

Article

Ultrasonic Spray Coating of Carbon Fibers for Composite Cathodes in Structural Batteries

Thomas Burns ¹, Liliana DeLatte ² , Gabriela Roman-Martinez ¹, Kyra Glassey ³ , Paul Ziehl ⁴,
Monirosadat Sadati ¹, Ralph E. White ^{1,*} and Paul T. Coman ^{1,*} 

¹ Department of Chemical Engineering, University of South Carolina, 301 Main Street, Columbia, SC 29208, USA; tburns@email.sc.edu (T.B.); romanmag@email.sc.edu (G.R.-M.); sadati@cec.sc.edu (M.S.)

² Department of Chemistry, Wofford College, 429 N Church St, Spartanburg, SC 29303, USA; delattele@wofford.edu

³ Department of Chemical and Environmental Engineering, University of Arizona, 1133 E. James E. Rogers Way, Tucson, AZ 85721, USA; keglassey@arizona.edu

⁴ Departments of Mechanical and Civil and Environmental Engineering, University of South Carolina, 301 Main Street, Columbia, SC 29208, USA; ziehl@cec.sc.edu

* Correspondence: white@cec.sc.edu (R.E.W.); comanpt@cec.sc.edu (P.T.C.)

Abstract: Structural batteries, also known as “massless batteries”, integrate energy storage directly into load-bearing materials, offering a transformative alternative to traditional Li-ion batteries. Unlike conventional systems that serve only as energy storage devices, structural batteries replace passive structural components, reducing overall weight while providing mechanical reinforcement. However, achieving uniform and efficient coatings of active materials on carbon fibers remains a major challenge, limiting their scalability and electrochemical performance. This study investigates ultrasonic spray coating as a precise and scalable technique for fabricating composite cathodes in structural batteries. Using a computer-controlled ultrasonic nozzle, this method ensures uniform deposition with minimal material waste while maintaining the mechanical integrity of carbon fibers. Compared to traditional techniques such as electrophoretic deposition, vacuum bag hot plate processing, and dip-coating, ultrasonic spray coating achieved superior coating consistency and reproducibility. Electrochemical testing revealed a specific capacity of 100 mAh/g_{LFP} with 80% retention for more than 350 cycles at 0.5 C, demonstrating its potential as a viable coating solution. While structural batteries are not yet commercially viable, these findings represent a step toward their practical implementation. Further research and optimization will be essential in advancing this technology for next-generation aerospace and transportation applications.

Keywords: structural batteries; ultrasonic spray coating; multifunctional energy storage; carbon fiber electrodes; lithium-ion batteries



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1. Introduction

Structural batteries have gained significant attention in recent years due to the multifunctionality of carbon fibers, which can store electrical energy through electrochemical reactions and provide significant strength, making them ideal candidates for mechanical and aerospace applications [1–3]. Although their energy density is lower than traditional batteries, this metric ought to be calculated differently. Structural batteries do not require large, heavy current collectors and can replace structural parts of vehicles, meaning the energy density should be evaluated at a system level and not as standalone units [4].

The electrochemical mechanism of lithium-ion batteries (LIBs) is well studied with apt mathematical models for analysis [5]. However, fully harnessing and optimizing the energetic capabilities of lithium ions in structural batteries involves much more testing and experimentation.

Many studies have investigated the electrochemical and mechanical performance of structural batteries. Johannisson et al. [6] were able to construct and test a structural battery with an energy density of approximately 25 Wh kg^{-1} . Scholz et al. [7] also determined that the specific energy for a structural battery must be at least 51.8 Wh kg^{-1} to make notable contributions towards powering electric devices. Another standard needing to be met by structural batteries is the ability to cycle for hundreds if not thousands of times with significant power densities and capacity retention. However, carbon fiber electrodes can barely withstand tens of cycles, reinforcing the necessity of further research [3].

Chen et al. [8] provided an excellent compilation of data and analysis for the progress and outlook of structural batteries. Structural batteries, while making progress, must navigate the tradeoff between mechanical strength and electrochemical performance. Thomas et al. [9] produced a functioning soft pouch; however, the battery's viscoelastic shear bonding reduced the cell's strength by 50% in comparison to a conventional LIB. For structural cells to produce enough electrochemical potential to compete with conventional cells, the mechanical strength provided will be significantly reduced. Chen et al. [8] discussed the DARPA structural battery that enhanced system performance but ultimately did not yield any mechanical capabilities. Furthermore, the authors discussed the results of interlocking rivet configuration NCM/graphite batteries that exhibited a specific energy of 131 Wh kg^{-1} , an 80% capacity retention after 800 cycles, and a bending stiffness of 12 N m^2 . Consequently, these batteries also exhibited a 40% decrease in energy density.

While reviewing the current state of the academic field surrounding structural batteries, the rapid pace of improvement is evident. However, further research on how to optimize the mechanical strength, electrode-to-electrolyte interface, and energy density is necessary. The optimization of active material coating methods is integral in the pursuit of creating a competitive structural battery cell.

The prominent method of slurry casting active material onto structural bases has dominated the field of LIB production. Slurry casting generally consists of four steps, with specific parameters varying from adaptation to adaptation: the preparation of the solvent, the casting of the active material (often by calendaring), active material drying, and then shaping, such as punching out a coin cell [10]. While this technique is one of the most widely used and optimized techniques in the field, it does not come without disadvantages. The active material drying process is not in tandem with the active material application, which causes this process to be more time consuming. Furthermore, with the lack of clear parameters, this technique allows for a wide variety of resulting battery efficacy, thereby producing a widely varied industry standard.

One of the challenges remains achieving a good coating of the carbon fibers with active materials while maintaining structural integrity and electrochemical performance [1,11]. Among the coating methods explored, electrophoretic deposition (EPD) has been prominent. First used in early 2000s for Li-ion battery coatings, EPD can deposit a wide range of materials, including ceramics, polymers, and carbon nanotubes. It offers uniform coatings with controlled parameters and adjustable thickness. However, the conductive materials and parameters used to ensure a controllable process are complex. Moreover, since EPD requires an electric field, the coating process becomes difficult and complicated when upscaling to larger structural cells. This method is more suitable for small-scale projects, which may result in slurry waste. EPD coatings have achieved specific capacities of $62 \text{ to } 108 \text{ mAh/g}_{\text{LFP}}$ obtained at 0.1 C and a capacity retention of 62% after 500 cycles or 47% after

1000 cycles as compared to BOL (beginning of life) for a coating with LiFePO_4 (LFP) cathode material [1]. However, improvements to the EPD coating have been demonstrated in the literature. By adding nanosheets, it was shown that the specific capacity was increased to $130 \text{ mAh/g}_{\text{LFP}}$ at 0.1 C and BOL and $90 \text{ mAh/g}_{\text{LFP}}$ at a 1 C rate along with a capacity retention of 80% after 500 cycles [12].

Another method for coating that was demonstrated to offer good-quality coating was the vacuum bag hot plate method [11]. In this method, the carbon fibers are inserted into a pouch cell together with a slurry containing the active material, a conductive filler (C-45, for example) and a binder (PVDF, for example). The authors in ref. [11] showed in a half-coin-cell configuration with a Li/Li^+ reference electrode that coating LFP using this method can achieve 140 mAh/g at BOL and about 144 mAh/g at 0.33 C and 102 mAh/g at 1 C when the capacity retention dropped to 80% after 300 cycles.

Another method for coating is the layer-by-layer (LbL) deposition, which is a multi-step process involving the alternate dipping of carbon fibers into positively and negatively charged solutions to build up multiple layers [13]. The LbL method allows for precise control over the thickness and composition of the coating by varying the number of bilayers. This technique is particularly advantageous because it ensures uniform deposition of lithium oxide particles (LFP) onto the carbon fibers, which is crucial for achieving good electrochemical performance. The use of carboxymethylated cellulose nanofibrils (CNF) as a binder and polyethyleneimine (PEI) to modify the surface charge of LFP particles facilitates strong electrostatic interactions and robust layer formation. Furthermore, the LbL method incorporates a carbonization step, which converts the organic and insulating binders into an electrically conductive network. This step not only enhances the electrical conductivity of the coating but also removes organic materials that could induce side reactions with the electrolyte, thereby improving the electrochemical stability of the electrodes. The carbonization process is typically performed at temperatures around 450°C , which has been found to optimize the electrochemical properties of the electrodes, resulting in specific capacities exceeding $100 \text{ mAh/g}_{\text{LFP}}$ at a C rate of 0.1 C . The electrochemical properties achieved by this method are promising, but upscaling this method offers challenges for commercial viability such as maintaining the high temperatures required for optimal carbonization.

A similar and a one-step dip-coating mechanism proven to show a good electrochemical performance was demonstrated by Petrushenko et al. [14]. Their method consisted of dipping bare carbon fiber tapes in a slurry of LiFePO_4 , C-45, and PVDF. This method was developed as a motivation to produce reel-to-reel coating of tows of carbon fibers. The authors demonstrated that the capacity of LiFePO_4 cathodes dip-coated on the carbon fiber reached values of $55 \text{ mAh/g}_{\text{CF}}$ (per gram of carbon fiber) with a capacity retention of 90% after 20 cycles. It was later proven in this study that the dip-coating of LFP on carbon fibers results in specific capacities of $84 \text{ mAh/g}_{\text{LFP}}$ and a capacity retention of 82% after 30 cycles.

Electrode epoxy impregnation coating was also shown to provide an efficient electrode. Moyer et al. [15,16] demonstrated a method that the authors state “locks-in” the interface between the carbon fiber and the active material by using a slurry consisting of LFP, carbon black, PVDF binder, and carbon nanotubes on a carbon fiber weave. They demonstrated that at the full-cell level, the specific capacity reaches $20 \text{ mAh/g}_{\text{FULL CELL}}$ and a good capacity retention of 80% for more than 100 cycles. Another impregnation method with an unrolling station was demonstrated by Yücel et al. [17]. Carbon fiber tows were fed through a small slot used to control the thickness of the added active material. This slot had a pressured feed of active material that would impregnate the carbon fibers as they were unrolled. The authors obtained a good specific energy of about $151 \text{ mAh/g}_{\text{LFP}}$ at $\text{C}/14$ with a capacity retention of 81% after 100 cycles in their best scenario (when the coating was dried at 140°C) but 54% after 100 cycles at 60°C .

Another recent spray-coating method was published by Yücel et al. [18]. The authors used the classical lab spray gun to spray slurry on the carbon fibers that were placed on a hot plate heated at 100 °C, with a N₂ inlet. The authors showed that the spraying method can deliver over 140 mAh/g_{LFP} at rates under 1 C, similar to the ones with the enhanced electrophoretic method, while demonstrating a capacity retention of 77% after 100 cycles. That indicates that the spraying method might be an appropriate method to enhance energy density. Additionally, spraying is a good method for upscaling to larger surfaces of CF that would need to be coated. However, the human aspect of this method means reproducibility would be difficult to achieve, with its varying electrode performance making it commercially unviable.

In this paper, an ultrasonic spray-coating method is explored, utilizing computer numerical control equipment. This technique allows for coating large surfaces in an environmentally friendly manner with minimal slurry waste. The ultrasonic spray nozzle follows a programmed route, atomizing the carbon fiber surface, and can execute predefined spraying routines. Furthermore, the ultrasonic spray-coating technique results in a completely superficial coating, minimizing any permeation of the active material into the carbon fiber. Not only is this an asset to a structural cell application, but this also ensures that coating of only one side of the carbon fiber base is easily achievable. This technique also allows for easy replication, as the process is relatively straightforward and can be pre-programmed to produce ideal coatings. Compared to other methods, ultrasonic spray coating offers superior control, efficiency, and scalability, making it a good commercial candidate for coating carbon fibers in structural batteries.

2. Materials and Methods

Toray T800S 12K carbon fiber was chosen for electrode construction. The slurry for coating the carbon fiber was prepared using 80% LFP, 11% carbon black, and 9% PVDF binder. This combination was then added to 1-methyl-2-pyrrolidone (NMP) in a 10:1 ratio of NMP to active material. Materials were gathered and slurry preparation carried out for this study in accordance with that in ref. [14]. Replication of the dip coating technique seen in ref. [14] was also performed for a surface comparison with the ultrasonic technique.

2.1. Ultrasonic Coating

For the ultrasonic coating, a SONO-TEK ExactaCoat machine (Milton, NY, USA) with the ultrasonic head configuration was used to spray coat swatches of carbon fiber tows inside an encased fume hood. This machine is manufactured by SONO-TEK at their industrial headquarters in Milton, New York, USA. The attached nozzle (AccuMist™ nozzle) transfers high-frequency sound waves into mechanical energy to create standing waves in the mixture passing through the nozzle end, which atomizes the active material as it exits the nozzle. The operating frequency of the nozzle dictates the nominal droplet size by fluctuating the vibrations felt in the mixture. The vibrations created a soft spray with a homogeneous droplet size at a constant frequency, resulting in a uniform surface coating. Two different syringe pumps were used: the standard syringe pump and the SonicSyringe pump shown in Figures 1a and 1b, respectively. The SonicSyringe works similarly to the head nozzle, using sound waves to maintain a uniform mixture within the syringe during the coating process.

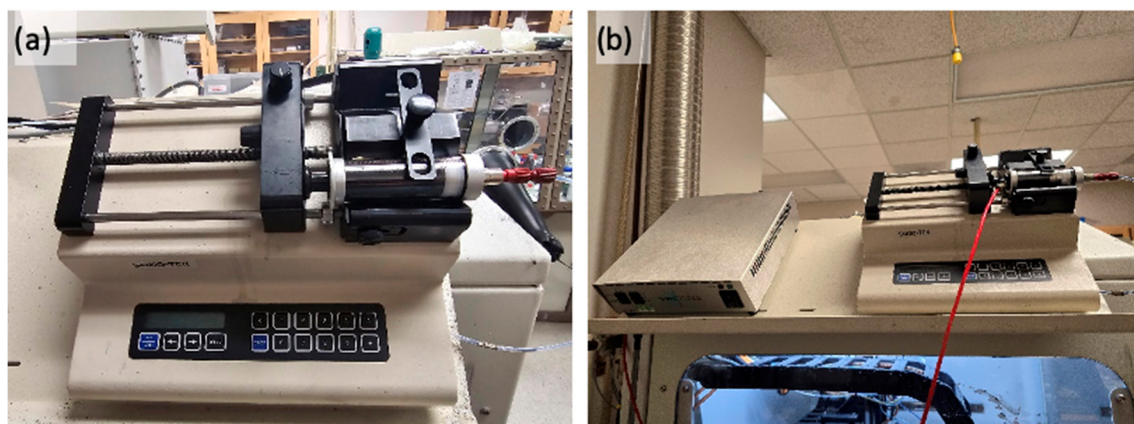


Figure 1. SONO-TEK ExactaCoat syringe configurations for (a) standard syringe and (b) SonicSyringe operation.

Bare Toray T800S 12k carbon fiber tows were laid taught across a stainless-steel 7 cm by 7 cm frame and secured with painter's tape (Figure 2). This swatch of carbon fiber was centered in the ultrasonic machine on a curing plate. The XYZ tracing system, installed with the machine, was used to input the dimensions of the swatch into the software by mapping three corners of the area to be coated. A working distance of 45 mm in the Z direction was input manually. The software used for the Exactacoat machine is Windows-based and is called Pathmaster, the version used was 1.0.3. In the Pathmaster software, this 45 mm value is based on the internal tracing dimensions of the nozzle, meaning 0 mm correlates to the highest position the tracing system can achieve. Due to this, the actual distance from the nozzle to the CF swatch was 1.75 cm. The ultrasonic nozzle traced the area in the Y direction at 10 mm/s, coating columns 2 mm apart in the X direction and spraying at a rate of 6.5 mL/h.

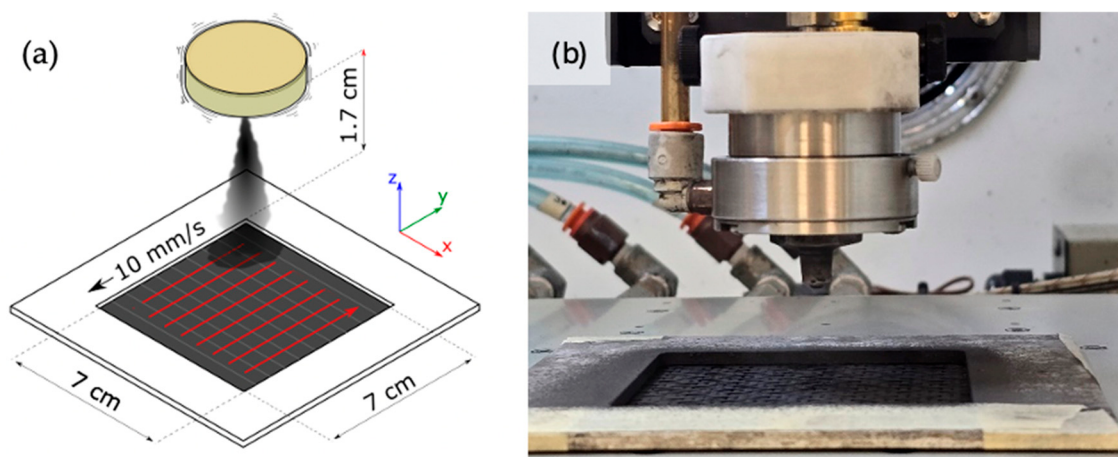


Figure 2. (a) Sketch showing the vibrating ultrasonic head, the zig-zag coating path, the fiber array direction, the coating velocity, and the overall dimensions; (b) the real-life system.

The nozzle utilizes a centering head that creates a cone of air to adjust the spray width. Air pressure for the centering head was set at 2.50 kPa. A total of 11 passes were made, resulting in a coating of 5 mL on one side of the carbon fiber swatch. The curing plate cured the slurry in tandem with the spray application at a temperature of 50 °C. The ending position of the nozzle head was raised to a distance of 25 mm in the system to prevent the heat from curing the slurry inside of the nozzle head. The temperature, nozzle speed and distance, spray rate, and centering air pressure were chosen due to the increased control

at these lower rates. A faster coating with similar or superior results is likely achievable and should be explored further. Once the slurry application was complete, the curing plate remained at 50 °C, and the swatch sat in the ultrasonic coating machine for 24 h to ensure complete curing.

2.2. Coin Cell Assembly

After ultrasonic coating, the cathodes were analyzed using a Zeiss Gemini 500 Field Emission SEM to observe the coating quality, thickness, and penetration depth. The sheet was then used to assemble coin cells in accordance with the procedure described by Petrushenko et al. [14]. For the electrolyte, 100 μL of a 1.0 M solution of LiPF_6 dissolved in 1:1:1 EC/DMC/DEC was used. The lithium disk anodes used in the coin cells had a diameter of 16 mm and a thickness of 0.6 mm. The areal active material loading on the carbon fiber in the coin cells was found to be 5.7 mg/cm^2 . The coin cells were left to sit in the argon environment in the glovebox for 24 h and then tested using an Arbin MSTAT battery cycler. The half-cells were cycled from 2.75 V to 4.0 V vs. Li/Li^+ with a small initial current of 715 μA to promote the formation of a solid electrolyte interface (SEI). After SEI formation, the current was increased to a rate of 0.5 C for electrochemical testing, and the cells were cycled until end-of-life (EOL). A specific theoretical capacity of 170 mAh/g for LFP was used to determine C based on the mass of coated active material on the carbon fiber. This correlates to an areal capacity of 0.98 mAh/cm^2 used for cycling. One cycle included a charge and discharge with no rests in between cycles or changes in C rates.

3. Results

3.1. Scanning Electron Microscopy Analysis

Surface analysis of both dip-coating and ultrasonic coating showed significant differences. The uniformity of the dip-coating surface was subpar; there were areas of the swatch with a thick coating and areas with bare carbon fiber exposed. The coating seen in Figure 3a could alter the amount of contact between the electrode and the separator. This lack of contact could potentially create pockets that could encourage lithium plating. Another topic of interest is the adhesion of active material to the carbon fibers, especially regarding systems using multiple layups and active material layering. Dip-coating ensures more penetration (as is the case with electrophoretic method), but the accessibility to the electrochemical active surface area might be blocked by the tri-dimensional distribution of the active material, creating a hybrid electrochemical area.

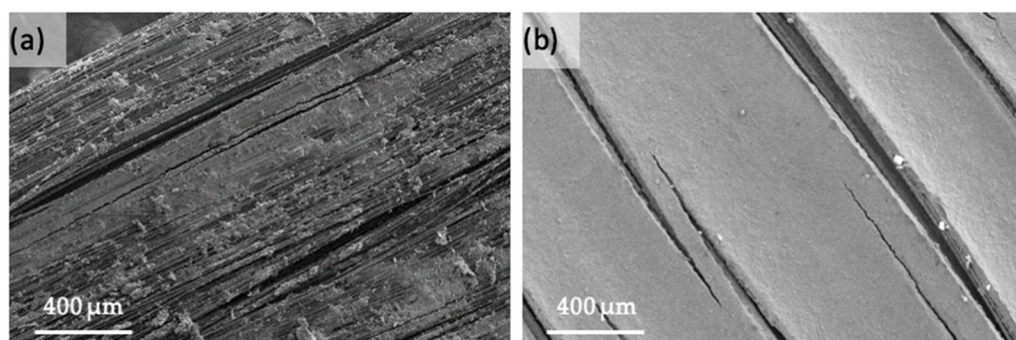


Figure 3. Zeiss SEM pictures at 428–459 magnification comparing the surfaces of (a) dip-coated carbon fibers and (b) ultrasonically coated carbon fibers.

The ultrasonic coating seen in Figure 3b appears uniform. Some portions of the swatch have slight cracking between the carbon fiber tows, but cracking is very minimal in relation to the swatch area. There is nearly no exposed carbon fiber, and the thickness of the coating

is uniform throughout the sample. Another important note is that the uniformity from tow to tow varies widely in the dip-coated sample, whereas the carbon fiber tows in the ultrasonic sample are nearly identical. In a scenario where the cathode production is upscaled, uniformity between samples would be necessary to ensure equal performance in all LIBs.

The dip-coated surface is wildly varied and irregular, while both the spray and ultrasonic coatings are extremely uniform even at a much deeper magnification, as seen in Figure 4. It is important to note that the slurry itself appears to have the same uniformity in particle size and its homogeneous composition on both samples. This indicates that the difference in coating uniformity is a result of the coating technique and not the active material or carbon fiber base.

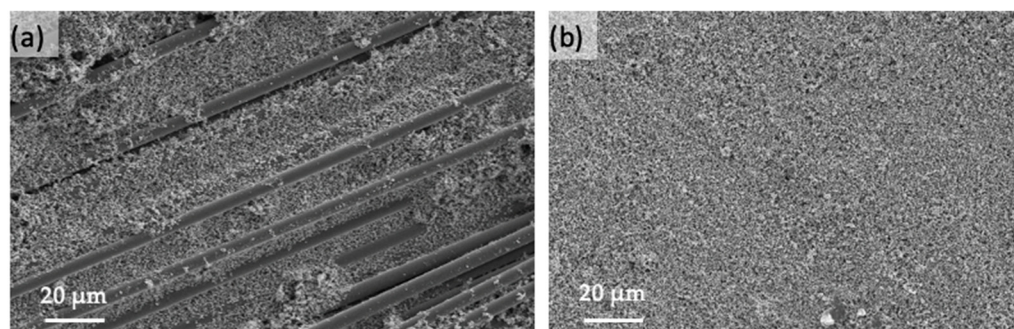


Figure 4. Zeiss SEM pictures at 5.00 K magnification comparing the surfaces of (a) dip-coated carbon fibers and (b) ultrasonically coated carbon fibers.

In Figure 5a, the permeation of the active material into the dip-coated carbon fiber can be seen. While not complete permeation, there is a significant difference between the permeation resulting from the dip-coating technique, as shown in Figure 5a, and the lack thereof from the ultrasonic technique, as shown in Figure 5b. However, it is important to note that the permeation in the dip-coated tows is not uniform throughout; there are sections with deeper permeation and sections with relatively shallow permeation. This could be a reason for the uneven surface coating. In a structural cathode composite, active material permeation may be crucial in handling any bending or warping of the structural cell. In Figure 5b, a complete surface coating of the active material resulting from the ultrasonic coating technique can be seen. There appears to be no deep permeation of active material into the carbon fiber, but the surface coating of the active material is uniform and even, exhibiting a thickness of about 1–2 μm , as seen in Figure 5c. While active material impregnation of carbon fiber might be desirable in structural cells for flexibility, a uniform and even surface coating of active material may be crucial for structural cells. The future of structural cells relies on solid electrolyte instead of liquid; full contact between the active material and the electrolyte will be necessary for a continuous ion transfer. With the uneven dip-coated surface, there could be a reduction in electrode–electrolyte contact. In contrast, the ultrasonically coated active material forms an excellent surface coating that optimizes the surface area present for contact with solid electrolyte. Further testing regarding the importance of active material permeation must be conducted, but this is beyond the scope of this study.

As seen in the labelled SEM images, the width of the dip-coated carbon fiber tow seen in Figure 6a (313.2 μm) is roughly double the width of the ultrasonically coated carbon fiber tow (145.3 μm). This thickness increase could stem from many factors, such as the increased permeation of the slurry into the carbon fiber with dip coating. Another contributing factor could be the excess of slurry applied to the carbon fiber tow, which is inevitable with the dip-coating technique or other methods, for that matter. The additional weight of the

slurry on the tow, specifically on the edges, could cause folding regardless of the tension applied by the dip-coating rods. Furthermore, the ultrasonically coated carbon fiber tow in Figure 6b is very compact and uniform in its fiber density. Again, this could be attributed to multiple reasons, but the two leading hypotheses are that the stainless-steel framework used during coating provides even tension across the tows, preventing warping, and that the simultaneous coating and curing prevents overloading of the tows. Maintaining the structural integrity of the base tows is crucial in ensuring the mechanical strength of the carbon fibers remains even after coating.

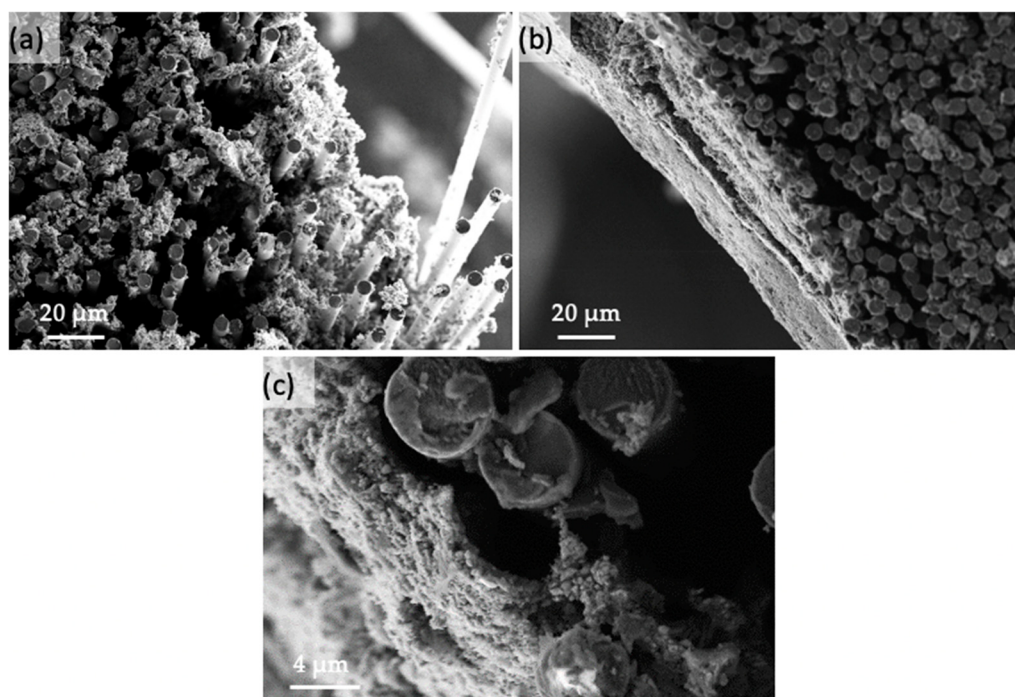


Figure 5. Zeiss SEM cross-sectional pictures of (a) dip-coated carbon fibers at 5.00 K magnification, (b) ultrasonically coated carbon fibers at 5.00 K magnification, and (c) ultrasonically coated carbon fibers at 30.00 K magnification. In (a), active material permeation can be seen throughout the carbon fiber tows, whereas in (b,c), the active material remains superficial.

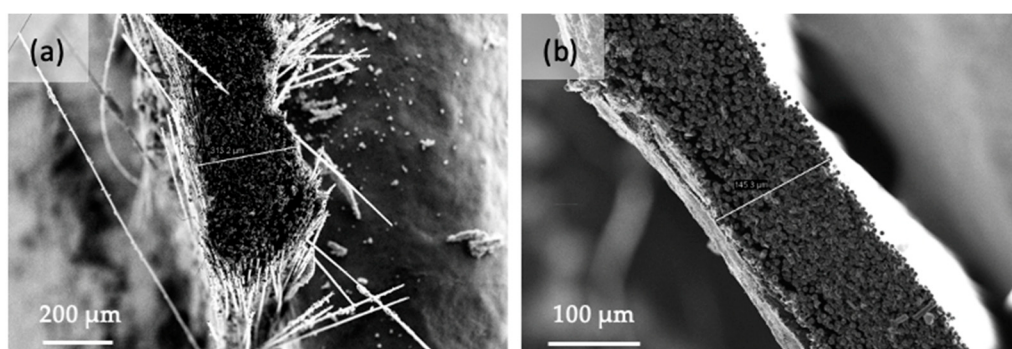


Figure 6. Zeiss SEM pictures at low magnification with labelled widths of (a) dip-coated carbon fiber tows and (b) ultrasonically coated carbon fiber tows. Figure (a) shows a width of 313.2 µm for the dip-coated carbon fibers, while Figure (b) shows a width of 145.2 µm for the ultrasonically coated carbon fibers.

3.2. Electrochemical Testing

For electrochemical testing following initial half-cell construction, cyclic voltammetry was performed on a LFP-coated carbon fiber produced utilizing the AccuMist™ ultrasonic

nozzle in conjunction with the SonicSyringe. Both the SonicSyringe and the nozzle caused further deagglomeration of the active material particles, yielding a uniform spraying distribution of particles, as seen in Figure 7.

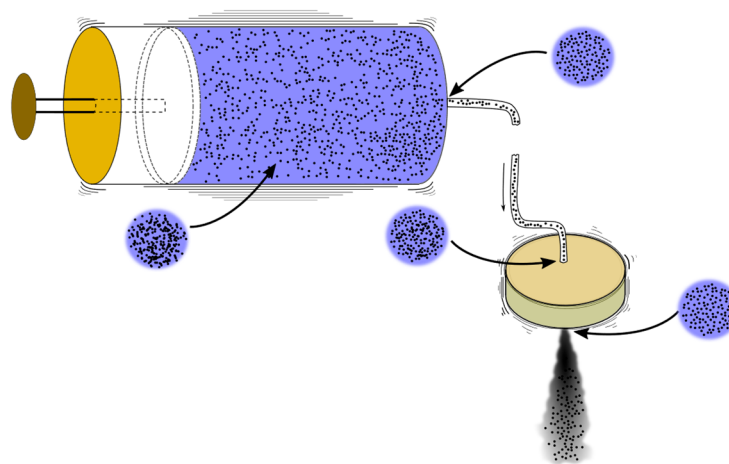


Figure 7. Sketch showing the deagglomeration of particles during ultrasonic vibration inside the head of the nozzle.

The cell was swept over five cycles between 2.5 V and 4.5 V, sweeping at a rate of 0.001 V s^{-1} . The upper and lower voltage limits were chosen to analyze the possibility of unexpected electrochemical reactions unique to an LFP carbon fiber matrix. An initial solid electrolyte interphase (SEI) was seen at 4.25 V during the first cycle, as shown in Figure 8. Following this, the cell stabilized and showed promising reversibility, with the oxidation and reduction peaks being close in magnitude. The increase in current at the peaks for cycles 2–5 showed a predictable increase in resistance as the cell was cycled. Despite stressing the cell with an upper limit of 4.5 V, its stability proves that ultrasonic coating offers competitive lithium-ion diffusion.

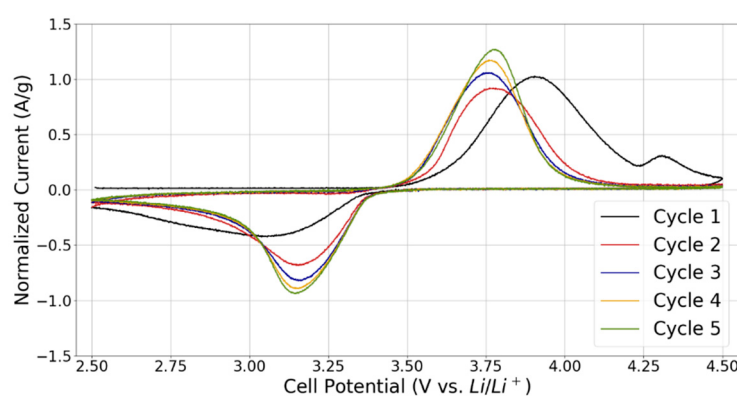


Figure 8. Cyclic voltammetry of an ultrasonic LFP coated half-cell at a sweep rate of 0.001 V s^{-1} between 2.5 V and 4.5 V vs. Li/Li^+ for five cycles.

To test the capacity of the ultrasonic coating, a half-cell was cycled between 2.75 V and 4.0 V, versus Li/Li^+ , at an initial rate of 0.1 C for the first two cycles to promote the formation of the SEI. After this, the current was increased to 0.5 C, and the cell was cycled until the capacity retention dropped below 80%. Figure 9 shows the lithiation and delithiation of the cell across 350 cycles at 50 cycle intervals. Cycle 1 shows the SEI formation, falling below the capacity of the following 250. The cell showed a recovery and increased capacity after the formation cycles, in contrast to the expected behavior of irreversible capacity loss following SEI formation as outlined in ref. [19]. A small amount of lithium ions was consumed during

the SEI formation [20]. However, the significant recovery may be attributed to the excess lithium ions available in the cell due to the LFP-coated electrode providing ions to the lithium disk. The atomized surface coating of the carbon fiber may have allowed for a more complete delithiation cycle, allowing some ions from the LFP to replenish the lithium metal. This process would also contribute to the stability shown by the cell. Following formation, a slow, consistent capacity drop occurred as expected with a tight window in potential between 3.2 V and 3.6 V up to 70 mAh/g for each cycle. This window of constant voltage is caused by the two-phase transition of LFP as lithium ions move across the cell during lithiation and delithiation [21]. Its persistence around 3.4 V again proves the strong stability of the cell.

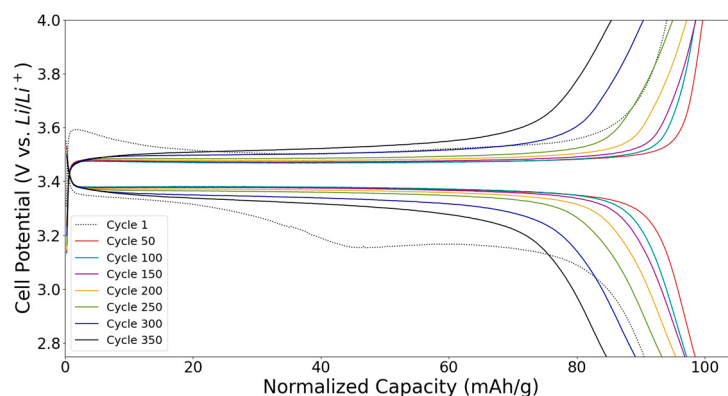


Figure 9. Lithiation and delithiation of an ultrasonic LFP coated half-cell across 350 cycles. An initial formation was performed during the first two cycles using a rate of 0.1 C. Current was then increased across the remaining cycles to 0.5 C.

The capacity drop of the half-cell per cycle is shown in Figure 10a for both the charge and discharge steps in each cycle, going up to 450 cycles following the schedule described above. Using the ratio of discharge to charge capacity for every cycle, the coulombic efficiency was found to remain at nearly 100% for the entirety of the 450 cycles, as seen in Figure 10b. A large drop in capacity can be seen in the initial formation cycles, followed by a quick recovery and stabilization. The half-cell reached a capacity of 99 mAh/g after 100 cycles and 97 mAh/g after 200 cycles before dropping to 73 mAh/g after 450 cycles.

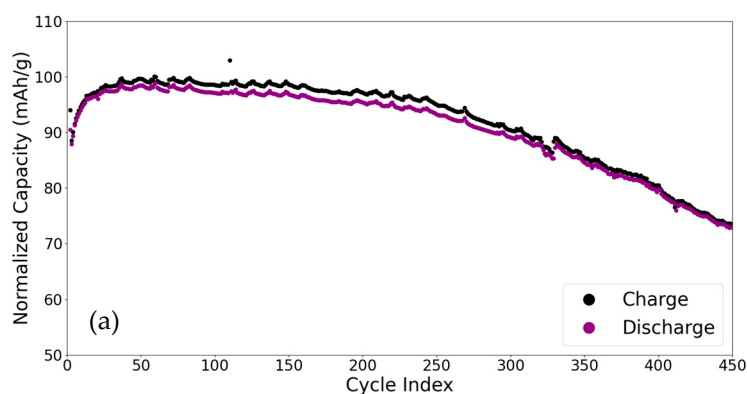


Figure 10. Cont.

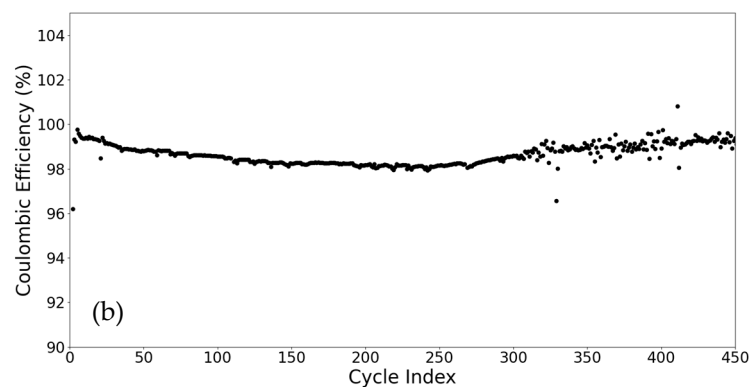


Figure 10. (a) Charge and discharge capacities with (b) coulombic efficiency of an ultrasonic LFP coated half-cell for 450 cycles at 0.1 C for the first two cycles and 0.5 C for the remaining cycles.

To show the effectiveness of ultrasonic coating with the SonicSyringe, a comparison of cell capacity is shown in Figure 11a, which includes other coating methods found in the literature as well as that performed with the regular syringe and dip coating during this study. Ultrasonic coating does not achieve the large initial capacity seen in other coating methods, but it maintains a capacity around 100 mAh/g for many more cycles than other methods explored in the literature. Comparing the capacity retention of each method in Figure 11b, the ultrasonic coating shows superior cell stability, reaching 350 cycles with a retention above 80%. Particle size and uniformity are likely to contribute to the success of the cell. The fine particles produced from the ultrasonic allowed a large active surface area to be available for lithiation, as seen in the SEM analysis. The particle size in conjunction with the uniformity of their dispersion on the carbon fiber may have also increased the stability of the microstructure, causing less severe irreversible deformities during cycling.

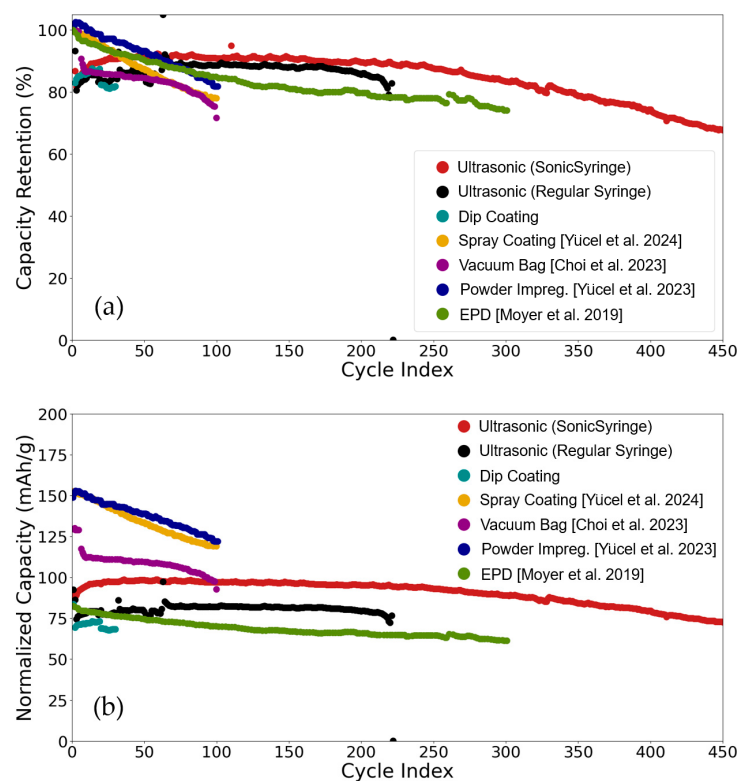


Figure 11. Comparison of (a) capacity retention and (b) capacity for the ultrasonic method against other coating methods found in the literature, including spray coating [18], hot press vacuum bag coating [11], powder impregnation [17], and EPD [22].

4. Conclusions

The ultrasonic spray-coating method presents a significant advancement in the fabrication of structural battery electrodes, offering an economical, scalable, and electrochemically efficient coating technique. By providing highly uniform surface coatings with minimal material waste, this method addresses key challenges in the development of multifunctional energy-storing structures. The integration of a computer-controlled tracing system ensures precise application, reducing variability and enabling large-scale production, a critical factor for the commercial viability of structural batteries. Electrochemical testing demonstrated that ultrasonic spray-coated samples achieved a specific capacity of 100 mAh/gLFP with 80% retention after 350 cycles, surpassing the performance consistency of conventional coating methods presented in the literature. The enhanced stability and uniformity of the coatings indicate that this approach could play a crucial role in advancing structural batteries from laboratory research to real-world applications. While the energy density remains lower than traditional Li-ion batteries, structural batteries offer system-level benefits by replacing passive structural components with multifunctional materials. Among currently explored techniques, ultrasonic spray coating emerges as the most promising for scalable manufacturing. These findings mark a step forward in enabling practical, high-performance structural batteries, paving the way for their integration into aerospace, automotive, and other weight-sensitive applications. Further research and optimization will be essential to fully unlock their potential.

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