DESIGN AND FABRICATION OF MULTI-FUNCTIONAL ENERGY STORAGE COMPOSITES INTEGRATING ULTRATHIN LITHIUM-ION BATTERY WITH ENHANCED ELECTRO-MECHANICAL PERFORMANCE

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Abstract: *Exponential advancement in the automotive and aerospace industry promotes the need for Multifunctional Energy Storage Composites (MESCs) to minimize the dependence on fossil fuels and reduce structural weight. This study proposes and evaluates a multi-functional carbon fiber reinforced polymer (CFRP) composite with an embedded lithium-ion polymer battery, demonstrating a structural integrity concept. Here electrospun epoxy-multiwalled carbon nanotubes (epoxy-MWCNT) nanofibers were incorporated precisely on the uncured CFRP surface to enhance adequate interfacial bonding and adhesion between the layers after curing. The mechanical and physical properties of modified CFRP have been evidenced to possess higher mechanical strength than the traditional CFRP composite. Commercial ultra-thin lithium-ion battery with higher energy density has been uniquely integrated into the core of the CFRP composite structure. Comparison with conventional CFRP composite and electro-mechanical testing ensured that the electrochemical property of the embedded battery was preserved in loading/unloading conditions, and the mechanical strength of the composite structure was not compromised.*

Keywords: Multi-functional Composites; Li-ion Battery, Electrospinning, Nanofibers, CFRPS

1. Introduction

Recently, structural batteries have emerged as a trendy concept for overcoming functional restrictions in load-bearing to achieve weight and volume savings in many structural domains such as airplanes, spacecraft, and commercial vehicles (1-3). These structural batteries provide energy storage capabilities. Rather than the energy storage elements themselves, the packaging material is critical for load-carrying capacity in these existing batteries. Numerous commercial sectors are attempting to lower the structural weight of their key products in order to mitigate global warming. Carbon fiber composites are increasingly being employed in structural components of motor vehicles, such as the body panels and chassis, to accomplish this goal. With the advancement of electric propulsion and the increasing demand for hybrid and electric vehicles on the market, issues for energy storage in these vehicles arise in terms of vehicle space. This also influences the vehicle's empty weight, as the battery system accounts for up to 25% of the vehicle's total body weight (4, 5). This creates relatively large storage space in the volume. In order to increase the vehicle's total storage, structural, and space economy, it is required to incorporate multi-functional composites into vehicle components. There are a number of methods for integrating electrical storage devices into composite structures. The first is the fabrication of the composite into a structural dielectric capacitor (SDCs) (6-8).

Contemporary Li-ion batteries are primarily designed for maximum energy storage performance at the expense of mechanical load carrying capacity and robustness. Li-ion pouch cells are constructed by stacking alternating anode and cathode layers separated by thin microporous polymer separator membranes (9). The advanced thin electrode films are made of copper and aluminum, which have a high structural composition. When these pouch cells are bent, applying the least mechanical load results in unjustified deformation and layer slippage. Due to the vacuum-sealed aluminum-polymer-laminate packing material, the structure has negligible strength. This study aims to synergistically syndicate the load-bearing capabilities of present battery elements to incorporate mechanical robustness into cells, resulting in significant volume and weight savings in the packaging. This work aims to fabricate structural load-bearing batteries using multi-functional energy storage composites (MESC) as an alternative technique. MESCs are a revolutionary energy storage device because they combine high mechanical strength with low weight and superior energy storage capabilities. MESCs are constructed by embedding Li-ion battery electrode materials in high-strength CFRP composites (10). This technique does not require altering Li-ion batteries' electrochemistry and can be included in standard industry designs, which are critical for engineering implementation. Due to the sandwich-style construction, the laminate's moment of inertia increases significantly, resulting in increased flexural rigidity. This mechanical robustness of the proposed CFRP composite structures enables them to be manufactured as multi-functional energy-storage devices for electric vehicles and other structural applications.

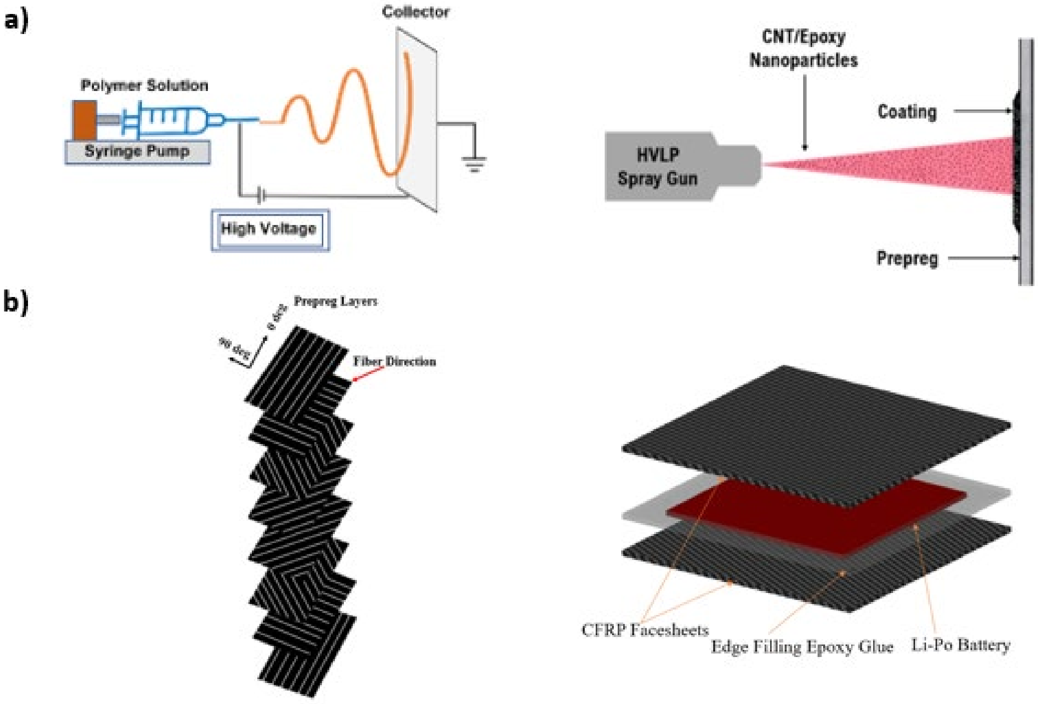

Figure 1: a) Electrospinning and air-spraying setup, b) stacking the prepreg layer (0°/90°/+-45°)2s., and integration of Li-ion battery in the core of the structure.

2. Methodology

2.1 Material Preparation

Masterbatch of MWCNTs/epoxy was synthesized using a previously published method by our group (10-12). Bisphenol A (50 - 99 pbw.%) and carbon nanotubes (5 wt.%) were mixed together, followed by the addition of Dimethylformamide (DMF) (1:4 volume ratio) and Triton X- 100 (20:1 volume ratio). Then the resulting mixture was sonicated for 10 mins in intervals of 45s and 30s rest between cycles. The same weight of more epoxy was added to the mixtures, and 15 mins stirring was done, followed by sonication. Finally, to have a uniform viscous mixture, the curing agent was mixed at a ratio of 15:1 and stirred at 50°C for 2 hours to obtain a homogeneous solution. The uniform mixture was then degassed and rested for 16 hrs prior to electrospinning. A syringe with a needle gauge of 26 G was filled with MWCNTs/epoxy mixture for electrospinning onto the CFRP layer. The Epoxy-MWCNT solution was air-sprayed onto the CFRP layers as an alternative method.

2.2 Electrospun CFRP Facesheets Sample Preparation

Electrospun and air sprayed Epoxy-MWCNT nanofibers were deposited onto (10 x 10 cm) precut prepreg layer with a plain weave pattern (SE70 Gurit Holding AG, Wattwil/ Switzerland). To decrease the void ratio, a hand layup process was used, followed by vacuum bagging. The stacking sequence on one side of the cell is $[0/90/\pm45]_{2s,}$ as reported in our previous work (13, 14). Square slots were cut before laying up in the stacking. A total of twenty layers were employed to create a single sample. Before curing, the samples were vacuum bagged for 60 minutes to ensure optimal adhesion between the layers. The sample was completely cured by placing it in a programmable oven (Easy Composite, UK) set to 120^{o}C for 25 minutes and vacuuming to a pressure of less than 1 bar. While maintaining the pressure, the samples were cooled to room temperature. The samples were given a smooth finishing at the edges for appearance. After curing, the sample had a final thickness of 0.52 cm.

2.3 Assembly of the Batteries Inside the CFRP Samples

The fabrication method of the MESC cell was operated sequentially. Two CFRP facesheets with the rectangular slot in the middle are placed flat on the surface. A prepreg sheet of the exact dimensions with a slot space is employed as the adhesion between the two surfaces. The edges of the two surfaces are heated at a temperature of 70^{o}C by means of a heat gun. The prepreg sheet is placed on one side of the CFRP face sheet, and the pouch cell battery is placed at the center of the surface. Due to the heated surface, the epoxy in the uncured prepreg sheet enables it to act as an epoxy adhesive, and both the surfaces are then joined together. The sample is then compressed in a heated hydraulic press to melt further the epoxy resin in the middle layer of the sample and helps to fuse both the surfaces of the facesheets together securely (100^{o}C, 0.5 MPa pressure). The sample remained to cool down to room temperature under the same temperature. During this phase, the epoxy solidified and thus equilibrated the stack mechanically. Edges of the sample were then sealed off with additional epoxy adhesive for a tidier appearance and homogeneity of the two surfaces. This method produced six samples with electrospun, air sprayed enhanced CFRP, and control CFRP sheets.

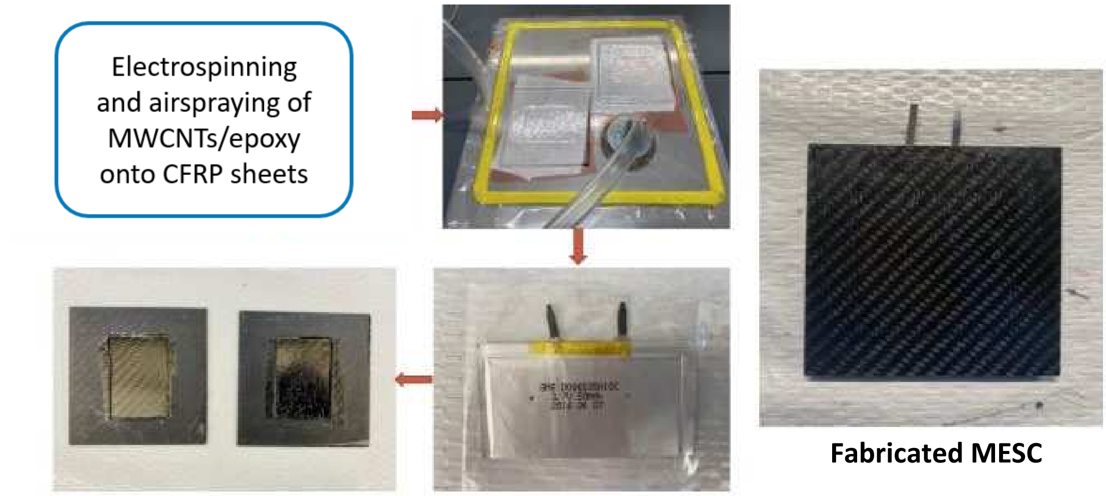

Figure 2: Hand layup method to fabricate MESC by enhanced CFRP facesheets.

3. Test Methods

3.1 Quasi-static Three-Point Bending

Three-point bending is performed to define the inter-laminar bond strength characteristics of the various types of CFRP-based MESC samples. For mechanical testing, a cylindrical roller is used as an applicator on the fixture for three-point bending (2 kN servo-hydraulic load frame, Integrated Resources Inc. MTS machine). This testing procedure followed the ASTM C393 standard technique. Flexure testing generates tensile stress on the convex side and compression stress along the midline, resulting in a shear stress area. The force required to bend the beam is measured under three-point loading circumstances. The cylindrical load applicator is positioned with its axes parallel to the load applicator axis and parallel to the specimen axis. At the mid-span of the sample, the cylindrical load applicator applied a vertical downward force along the line. The evaluation of adequate flexural rigidity of the sample is validated through the reinforcement of the inter-layer-shear inhibition capabilities. The load on the sample was applied at 3.33 mm/min. A linear variable differential transducer was employed throughout the experiment to measure the vertical displacement at the mid-span. The initial loading results were recorded, and the sample was subjected to continuous loading on the mid-span for 12 hours. This testing aims to analyze the battery characteristics for a period of time of continuous bending.

3.2 Electrochemical Characterization

The MESC samples were initially subjected to a continuous maximum bending load for 12 hours. The sample pouch cell battery was subjected to a bending load until deformation and when it was not under any loading and then tested. The initial purpose was to conduct an in-situ testing environment wherein the samples were subjected to bending loading and, at the same time, evaluated for electrochemical performance. However, due to the logistical constraints, the samples were subjected to the above specific bending cycle and then tested later. The samples were initially exposed to a slow calibration cycle between 3.0 V and 4.2 V to define the C rate. It is also the current at which the battery is discharged in 1 h. The initial discharge capacity of each sample at the beginning of life (BoL) was measured under constant current (CC) cycling at C/10.

The pouch cell battery is used for this testing to record the base performance. This testing calculates the expected capacity on the amounts of active materials added. The depth of discharge (DoD) is measured from the voltage difference while the current is interrupted. This charge-discharge cycle was repeated to measure the cell life performance by increasing the cycles of different sample types to compare the discharge capacity retention properties.

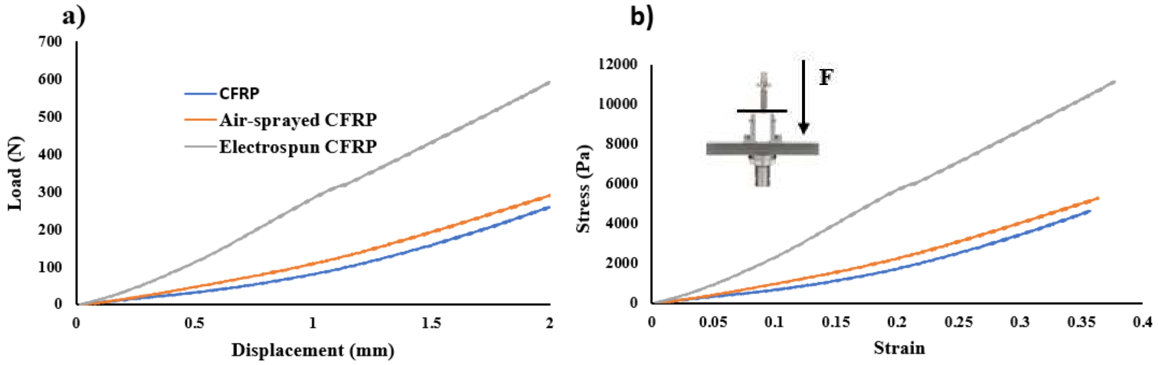

Figure 3: a) Load vs. displacement and b) strain vs. stress curves for different types of MESCs.

4. Results and Analysis

4.1 Quasi-static three-point bending test

The three-point bending for eight cell samples was performed to evaluate the mechanical performance of the MESC within themselves and the control pouch cell battery. The Interlaminar Shear Strength (ILSS) test sampling is much more insistent than the other shear tests. The values for flexural stress, elasticity in bending, and flexural strain are observed. The results are sensitive to the testing method based on the specimen, loading geometry, and strain rate. The stress required to fracture the samples yields a stress-deflection curve slope in this testing. The bending moment varies from zero at the support and maximum at the center. The electrospun CFRP sample has shown the highest load of 695.47 N, as shown in figure 3(a). The displacement had a limit of 2 mm as it is assumed to be the ideal state where the battery can take the maximum load without affecting its electrochemical characteristics. There is also an observation here that deformation of layers in the top did not occur and this maximum load could be further extended given there were no constraints on the displacement side. On the other hand, the control sample fabricated as a conventional CFRP sample shows the lowest peak load of 289.38 N with subsequent deformation in its top layers. The air sprayed CFRP sample displays a peak load of 365.25 N and lesser damage in the top layers than the conventional CFRP sample. The pouch cell battery could not register a peak load of more than 6 N before being completely deformed.

4.2 Flexural Rigidity

Flexural testing is the most practical method for gauging fiber-resin interface and matrices to assess the improvements in interlaminar properties. The electrospun CFRP sample demonstrated high flexural strength and strain compared with the control CFRP, and the air sprayed CFRP samples. The primary considerations in this sample were previous research experimentation revealed nano reinforcements integration causes a considerable increase in

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the flexural properties. The packaging of the lithium-polymer battery shows a flexural strength of 410.27 psi and goes onto total deformation, while the control and the air-sprayed samples show a flexural strength of 230.15 psi 390.31 psi, respectively.

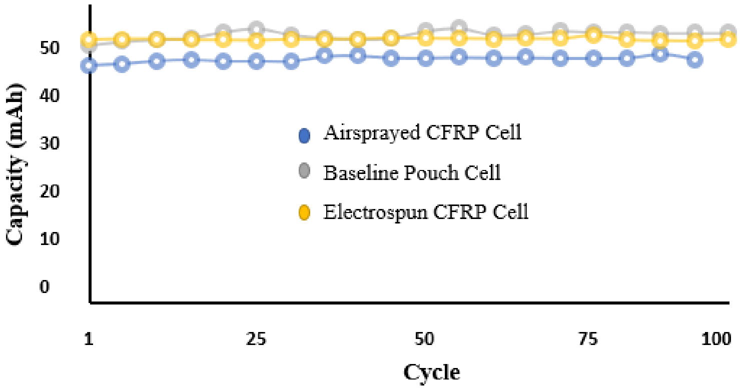

Figure 4: Battery performance inside different MESCs under loading conditions.

4.3 Electrochemical Analysis

The batteries were cycled using Landt battery analyzers (Landt Instruments), and all the electrochemical testing was performed at a temperature of 30° C. Each MESC was cycled 100 times at a rate of 1C. After each cycle, the charge-discharge rate and battery capacity were determined. Additionally, the baseline pouch cell battery was cycled before and after applying bending pressures. It demonstrated that the battery integrated into the electrospun CFRP enhanced MESC performs identically to a conventional pouch cell, as shown in figure 4. Similarly, the controlled and air played CFRP enhanced MESC significantly decreased battery performance under loading conditions. This demonstrates that electrospun CFRP cells are the optimal construction for the MESC in terms of overall testing properties.

5. CONCLUSION

The embedding of LiPo batteries within the sandwich CFRP surfaces' core in different orientations and characteristics presents a novel form of structurally integrated batteries in a distinctive material with a vertical integration process. In this process, the fundamental mechanical properties of the CFRP materials make the industry-standard Li-ion battery much more robust in a structural applications environment under the architecture of MESC. The highest strength in electrospun CFRP sandwich structures did not affect the batteries when the possibility of the malfunction was localized metal gouge to the structure's core. There was a significant increase in the mass of the sandwich composite with the inclusion of the LiPo batteries since they are much denser and can increase the self-weight of the structure and also can reduce the stiffness and strength at the same time. The design consideration, in this case, can be an equilibrium of particular properties and aim at increasing the energy storage density for obtaining lightweight structures. This was proven correct by the electrospun CFRP samples, which were comparatively lighter in weight than the air-sprayed and conventional samples and thinner in construction, resulting in maximum mechanical strength. The process of electrospinning the conventional CFRP facesheets is also economical and can open a versatile range for this kind of device in automotive structural applications and areas where flexibility can be a challenge.

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