the metal and the polymer at the interface and the metallization adheres well to the matrix, thus enhancing the ability of the matrix to transfer the AE signals to the sensor. These observations confirm the ability and the easiness of PVDF sensors to be embedded.

Even though the future of smart structure technology seems promising, its application to engineering systems cannot be accomplished until its integrity issues are fully understood. As sensors are integrated into complex structures, questions arise on the mechanical behavior and integrity of the structure. Therefore, the first step in the development of a reliable damage detection system based on AE technique has to be an investigation of the mechanical properties of the resulting structures.
A comparison between typical flexural load-displacement curves of $A$ and $B$ specimens is shown in Fig. 4. As it can be noticed, the behavior of specimens with and without embedded PVDF sensors is almost identical. The failure of aramid-reinforced epoxy composites in three-point bending can occur by either one of two modes being failure by longitudinal fracture of fibers in the tensile side or by shear delamination. The transition between these failure modes is controlled by the fiber volume fraction (Davidovitz et al., 1984). The curve of the fiber fracture mode is characterized by a yielding stage and a longer ultimate deflection whilst the curve for delamination mode is characterized by a main delamination event followed by a number of secondary delamination. Furthermore, the span length was long enough so that the specimen does not fail in shear and short enough to neglect the effects of large deformations. The composite specimens tested exhibited curves characterized by a yielding stage and a long ultimate deflection, during which the composites failed gradually, thus indicating an increased energy absorption and a good damage tolerance. The flexure curve is strongly non-linear and this behavior is consistent with the non-linear compressive behavior of composites reinforced with aramid fibers (Zweben, 1978; Mittelman and Roman, 1990).

In this case, it can be seen that the failure of both sides, namely tension and compression, may occur during flexural loading. This is due to the fact that Kevlar fibers are characterized by poor transverse and shear properties thus providing a composite, which shows poor strength in compression. This aspect can be enhanced by fiber crimps, which tend to lower the composite compressive strength since they make easier the appearance of kinking. This is not properly the case because in satin-woven fabrics, the crimp ratio is generally smaller if compared, for instance, with plain-woven fabrics. Nevertheless, some kink bands appeared in the specimens tested, as it will be explained later but the catastrophic failure was localized in the outer ply on the tensile side. The interface plays an important role in the behavior of these composites. For aramid/epoxy composites, the interface is thought to be a particularly weak link (Penn and Liao, 1984).

As regards the effect of PVDF sensors on the mechanical properties of composites, the two specimen types exhibited a similar three-point bending strength and flexural modulus (within 3% of each other in both cases). Therefore, from the results it can be inferred that the embedment
of PVDF sensors does not seem to have any remarkable influence on the damage evolution during three-point bending (Figs. 5-6) in spite of the test conditions’ severity, since the sensor embedded takes up most of the specimen’s cross section.

Fig. 5. Average three-point bending strength for specimen types A and B.

Fig. 6. Average three-point bending modulus for specimen types A and B.

Final failure of composites is considered to be due to the accumulation of different failure modes such as matrix cracking, debonding and fiber breakage. The peak-amplitude distribution (PAD) of AE signals can be successfully used to characterize the different damage mechanisms in composite materials since it depends strongly on the material as well as on the deformation mechanism present thus providing insight into how the structure behaves under a given loading condition. In this work, AE events generated during three-point bending were recorded until final fracture occurred. Figure 7 shows typical amplitude distribution plot for specimens A. The same curve for specimens B is shown in Fig. 8. As it can be noticed, typical load-displacement curves and AE activity patterns for both types are almost identical. In order to better understand the failure of these composites and to verify whether or not embedded PVDF sensors are able to follow on line the structural changes occurring, the cumulative event distributions have been

Fig. 7. Load and amplitude distribution for A specimen.
Fig. 8. Load and amplitude distribution for B specimen.

Fig. 9. Typical load and cumulative AE events versus time for specimen B.

divided in different intervals because, during the flexural tests, the testing machine was periodically stopped and the specimens examined by SEM so as to correlate the different PADs detected to the actual damage modes and progression (Fig. 9). The PADs corresponding to the different intervals labelled 1°-5° in Fig. 9 are shown in Fig. 10. During the first part of the loading (intervals 1 and 2), the PAD shows that events are generally situated in the range of 43-50 dB, which in literature is, in this condition, well documented as related to matrix microcracking and matrix-matrix friction. They also show some events around 55-63 dB characterizing the interface de-adhesion. This behavior can be explained referring to the low strength of the aramid/epoxy inter-
Fig. 10. Evolution of a typical PAD during flexural loading for specimen B.

Fig. 11. SEM micrograph showing the cross section of a B specimen in the as-manufactured stage.

Fig. 12. SEM micrograph from flexural tests showing longitudinal cross section at the mid span at the end of interval 2.

face, while fiber failure seems to be negligible in this stage (no significant events characterized by high amplitude values). These sensors seems to be insensitive to interfacial debonding due to fiber straightening, which is supposed to happen at the onset of loading. In fact, during the very first part of the loading, few signals or even no signals were detected. This behavior seems to contradict the experimental evidence that in composite materials AE is noted from the beginning of loading. This implies that a damage-producing mechanism should be already active at low stress levels. Since both matrix and fibers are not active at such low stress levels, it is reasonable to assume that some damage develops at the interface between fiber and matrix. Two different mechanisms can account for the occurrence of this kind of damage. The first is related to a fiber straightening process, during which the strains produced are higher than the failure strain of the
matrix, thus resulting in interfacial debonding. The second is due to residual stresses at the interface developed during the manufacturing stage of the composites as a result of different coefficients of thermal expansion. The SEM observations agree with what is suggested by the PAD, as it can be seen in Fig. 12, where de-adhesion in the warp fiber bundle and small matrix cracks can be seen. In order to make a comparison, Fig. 11 shows a SEM micrograph of a composite in the as-manufactured stage.

Fig. 13. SEM micrograph from flexural tests showing longitudinal cross section at the mid span at the end of interval 3.

Fig. 14. SEM micrograph showing cracks in the weft fiber bundle.

Fig. 15. SEM micrograph showing some fiber kinking.

Fig. 16. SEM micrograph from flexural tests showing longitudinal cross section at the mid span after final failure occurred.
The stage that leads to the first drop in the load, is characterized by a large number of events situated in the amplitude range 40-65 dB, characterizing the growth of transverse cracks in weft fiber bundle and some delaminations (see Figs. 13 and 14), and events with high amplitudes (80-90 dB), which belong to fiber fracture located mainly in the outer ply on the tensile side. Fiber fracture occurs late during the loading due to the small scatter of aramid fiber strength. This behavior is strongly dependent on the type of the weaving structure used. In this stage, the tensile failure is the dominant failure mode even though micro-damage and fiber kinking were observed on the compression side of the specimens (Fig. 15).

As loading continues, the corresponding failure is characterized by the growth and accumulation of the micro-damages discussed above, as PAD and SEM observations confirm. Figure 16 shows the longitudinal cross section at the mid-span after final failure occurred. It is evident the accumulation of the several damage mechanisms discussed so far. These results are also con-
firmed by the duration distribution of the AE events, as can be seen in Fig. 17, which shows the evolution of the duration distribution for a $B$ specimen (the intervals being the same as Fig. 9). As loading continues, most of the signals are located at 0-200 $\mu$s, which are values typical of matrix cracking while longer durations begin to be significant during the third interval and up to final fracture (delamination and debonding), where the occurrence of even longer durations should be attributed to fiber ruptures.

The embedded PVDF sensors were also able to localize the AE events (see Fig. 18). Due to the type of loading, most of the signals are located at the center of the specimen even if the AE signals seem to be more spread around the point of the maximum applied load, probably due to the enhanced toughness of Kevlar fibers.

**Conclusions**

Results from this work indicate that the embedment of PVDF sensors in aramid/epoxy laminates affects negligibly the mechanical response of reference materials. In fact, the two specimen types ($A$ and $B$) exhibited very similar three point bending strength and flexural modulus (within 3% of each other in both cases). The mechanical characterization was preceded by SEM analysis, which enabled to confirm both the possibility of successfully embedding PVDF sensors in composite laminates and the sensor’s ability to withstand the composite curing temperature without experiencing significant degradations (especially depoling). The mechanical tests were monitored on-line through the use of AE. A parametric off-line analysis of the signals detected showed that different failure mechanisms have different characteristics, which can be properly studied and identified using the AE responses. Through these investigations it can be concluded that PVDF film sensors can be effectively used as acoustic emission sensors for a SHM system. However, a more in-depth investigation of AE through the use of techniques such as statistical pattern recognition is necessary. These results are nonetheless promising in view of the development of a reliable, low cost, effective and in situ damage monitoring system.

**References**


C. Caneva, I.M. De Rosa, F. Sarasini (2007), Strain, accepted for publication.


M.A. Marcus (1982), Ferroelectrics, 40, 29-41.


A. Mittelman and I. Roman (1990), Composites, 21, 63-69.


