Development and Characterization of Piezoelectric Particle-Enhanced Self-Sensing Hybrid Composites

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Keywords: *self-sensing; smart composite; hybrid composite; piezoelectric materials; structural health monitoring*

1. Abstract

Self-sensing multifunctional polymer composites were fabricated using zinc oxide particles and copper-mesh electrodes. Glass and carbon fabrics were used as reinforcing materials in different laminate configurations. Due to the piezoelectric properties of the zinc oxide filler, an electrical voltage was generated under flexural loading, enabling the simultaneous realization of mechanical load-bearing and self-sensing functions. Based on the electromechanical tests, the hybrid composites containing carbon fabric produced a higher piezoelectric response at the same deformation level. The carbon fabric layers may have contributed to the collection and transfer of the generated electrical signal, thereby improving the sensing efficiency of the composites. During interlaminar shear strength testing, a mixed failure mode caused by both shear and tensile stresses was observed in the composites reinforced only with glass fabric. In contrast, damage in the hybrid composites mainly occurred as shear-induced delamination propagating along the laminate interfaces.

2. Introduction

Nowadays, the further development of conventional structural composites with favorable mechanical properties has led to a growing demand for multifunctional and so-called smart composites [1]. These materials are not limited to load-bearing applications; they may also be capable of detecting various environmental and mechanical influences and transmitting the corresponding response signals. As a result, the specific performance of composite structures can be further improved, their structural efficiency can be enhanced, and real-time monitoring of structural condition may also become possible [2].

One promising approach is the development of self-sensing composites, which are becoming increasingly important in modern engineering structures, particularly in applications where safety and reliability are critical requirements. Continuous monitoring of structural integrity and loading conditions can contribute to predicting failure processes in fiber-reinforced polymer composites, whose damage and failure behavior are often difficult to predict [3]. The use of composites with integrated sensing functionality enables the structure itself to provide information about its loading state, deformation, or damage evolution [4, 5]. One possible way to achieve self-sensing functionality is to incorporate piezoelectric particles into the composite matrix. This approach can be particularly advantageous, as the sensing function is not introduced as a separate sensor but is integrated directly into the material architecture. At the same time, the incorporation of functional fillers, conductive layers used as electrodes, and different reinforcing fabrics may alter the internal structure and interlaminar behavior of the composite. Therefore, during the development of self-sensing capability, not only the magnitude of the electrical response but also the preservation of mechanical integrity must be considered as a key aspect [6].

A particularly important aspect in the case of hybrid glass- and carbon-fiber laminates is how layers with different mechanical and electrical properties influence load transfer, damage mechanisms, and the measurable sensing signal [7, 8]. The electrically insulating nature of the glass fabric may be advantageous for measuring the piezoelectric response [9], while the greater stiffness and conductivity of the carbon fabric may affect both the structural behavior and the efficiency of signal collection [10]. In addition, the presence of the copper mesh incorporated as electrodes creates additional interfaces that can influence both the interlaminar shear strength and the failure mode.

The aim of this study was to develop and characterize self-sensing hybrid composites containing piezoelectric ZnO particles. The investigated hybrid composites were based on glass- and carbon-fiber reinforcements combined with an unsaturated polyester matrix, into which piezoelectric particles were incorporated to enable the measurement of the electrical response generated under mechanical loading. The developed composites were evaluated from two main perspectives. First, their mechanical performance was assessed by short-beam testing, focusing on interlaminar shear strength and failure modes. Second, their self-sensing behavior was analyzed using force–voltage and deformation–voltage relationships. These two complementary approaches enabled the evaluation of how piezoelectric ZnO particles, copper-mesh electrodes, and hybrid laminate configurations consisting of glass and carbon fabrics collectively affect the mechanical integrity and sensing performance of the composites.

3. Materials and methods

3.1. Materials and manufacturing

In our study, we tested three different types of self-sensing composites (Type A, Type B, Type C), as shown in Figure 1.

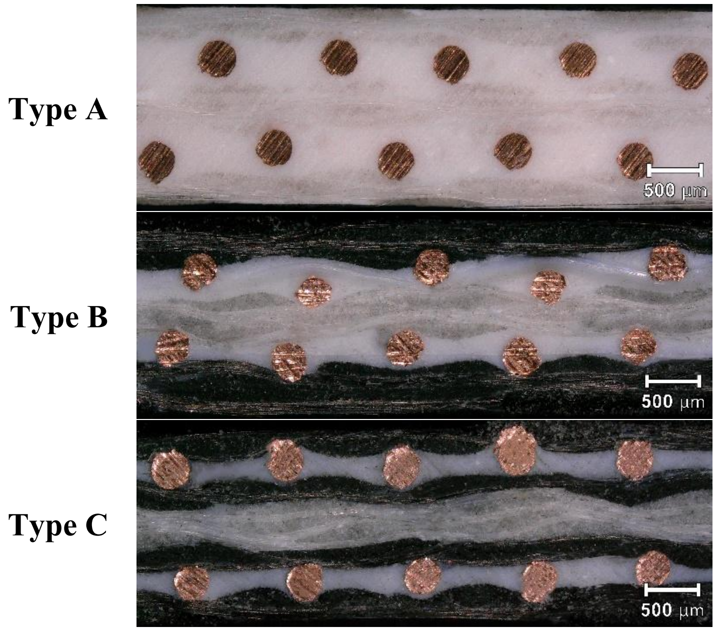

Figure 1. Types of the tested polymer composites. Their structure:
Type A: 2 layers of GF – copper mesh – 3 layers of GF – copper mesh – 2 layers of GF,
Type B: 2 layers of CF – copper mesh – 3 layers of GF – copper mesh – 2 layers of CF,
Type C: 1 layer of CF – copper mesh – 1 layer of CF – 3 layers of GF – 1 layer of CF – copper mesh – 1 layer of CF.

The glass fabric (GF, UNIQUE Textiles, s.r.o., Czech Republic) used was plain-woven with an areal weight of 220 g/m². The carbon fabric (CF, SGL Technologies GmbH, Germany) was plain-woven with an areal weight of 200 g/m². The copper meshes (Copper Age Kft., Hungary) were made of high-purity (over 99%) copper with a mesh density of 1 mm and a thickness of 0.5 mm. Before lamination, the surface of the copper meshes was treated with a 10% citric acid bath (Parma Produkt Kft., Hungary) at 80°C for 30 minutes. The piezoelectric self-sensing function was provided by zinc oxide (ZnO, Molar Chemicals Kft., Hungary) particles, which were mixed with the resin at a concentration of 20 m/m% based on our preliminary results. The composite matrix was Distitron 5119 ESX20ZQ unsaturated polyester resin (UPR, Polynt Composites Poland Sp. z o.o., Poland), which was used with Peroxan ME 50 LX (PERGAN GmbH, Germany) as a crosslinking agent at a 100:1 ratio.

During the fabrication of the composites, we first dispersed the ZnO particles in the UPR by mechanical stirring for 10 minutes, then by ultrasonic stirring for 5 minutes, using a Bandelin Sonopuls 4200 HD UH ultrasonic homogenizer (BANDELIN electronic GmbH & Co. KG, Germany) at 20 kHz, and 100 W of power. We then prepared the composites by hand lamination and cured them using a T30-type hot press machine (Metal Fluid Engineering s.r.l., Italy). First, we cross-linked them at 80°C under 100 bar hydraulic pressure (25 bar on the composite sheet) for 1 hour, then post-cured them in a drying oven at 100°C for 1 hour. The cured composites were cut to 30 mm x 100 mm x 2 mm for self-sensing function tests and to 20 mm x 10 mm x 2 mm for short-beam tests using a Mutronic Diadisc 4200 diamond-tipped precision saw.

3.2. *Testing and characterization*

For testing the self-sensing functions, the test specimens were bent using a Zwick Z005 (Zwick GmbH & Co. KG, Germany) tensile tester machine in a three-point configuration. We tested 5 specimens per sample with an 80 mm support distance, loading and unloading over 10 cycles at a test speed of 100 mm/min. During loading, the electrical voltage generated by the composite was recorded at room temperature using an Agilent 34970A (Agilent Technologies, USA) digital multimeter data logger, with a sampling time of 300 ms.

We used a Keyence VHX-5000 optical microscope (Keyence Corporation, Belgium) for the visual analysis of the composites.

Short beam tests were performed in accordance with ISO 14130 on 5 test specimens per sample using a Zwick Z005 (Zwick GmbH & Co. KG, Germany) tensile tester. The interlaminar shear strength (ILSS) of the samples was determined based on (1):

$$\tau = \frac{3}{4} \cdot \frac{F}{b \cdot h} \tag{1}$$

where τ is the interlaminar shear strength, F is the maximum force at failure, b is the thickness of the test specimen, and h is the width of the test specimen.

4. Results and discussion

4.1. Testing of self-sensing composites

During testing of the self-sensing functions, we defined five load levels (50, 75, 100, 125, and 150 N) that were applied equally across all composite types. The resulting force-voltage and strain-voltage curves are shown in Figure 2.

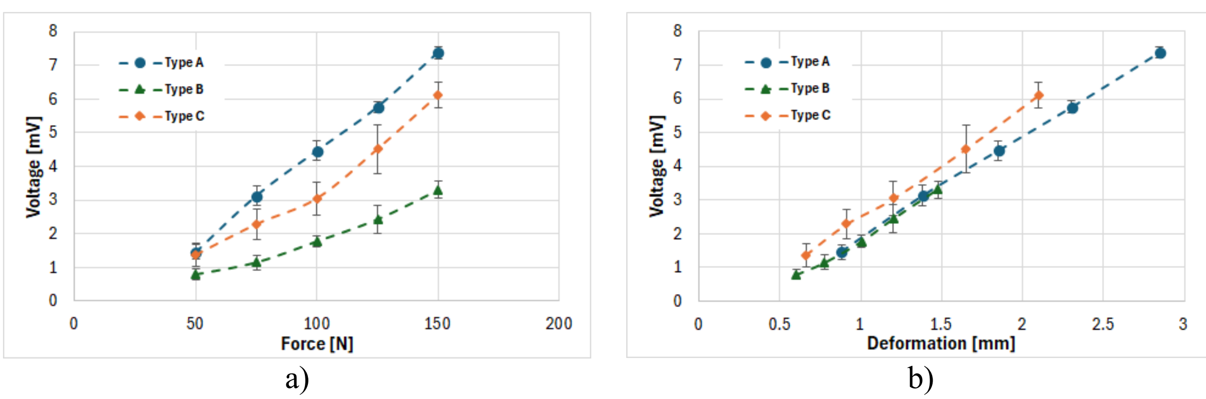

Figure 2. Testing of self-sensing functions: a) stress-voltage curves and b) deformation-voltage curves of composites.

According to the force-voltage curves (Figure 2a), we noticed that all three types of composites had nearly linear electrical sensitivity within the tested force range. The largest signal response to the load was observed for Type A, which is a composite containing only glass fabric. The smallest signal value was observed for the Type B composite under the same force values. However, it is important to note that a direct comparison of these results may provide a misleading picture, as the composites containing carbon fabric had higher flexural stiffness; therefore, it is advisable to evaluate the results in terms of strain as well. As a solution to this, we also examined the stress-voltage curves (Figure 2b). These results allow for a comparison that is less distorted by mechanical behavior and more relevant from the perspective of sensor response. It can be seen that the strain-voltage sensitivity of Type A and Type B composites has nearly identical characteristics, which is because the piezoelectric particles dispersed in the matrix primarily generate a measurable electrical signal between the copper mesh serving as the two electrodes. Type C produced better results than Type A and Type B. This can be explained by the fact that, although to a minimal amount, the carbon fibers also participated in the collection and transfer of the electrical signal, thereby improving the efficiency of charge collection and signal transfer.

4.2. Mechanical Properties of Composites

The aim of the short-beam test was to compare the interlaminar shear strengths; the results are shown in Table 1.

Table 1. ILSS Results for Composites.

Composite name	ILSS (MPa)
Type A	9.8 ± 0.6
Type B	6.8 ± 0.8
Type C	6.2 ± 0.4

Based on the results, the Type A specimen had the highest interlaminar shear strength. This can be explained by the fact that, in this laminate, the copper mesh was located only between glass fabric layers, resulting in a more homogeneous layer structure. In the case of the Type B and Type C test specimens, the addition of carbon fabric layers resulted in lower ILSS values. This is due to the differing stiffness and deformation behavior of the CF and GF layers, which caused stress concentration at the layer boundaries, thereby promoting interlaminar delamination. The lowest value was obtained for the Type C sample, suggesting that, in this layer arrangement, the positioning of the carbon fabric and copper mesh was less favorable for interlaminar shear stress.

The force-deformation curves measured during the ILSS test are shown in Figure 3.

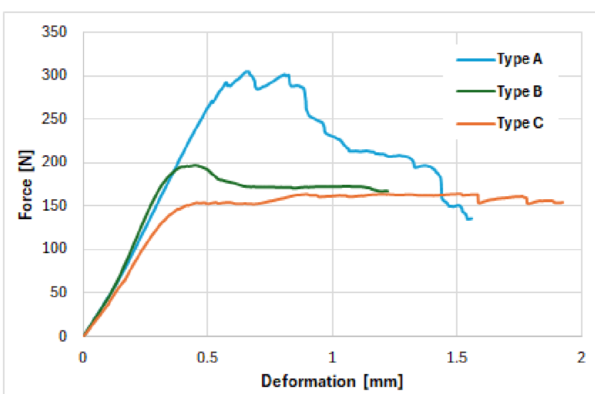

Figure 3. Typical curves during the short beam test.

The Type A composite showed the best load-bearing performance, reaching the highest ILSS among the investigated configurations. The initial region of the force–deformation curve was almost linear, followed by progressive damage accumulation as the load increased. After the peak load, the force gradually decreased, suggesting the development of interlaminar damage and delamination. Lower peak forces were recorded for the Type B and Type C specimens. Type B showed a rapid initial increase in force and reached its peak at approximately 180–200 N, followed by a gradual loss of load-bearing capacity. In contrast, Type C reached a lower peak force but showed a more stable response. After the peak load, the force remained nearly constant, suggesting a more gradual, controlled damage process.

The interlaminar shear strength of Type B was slightly higher than that of Type C, suggesting that the stacking sequence and structural arrangement of Type B provided more effective load transfer between the layers. However, both carbon-fabric-containing composites had lower ILSS values than the Type A composite, which was reinforced only with glass fabric. This difference can be explained by variations in adhesion between the different fabric types and by the interfacial behavior of the glass/carbon fabric interfaces.

The failure modes observed after the interlaminar shear strength tests are presented in Figure 4.

a) b) c)

Figure 4. Failure modes of the specimens after short-beam testing: a) Type A, b) Type B, and c) Type C.

In the case of Type A, a combined failure mode resulting from shear and tensile stresses caused by bending was clearly visible, leading to a mixed failure. In the case of short-beam or interlaminar shear strength testing, this is not considered an ideal or applicable mode of failure for composite materials, as the measured result is not determined only by the interlaminar shear strength. In the case of Type B and Type C specimens, damage occurred in the form of delamination propagating along the layers. In these specimens, a single mode of failure, predominantly due to shear, was observed, which is an ideal and acceptable failure mode in ILSS testing. This confirms that, in the case of composites containing carbon fabric, failure occurred largely along the layer boundaries, resulting purely from shear stresses. It is also interesting to note that, in both hybrid composites, failure initiated at the interface between the carbon and glass fabrics and propagated along it. This suggests that, in the case of the Type C composite, the incorporation of the copper mesh weakened the structure by a smaller degree than the interface between the different fabric types. This may be an important design consideration for hybrid composites into which we intend to integrate a conductive copper mesh or other functional layer.

5. Conclusions

In this study, self-sensing hybrid composites containing piezoelectric ZnO particles were developed and characterized, in which the sensing function was realized as an integral part of the material structure without the use of externally embedded sensors. The investigated material systems consisted of glass- and carbon-fiber reinforcements, an unsaturated polyester matrix, and piezoelectric particles incorporated into the matrix, enabling the measurement of the electrical response generated under mechanical loading.

Based on the force–voltage measurements, an approximately linear response was observed for all three composite types within the investigated loading range. According to the force-dependent results, the highest voltage response was measured for the Type A composite, which contained only glass fabric reinforcement, whereas the lowest signal values were obtained for the Type B composite at identical force levels. However, due to the higher flexural stiffness of the hybrid composites containing carbon fabric, the force–voltage-based comparison alone was not considered fully representative of the sensing performance. Therefore, the voltage response was also evaluated as a function of deformation, allowing a more relevant comparison of the electromechanical behavior of the composites.

Based on the deformation–voltage results, nearly identical sensitivity was determined for the Type A and Type B composites, indicating that the piezoelectric response was mainly generated by the ZnO particles dispersed in the matrix within the region between the two copper mesh electrodes. In contrast, a higher voltage response was measured for the Type C composite. This suggests that the carbon fabric layers may have contributed to the collection and transfer of the generated electrical signal, thereby improving the sensing efficiency of the hybrid structure.

The interlaminar mechanical behavior of the developed composites was evaluated by short-beam testing. The highest apparent interlaminar shear strength was observed in the Type A composite; however, based on the failure images, a mixed damage pattern from both shear and tensile stresses was observed in this specimen. This failure mode is not considered acceptable for ILSS testing. In contrast, in the Type B and Type C hybrid composites, damage occurred as delamination propagating along the laminate interfaces, indicating a predominantly shear-induced failure mode acceptable for ILSS testing.

The failure analysis further showed that, in the hybrid composites, damage occurred primarily at the interface between the carbon and glass fabrics rather than at the incorporated copper mesh layer. This indicates that, by using an appropriate stacking sequence, the weakening effect of the copper-mesh electrode can be reduced or eliminated, while the interface between dissimilar reinforcing fabrics may have a greater influence on interlaminar failure. This should be considered an important design aspect in the development of multifunctional hybrid composites into which conductive copper layers or electrodes are intended to be integrated.

6. Acknowledgements

The project supported by the Doctoral Excellence Fellowship Programme (DCEP) is funded by the National Research Development and Innovation Fund of the Ministry of Culture and Innovation and the Budapest University of Technology and Economics, under a grant agreement with the National Research, Development and Innovation Office.

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