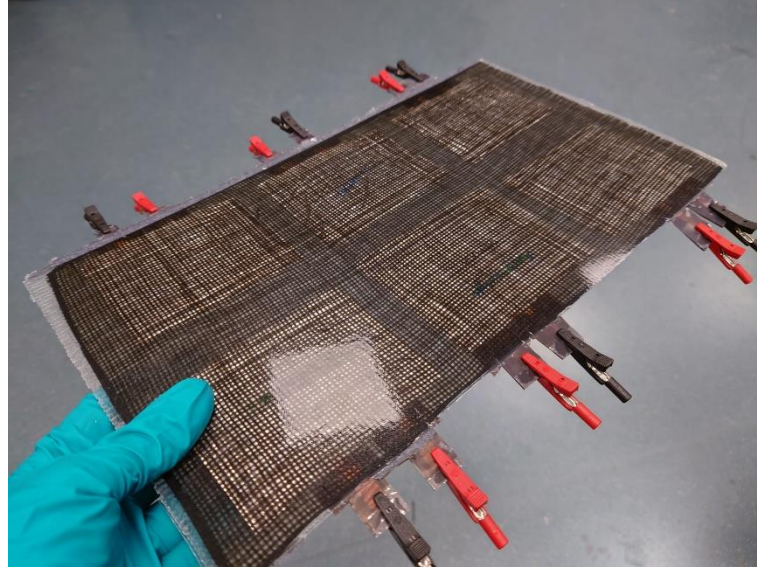




CHALMERS
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Improvements in structural battery cells processing: manufacture, characterization, and multicell demonstration

Master thesis in Industrial and Materials Science

Mohammad Siam Siraj
Samia Tasneem

DEPARTMENT OF INDUSTRIAL AND MATERIALS SCIENCE
Division of Material and Computational Mechanics

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MASTER'S THESIS 2022

**Improvements in structural battery cells processing:
manufacture, characterization, and multicell demonstration**

Final Report

Mohammad Siam Siraj
Samia Tasneem

| siamm@student.chalmers.se
| samiat@student.chalmers.se



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Improvements in structural battery cells processing: manufacture, characterization, and multicell demonstration
Final Report

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Industrial Supervisor: Dr. Fredrik Edgren, Volvo Cars
Chalmers Supervisor: Prof. Leif Asp, Department of Industrial and Materials Science
Examiner: Prof. Leif Asp, Department of Industrial and Materials Science

Master's Thesis 2022
Department of Industrial and Materials Science
Division of Material and Computational Mechanics
Chalmers University of Technology
SE-412 96 Gothenburg
Telephone +46 31 772 1000

Cover: Structural Battery multicell demonstrator.

Abstract

The road to a sustainable future involves the electrification of the transportation industry and one of the greatest challenges in it is electrical energy storage. Lightweight batteries serving dual purpose of storing electricity in structural load paths show great promise as a steppingstone in overcoming that challenge. While previous research successfully reported such multifunctional structural battery cells, the low energy density and lack of a manufacturing method to produce repeatable cells remain major concerns. This thesis introduces a robust manufacturing method comprising of the well-known vacuum infusion of composite materials to produce structural composite cells with repeatable performance. Manufactured cells are also presented in a six-cell structural battery composite laminate demonstrator where cells that can be connected in any desired configuration, performance of three such configurations have been portrayed experimentally. The composite cell components are carbon fiber cathode, lithium-iron-phosphate coated aluminum anode, and a separator material all infused with structural battery electrolyte (SBE) resin. The manufacturing method has a 100% success rate of producing working cells with 70% of them having repeatable and high performance. Cells with Whatman GF/A and glass fiber plain weave separators features an unmatched 30.0 Wh kg^{-1} and 41.2 Wh kg^{-1} energy densities, respectively. The effects of low pressure on SBE as well as the reduction in high state-of-charge surface impedance and charge resistance of individual cells have also been reported. With such performance, these multifunctional materials can reduce the burden from conventional batteries and lighter means of transportation or other goods can be achieved.

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Abbreviations

CF	Carbon Fiber
CFRP	Carbon Fiber Reinforced Polymer
GF	Glass Fiber
PW	Plain Weave
SBE	Structural Battery Electrolyte
SB	Structural Battery
Cu	Copper
Al	Aluminum
LFP	Lithium Iron Phosphate
Li	Lithium
EC	Ethylene carbonate
PC	Propylene carbonate
LiBoB	Lithium bis(oxalato)borate
LiTFS	Lithium tri-fluoromethanesulfonate
AIBN	Azobisisobutyronitrile
EIS	Electro-chemical Impedance Spectroscopy
GCD	Galvanostatic charge and discharge
FR	Freudenberg
FTIR	Fourier-transform infrared
SOC	State of Charge
BMS	Battery Management System

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1. Introduction

1.1. Background

Multifunctional lightweight composite materials pose an attractive research area. These novel materials have a huge potential to tackle some of our great challenges, particularly the problems related with minimizing environmental impact and maximizing energy efficiency in transportation by reducing weight of a vehicle. One such idea is materials which can store energy at the same time provide structural support by itself. The potential advantage of these “mass-less” energy storage is very attractive and can revolutionize future lightweight transport designs [1].

Structurally strong batteries that use solid-state electrolytes and load carrying electrodes and separators have not been widely researched. Recently, a structural battery (SB) composite with multifunctional performance has been demonstrated but still is in a very early state of research. Presenting the structural battery in a real-world use will open countless possibilities and accelerate research in this area, thus, making a demonstrator showcasing its multifunctional properties is important. The ambition is to develop a road path for lighter electrical cars, aircraft, and consumer goods equipped with these materials for the future [2].

1.2. Aim

This thesis aims to develop a reliable manufacturing process of state-of-the-art structural battery cells and to facilitate demonstration of the multifunctional use of structural batteries in an automotive application. The structural batteries will power an electrical equipment within a car door while providing mechanical support to all components behind the door pad. The scope of this thesis includes the following:

- Manufacture structural batteries using a robust manufacturing technique that produces cells with repeatable performance.
- Electro-Chemical and Mechanical characterization of the cells
- Build a laminate as a multicell structural battery demonstrator and assess its applicability
- Documentation and presentation of the findings

1.3. Tasks/ Deliverables

- A verified/proven method for cell manufacturing.
- A small-scale model of the automotive power window elevator system solely powered by the manufactured structural battery.

1.4. Ambitions

As a stretched ambition of the project, cell manufacturing steps was focused on automation and mass-production of the Structural Battery.

1.5. Limitations

Cell performances were based on the cell manufacturing methods introduced in this paper. Fixed cell dimensions were used for the samples, therefore, variations due to cell sizing were not investigated. Performance variables due to the state-of-the-art cell components (electrodes, separator, structural battery electrolyte) were not assessed in this research. Study of other key factors such as recyclability, durability, and safety aspects of the Structural Battery are out of the scope of this research project. Given the short time period of the thesis work, thermal aspects of the cells were not investigated.

2. Literature Review

2.1. Conventional lithium-ion batteries

Lithium-ion batteries have been used to store energy for the past two decades. Starting with its application in small electronic appliances such as mobile phones to its application in much bigger context such as electric vehicles, the Li-ion batteries have come a long way in these recent years.

A Li-ion cell is made up of a positive electrode (cathode), a negative electrode (anode) and an electrolyte consisting of lithium ions. A typical cell would have LiCoO_2 or LiFePO_4 as the cathode material and carbon as anode material [3]. The cathode and the anode need to be electrically insulated from each other by a separator. Most common separator is a microporous polymer membrane which allows the flow of the Li-ions. Figure 1 shows the components and operation principle of a cell.

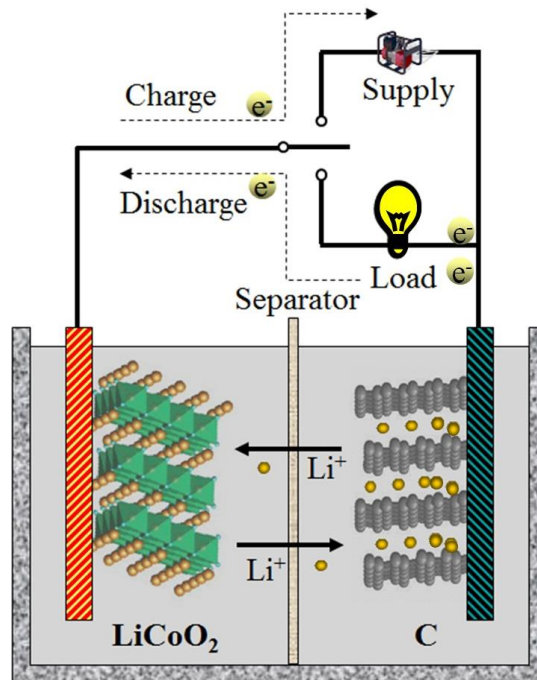


Figure 1. Working principles of a Li-ion battery [3]

During charging, the cell is connected to an external circuit with electric supply. The charging of the cell will cause the release of electrons at the cathode which flow in the external circuit to the anode. At the same time, the Li-ions move from the cathode to the anode via the electrolyte and are inserted in the carbon layers. The external electrical energy is thereby stored as chemical energy which can be used during the discharging. The electrons move back to the cathode through the external circuit providing the extra energy to generate power while the Li-ions move back through the electrolyte as well. The whole process involves chemical redox reactions which allow for the storage of electrical energy [3].

While Li-ion batteries are successful commercially today, scientists are still studying for ways to increase the energy density of the cells. The mass of the batteries compared to their capacities remain a major concern in the automotive and aerospace industry. Structural batteries seem to be the right step towards solving the issue by being a “mass-less” storage system.

2.2. Carbon fiber energy storage systems

The concept of structural battery is based on the idea of saving mass by integration of the load carrying property of a material with its energy storage property. The multifunctional performance of the battery can be judged by using the ratio of the saved mass to the total mass of a conventional battery mass having mono functionality. Carbon fibers that are sold commercially nowadays can be used for such multifunctional performance.

Research done on the positive electrode has shown potential of using carbon fibers coated with LiFePO_4 powder while using Polyvinylidene fluoride as binder. Carbon black has been also used for enhancing the electrical contact of CF with LiFePO_4 . This results in use of CF for both electrodes making the battery multifunctional [4].

Structural CF batteries have a common design principle which they share with conventional Li-ion batteries as shown in Figure 2. Differences between them include having CF as the negative electrode and possibly as coated positive electrode both enclosed in an electrolyte and solid matrix for ion conductivity and mechanically superior performance. Electrically insulating separators for SB can be made of glass fibers which contributes to the mechanical properties of the cells [5].

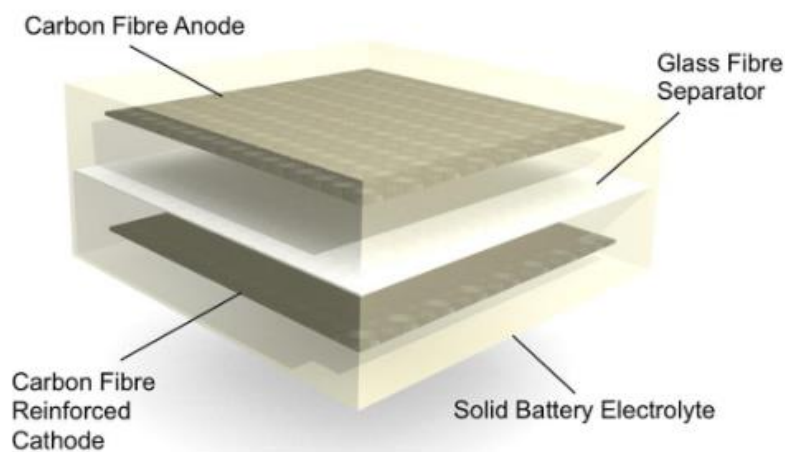


Figure 2. Structural composite battery schematic diagram [5].

Secondary Li-ion batteries have been using carbon in many graphitized forms since the beginning of the research in this sector. The property of graphite being able to intercalate lithium ions in its layers is the main reason why it is used in such application. This property along with the high specific mechanical properties make carbon fibers a suitable choice in

the manufacture of the structural battery composite layers. The electrochemical and mechanical properties of commercial carbon fibers have been researched and it was proved promising to be used as multifunctional composite material in the structural battery laminate [1].

The elastic modulus of carbon fibers has a wide range starting from 200 GPa to 600 GPa. Intermediate Modulus (IM) fibers (IMS65 and T800) have been found to have better electrochemical properties and their modulus ranges from 300-400 GPa. High modulus fibers, on the other hand, were having lower capacity which is due to the partial obstruction of lithium staging by the turbostratic graphitic structure [5].

The lithiation and delithiation of the fibers researched do not affect the tensile stiffness but does reduce the ultimate tensile strength. First lithiation reduces the strength by 20% but was observed to be recovered partially during delithiation resulting in a loss of 10% strength when compared to a pristine fiber. The fibers also swell during the charging in both axial and radial direction. In the axial direction, the swelling went up to 1% when the theoretical capacity limit was reached. Fully charged fibers showed swelling up to 8% in radial direction. More research needs to be conducted in order to understand the effect of swelling and shrinkage of the fibers during the electrochemical cycling [5].

2.3. Structural battery electrolyte

Structural battery requires an electrolyte which can perform mechanically as well as electrochemically. Traditional batteries use liquid electrolyte which cannot carry mechanical loads. To form a structural battery, it is important for the electrolyte to transfer mechanical loads between fibers along with conducting the ions. An electrolyte for this purpose has been developed which has multifunctional matrix and is called Structural Battery Electrolyte (SBE) [6].

It has been a challenge to develop a matrix which has the mechanical and electrochemical properties simultaneously. Better mechanical properties would result in poor electrochemical properties and vice versa. For a fiber composite to have a high performance in terms of mechanical strength, it is essential to have good adhesion between fibers and matrix. On the other hand, ion mobility at the fiber/SBE interface is a must to allow electrochemical reactions take place resulting in a demand for liquid electrolyte to reach the surface of the carbon fibers. Consequently, the fiber surface must be in contact with both the solid polymer phase and liquid phase to have the multifunctional properties [7].

Schneider et al. [6] first introduced such a matrix which fulfilled the requirement of the SBE. Conventional liquid electrolyte with the presence of Li salt was mixed with bis-phenol A based monomer which is also commonly used in the composites industry. Thermally induced polymerization was achieved by heat curing the mixture at the range of 80-90°C forming a structure with two percolating phases. The SBE developed was proven to have both the desired multifunctionality with 61 wt% of solid matrix and 39 wt% of liquid electrolyte flowing through it. The modulus of the SBE was reported around 500 MPa at ambient temperature with an ionic conductivity of $2 \times 10^{-4} \text{ S cm}^{-1}$. To avoid side reactions, SBE was kept in inert and dry environment in all steps [6]. This thesis used the

composition used in the research paper by Schneider et al. [6] and Asp et al. [2] which was proved to have the best results in terms of electrical conductivity and mechanical performance.

2.4. Electrochemical characterization and properties

Structural battery cell multifunctional properties have been demonstrated in a recent study. Carbon fibers were used as the negative electrode and LFP foil was used as positive electrode with a glass fiber separator in between. The cell was cured in SBE resulting in forming a SBE matrix holding the cell together. Two different GF separators were tested, GF plain weave and Whatman GF/A. [2]

Constant current was applied to the cells for galvanostatic cycling of the cells. Choice of current was based on the aim to reach a specific charging time. Longer charging time (20 h or more) required a very low current which leads to reaching the maximum capacity of the cells. Shorter charging times, on the other hand, can be used for looking into the resistivity and transport limitations of the cells. With a discharge time of 20 h or more (a C rate of 0.05 C), the cells with GF plain weave were found to have energy density of 23.6 Wh kg⁻¹ while the Whatman GF/A cells had 11.6 Wh kg⁻¹. The below table shows the specific capacities and energy densities of the cells:

Table 1. Electrochemical properties of structural batteries [2].

Separator	Whatman GF/A		GF plain weave	
	Battery cell	Active mat.	Battery cell	Active mat.
Specific capacity [Ah kg ⁻¹]	4.13	32.2	8.55	38.4
Energy density [Wh kg ⁻¹]	11.6	90.1	23.6	106
Calculated maximum energy density [Wh kg ⁻¹]	37.7	220	60.6	220
Specific power [W kg ⁻¹]	5.94	34.7	9.56	34.7
Total mass of cell [g cm ⁻²]	0.074		0.046	
Cell thickness [mm]	0.40		0.27	

Electrochemical characterization of the cell with Whatman GF/A separator is shown below:

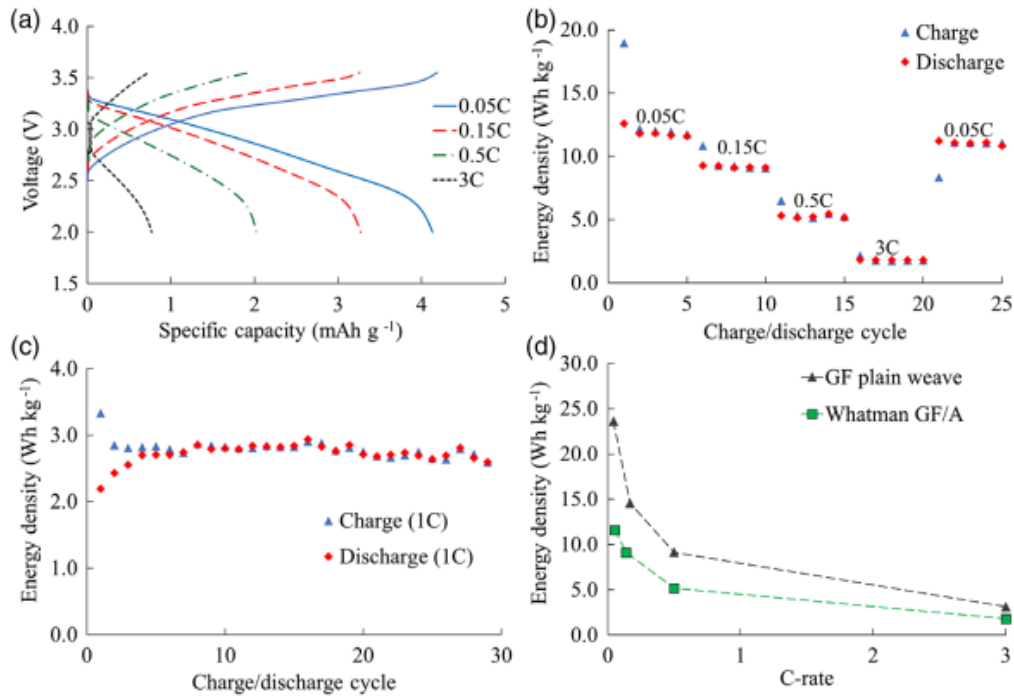


Figure 3. Electrochemical results of structural batteries as studied by Asp et al., 2021 [2]

As shown in Figure 3.a, a stable charge discharge profile is visible from the voltage profiles at different C rates. At different C rates, the charge discharge cycles with energy densities show that after many cycles, the capacity of the cell does not fade. Coulombic efficiency is observed in Figure 3.c where high-capacity charge retention can be observed at 1C. Lastly, it can be observed that GF plain weave has significantly higher energy density compared to the thicker Whatman GF/A cell.

The limitation for structural batteries is mainly the mass of active materials compared to the total mass and the amount of SBE when it is added manually. This thesis involved a major change to this method of manufacture to ensure even SBE spread out in order to maximize the capacity of the cells.

2.5. Mechanical characterization of composite energy storage systems

Mechanical properties of the full cell were also characterized in the same research as above. Tensile loading in both x and y direction was used. The Figure 4 below shows the load displacement curves for three samples: Whatman GF/A, GF plain weave with fibers extending in $\pm 45^\circ$ and GF plain weave with fibers extending in $0^\circ/90^\circ$.

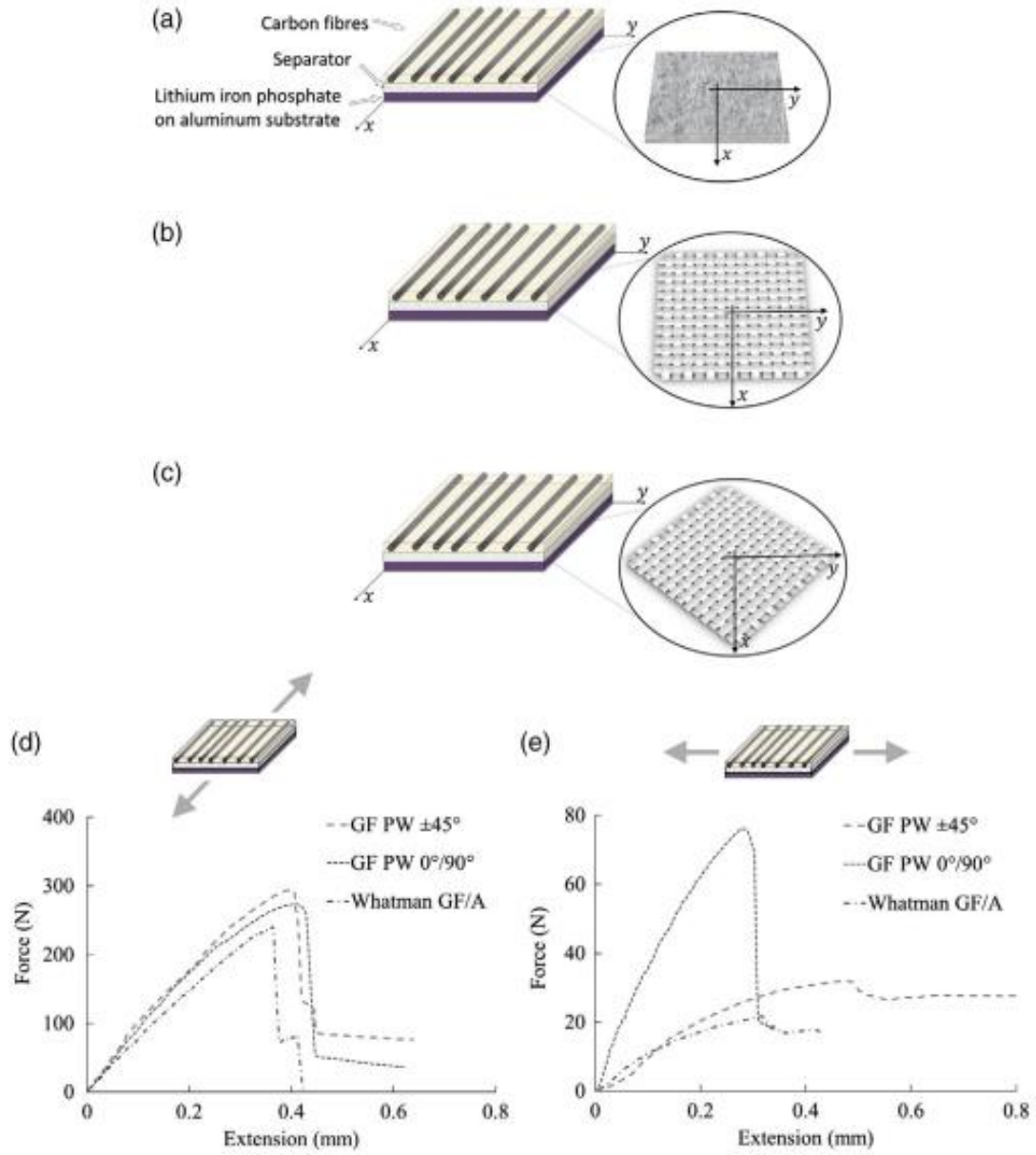


Figure 4. Structural battery separator layups and their mechanical responses as studied by Asp et al. [2].

As shown in Figure 4 above, highest modulus is in the x direction for all structural battery composites. This is because the CF as the negative electrode have a parallel orientation to the loading direction. Table 2 reports the mechanical properties of the cells.

Table 2. Mechanical properties of the latest structural batteries. [2]

Property	Separator type		
	Whatman GF/A	GF plain weave $\pm 45^\circ$	GF plain weave $0^\circ/90^\circ$
E_x [GPa]	18.3(± 0.9)	14.6(± 0.6)	25.4(± 3.3)
E_y [GPa]	2.9(± 0.5)	2.8(± 0.2)	13.3(± 0.7)
X [MPa]	>163	>312	>287
Y [MPa]	>16	>34	>72

The GF plain weave in $0^\circ/90^\circ$ has the highest modulus which is due to the fibers extending in the 0° direction. Both CF and GF fibers reinforce to provide the maximum amount of load carrying ability. On the other hand, it is observed that modulus in y direction is comparatively lower.

Based on the table above, if Whatman GF/A was replaced with a thinner and more mechanically stiff separator, the multifunctional performance of a structural battery cell is believed to be enhanced significantly. This thesis used Freudenberg separator in addition to the separators mentioned here to judge their multifunctional performance.

2.6. Composite cell manufacturing techniques

Carbon fiber composites are usually made with thermoset polymers for their superior mechanical performances. Thermosets are basically polymerized and stiffened by heating. This polymer matrix reinforced by carbon fibers are done by impregnating some sort of monomer and hardener mixture in their liquid phase into the fibers. An external pressure is required for keeping the fibers and matrix closely, simultaneously heat is also applied. This starts the curing process which polymerizes the monomers. The result is a Carbon Fiber Reinforced Polymer (CFRP). State-of-the-art composite structural batteries are being produced by researchers based on this principle.

Composite structural cell manufacturing techniques did not reach the full-fledged industrial mass production stage unlike the conventional Li-ion batteries. The multifunctional cell manufacturing is still very immature and done in research laboratory. A manual SBE wetting method studied at Chalmers University of Technology, currently produces CF cells with the highest specific capacities [2]. The process includes an assembly of cell components within a pouch bag enclosure which is then seal on the current collector end to keep the components in place, two sides of the pouch are kept open for drying and SBE addition. The assembled cells are then dried for a long period of time before they are transferred into an inert chamber for SBE wetting of the cells, after which the pouch bag is vacuum heat sealed. The process is shown in Figure 5.

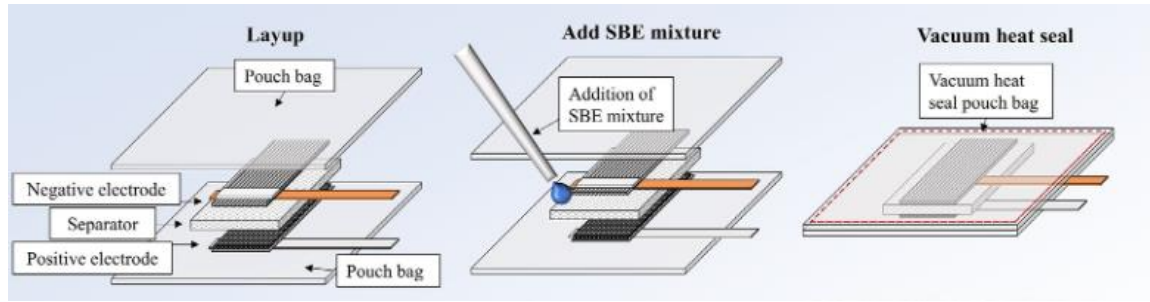


Figure 5. Cell assembly, SBE wetting, and pouch bag sealing of structural battery [2]

The sealed pouch cell is then taken out of the glovebox and cured at 90°C for 1 hour. Since an external pressure is required, simple plate clamping is used for the purpose as shown in Figure 6. After the curing process, the pouch cell is ready for electrochemical testing. Mechanical tests were done on cured cells without the pouch bags [2].

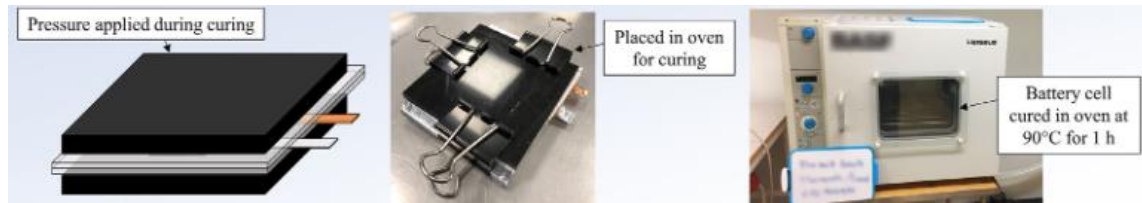


Figure 6. Curing process of structural batteries by Asp et al., 2021 [2].

Another study done at KTH Royal Institute of Technology has demonstrated a modified version of the well-known vacuum infusion method for wetting the carbon fiber electrode assembly. They used thin carbon fiber bundles as both electrodes for their structural morphing material with copper current collectors attached to the fibers with silver conductive glue. The process involves firstly placing the components in place on a flat mold. The assembly is covered with a kind of non-stick film and a distribution layer. Then the sample is vacuum bagged and sealed using vacuum sealing tapes. Both, the vacuum infusion method SBE was prepared in a dry and inert environment. Both UV and heat curing were investigated. The resulting composite was transferred into pouch bags to avoid side reactions of SBE [4].

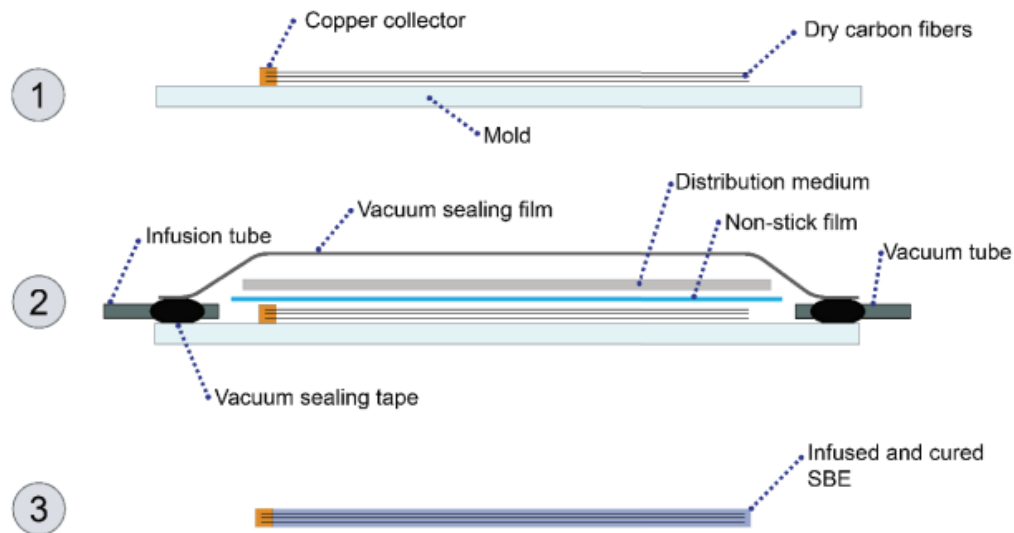


Figure 7. Vacuum Infusion for making shape-morphing carbon fiber actuator [4]

Though the vacuum infusion method was introduced for manufacturing carbon fiber structural morphing material, it does provide the pathway of adopting this manufacturing method for producing composite energy storing full cells [4, 8].

3. Methodology

This section explains the choices and manner of the tasks performed motivated by the issues faced during the research project. Project started with manufacturing CF structural cells using *vacuum infusion* which was modified and further developed in this research.

As one of the main objectives, a robust manufacturing method was the key focus. Several iterations of the processes were carried out until the process reached a stage where it produced 100 % working cells with repeatable cell results. Several factors were considered before the the manufacturing setup such as the reactivity of the SBE with air, amount of chemical waste, and possible handling contaminations of cell components.

The general well known procedure of vacuum infusion setup of composite materials was adopted and modified according to the above factors. To start, a precise sizing of cell component followed by a careful assembly was important for repeatable cells and eliminating short circuits. Reliable vacuum bagging of the cell assembly along with all other infusion equipment needed proper drying before the infusion could take place. Since the SBE resin must not come in contact with air [7], its preparation was done in an inert argon gas environment inside a glovebox and transferred into a syringe as a container of infusable substance. Eliminating the use of glovebox for the infusion process was a major challenge. A fully sealed infusion setup was designed (section 3.3.2) that could be carried outside the glovebox after the prepared SBE solution was incorporated into the infusion setup. For this, the vacuum bagged assembled cells would have to be taken in and out of the glovebox for sealing the setup using clamps. This meant the SBE infusion of the cells could be done outside and, as a result, reduce the complexity of setting up a vacuum infusion system inside the glovebox. After the SBE is infused under low pressure, the cell is cured in an oven. Debugging is done by again taking the cured cells inside glovebox where the cells can be tranferred into a pouch bag before taking out for cell conditiong and testing.

The entire thesis methodology can be summarized in a flow chart shown in Figure 8 below. The figure shows the main steps in chronological order, separately identifying in blue boxes the processess done in inert environment. The detailed methods of manufacturing and testing are explained in later sub-sections.

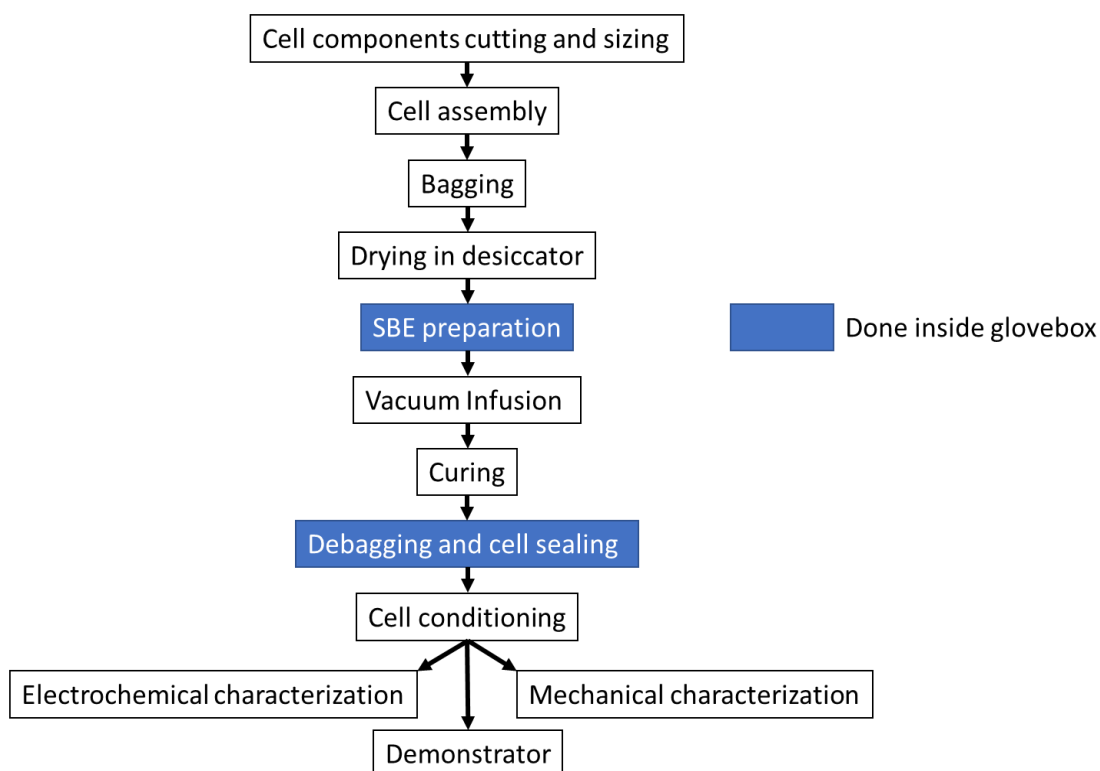


Figure 8. Flowchart of thesis methodology. Steps marked with blue boxes were carried out under inert environment (inside a glovebox)

3.1. Materials and equipment

The materials chosen for the different components of the cells are based on literature studies [2, 4, 7, 8]. State-of-the-art materials, and the equipment used in this research have been divided into subcategories.

Anode

Negative electrode materials:

- TeXtreme® T800S unidirectional Carbon Fiber is used as the negative electrode. The CF tow used has a fiber count of approximately 12,000 and 1.4 cm to 1.35 cm of tow width.
- Copper current collector (Cu foils from Sigma Aldrich).
- PELCO® conductive silver paint from TED PELLA INC for binding current collectors with the electrodes.

Cathode

Positive electrode materials

- Commercial Lithium Iron Phosphate (LFP) coated on Al foil is used as the positive electrode. Rated at 1.0 mAh/cm².
- Aluminum current collector (kitchen Al foil)

SBE

SBE constituents:

- Bisphenol A ethoxylate dimethacrylate (monomer)
- Heat curing initiator Azobisisobutyronitrile (AIBN)
- Lithium tri-fluoromethanesulfonate (LiTFS)
- Lithium bis(oxalato)borate (LiBoB)
- Ethylene carbonate (EC)
- Propylene carbonate (PC)

Separator materials

Three different types of electrode separators were used in the manufacturing of cells and their performances were investigated:

- Whatman GF/A (Micro glass fiber separator)
- Glass Fiber (GF) plain weave (PW) 25 g/m²
- Freudenberg battery separator (23 μm thickness)

Structural battery demonstrator laminate

Materials used for the layups of the laminate:

- 400 and 90 gsm 0/90° glass fiber weaves
- Bcomp ampliTex 200 gsm 0/90° low twist flax fiber weave
- RAMPF EI-2500 Epoxy
- RAMPF EH-2970-1 Hardener

Equipment for cell manufacturing

Separate sizing and handling equipment were used for the two electrodes and electrode components, and another set was used for general purposes. This was done to avoid mixing of electrode material dust or particles with each other and also to avoid contaminations of cells. This separation of handling and cutting equipment has proven to reduce the short-circuit problems faced during the initial period of the research project. A full list of equipment can be found in the appendix section A.2.

Electrochemical testing hardware

Galvanostatic charging and discharging, and Coulombic efficiency tests were done using Neware Battery Tester. Gamry Multichannel Potentiostat was used for the EIS and the battery pack testing for the demonstrator laminate [2, 9].

Mechanical tester

The mechanical testing is carried out using a Deben Mechanical Tester [2].

3.2. Structural battery electrolyte (SBE) preparation

Since the SBE used contains Li salts, to avoid side reactions, preparation of SBE is completely carried out in an inert and dry environment (inside the glovebox at conditions less than 1 ppm O₂ and 1 ppm H₂O). Firstly, the liquid electrolyte was prepared in large quantity and the same recipe was used for all electrolyte production throughout this project. The liquid electrolyte consists of EC and PC (50:50 wt %) with LiTFS and LiBoB were dissolved in the correct molar ratio of 1 M. The exact composition of each batch of electrolyte prepared is given in appendix section A.3.

For making the SBE, small quantities of this electrolyte were then used and mixed with monomer in equal amounts (50:50 wt %). The monomer stored outside glovebox in a fridge were vacuum dry cycled 3 times before bringing it inside. Heat curing initiator (1 wt % of monomer) was added to the mixture and stirred to trigger the reactions. To keep the chemical waste to a minimum during infusion, some adjustments were made to the infusion setup (section 3.3.2).

After mixing, the SBE samples were degassed at a low pressure (-0.2 bar Δ pressure). This was done to speed up the degassing of SBE. To do this, the sample container lid was replaced with a perforated lid and the sample was then placed inside the transfer chamber of the glovebox where the pressure could be controlled in an argon environment. Three minutes was sufficient for the sample to be completely bubble free. The sample was then brought back into glovebox and transferred into a syringe which then was ready for incorporating with the infusion setup. Process of SBE transfer into a syringe and its integration into the infusion setup is shown in Figure 9. A full process summary is given in Figure 10.



Figure 9. SBE transfer into a syringe (left). Integrating the syringe into the infusion setup (right).

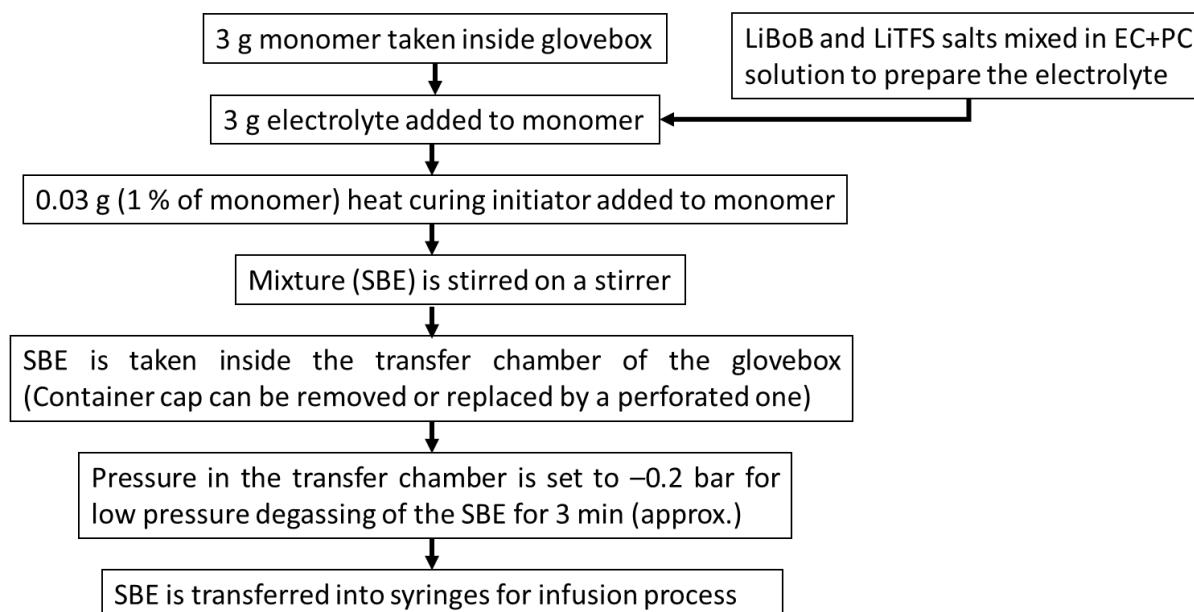


Figure 10. Flow chart of the SBE preparation process.

Fourier transform infrared (FTIR) spectroscopy for chemical composition

A Fourier transform infrared (FTIR) spectroscopy was done on the SBE samples to check if there were any changes to the constituents of SBE because of low pressure degassing. A batch of SBE was prepared and divided into two samples. One was degassed at -0.2 bar pressure difference and the other was not. Both samples underwent FTIR spectroscopy and while testing, they were exposed to moisture. There was no way to avoid contamination at the time of testing.

The instrument used for this spectroscopy includes Bruker Hyperion3000 microscope with a Bruker Vertex70v spectrometer. The sample droplets were added to the attenuated total reflectance (ATR) accessory with a diamond crystal. Reference scans were first run

followed by scans for the samples. The spectrum data collected was analyzed to see the difference in the two samples.

3.3. Cell manufacturing

Cell manufacturing in this project involved assembly of cell components, making of the SBE, vacuum infusion of SBE into the cells, curing, followed by sealing processes of the cured cells. Cell components were assembled, vacuum bagged and dried in an oven at 50°C 14 hours approximately (overnight) inside a desiccator. Electrolyte, monomer, and initiator needed to be mixed in an oxygen and moisture free environment (inside glovebox) then taken out in a sealed manner for the infusion process outside the glovebox. After infusion, the cells were cured at 90°C for 2 hours. Cured cells were then taken inside glovebox and sealed in a pouch bag (PET: Al: PE, 12 μm : 9 μm : 75 μm thickness) as used by Xu et al. [7].

3.3.1. Component sizing and assembly

The components were assembled on a glass plate which was also used as the mold for the cells. Plain release plastics were taped on the glass plates (instead of any release chemicals to avoid contaminations) over which sized components were laid. Starting with cathode, followed by separator (4 cm $\times$ 6 cm) and then the anode is positioned with visual accuracy. The current collectors are covered with release plastic caps to avoid SBE from reaching them which are also taped with the glass plate to avoid movements of electrodes as seen in Figure 11.

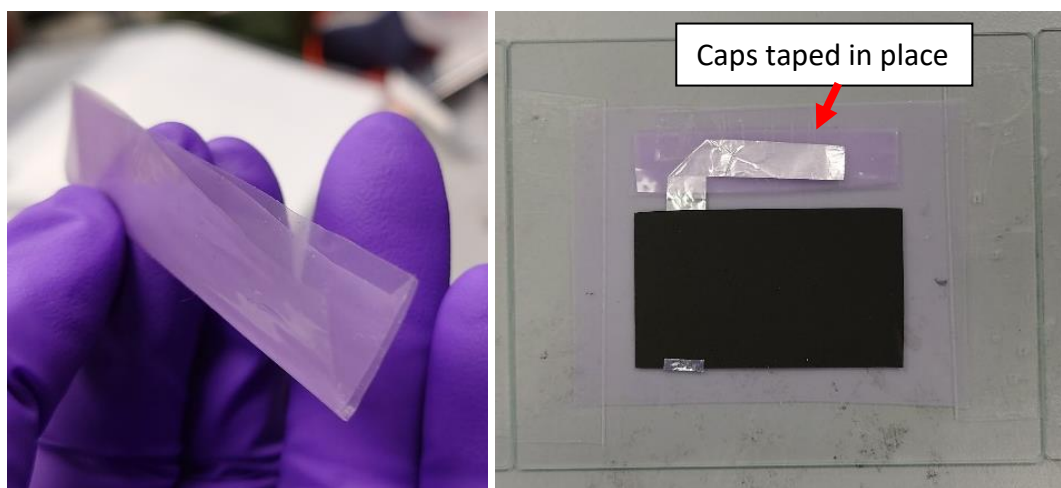


Figure 11. Release plastic caps for current collectors (left). The caps covering cathod current collector taped in place

Placement of the anode and cathode was very important for consistent performance of the manufactured cells. The CF anode was given extra importance because of the loose flying

fibers which needed to be checked for eliminating any possibilities of short circuiting. Figure 12 shows an assembled cell which used glass fiber plain weave as the separator material.

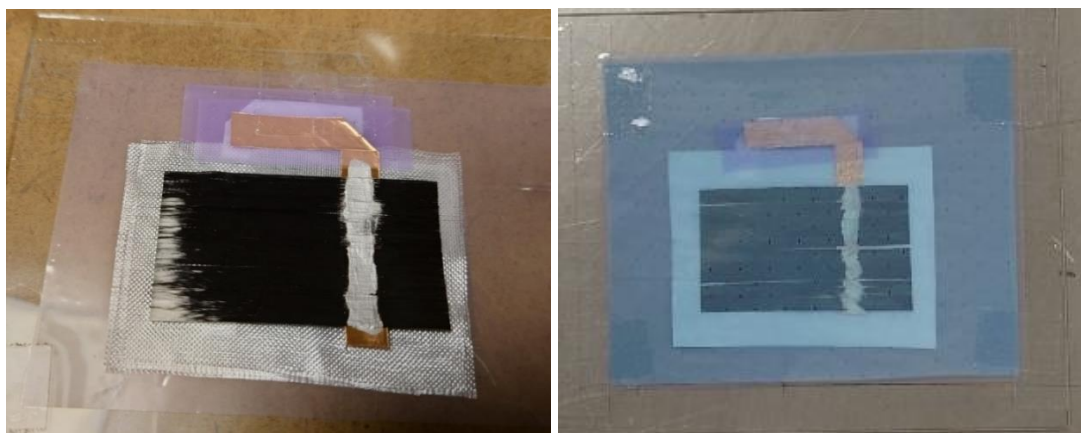


Figure 12. Assembled cell. An example of glass fiber plain weave separator cell assembly (left). Perforated release plastic taped tightly over cell assembly (right)

Assembled cell components are then covered with perforated release plastic followed by a layer of peel ply for infusing SBE. At early stages of this work, samples with additional breather fabric layer or infusion mesh layer were also used but were disregarded because of high chemical waste and no difference in cell performances; only advantage was they reduced the infusion times per cell. A schematic diagram of the full cell assembly can show the layups for vacuum infusion more clearly as in Figure 13.

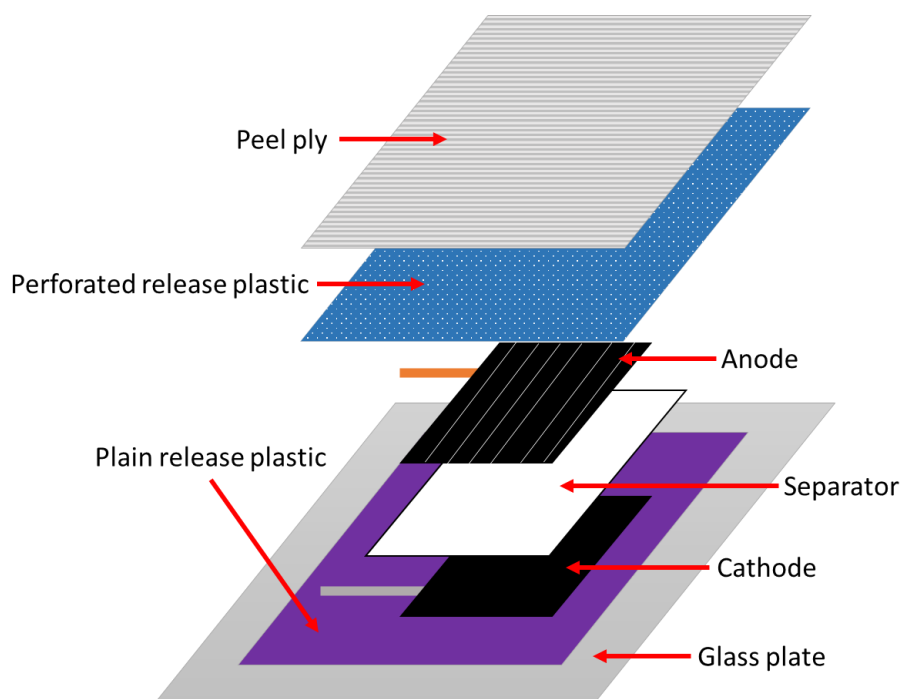


Figure 13. Schematics diagram of the full cell assembly for vacuum infusion.

Anode

Two CF spread tows were aligned in parallel on a glass plate to give a width of 2.7 cm to 2.8 cm and taped in place. Copper current collectors were placed underneath the tows and then bonded to the fibers using a conductive silver paint before cutting the to a length of 5 cm as seen in Figure 14.

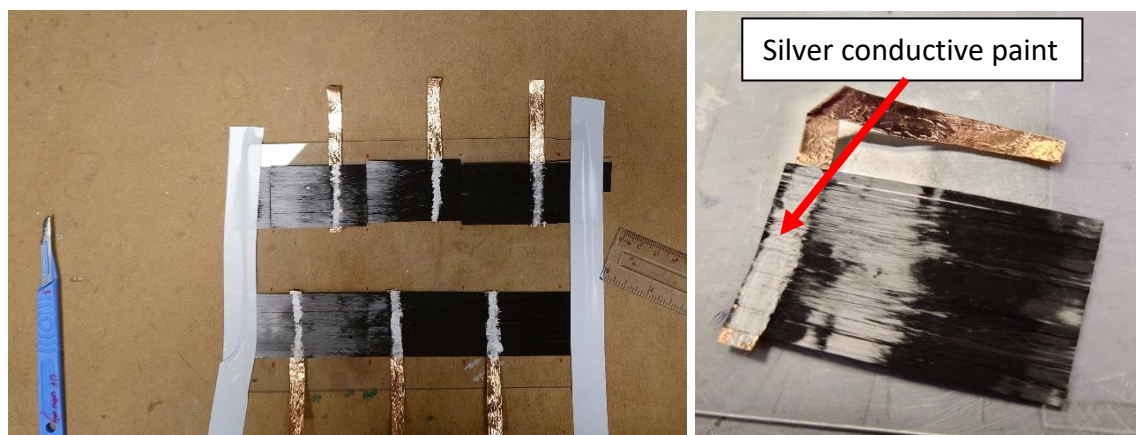


Figure 14. CF electrode assembly process (left), CF electrode assembly (right)

The schematic diagram of the CF anode is given in Figure 15. It shows the sizing of the anode and its assembly used for all the cells manufactured in this study.

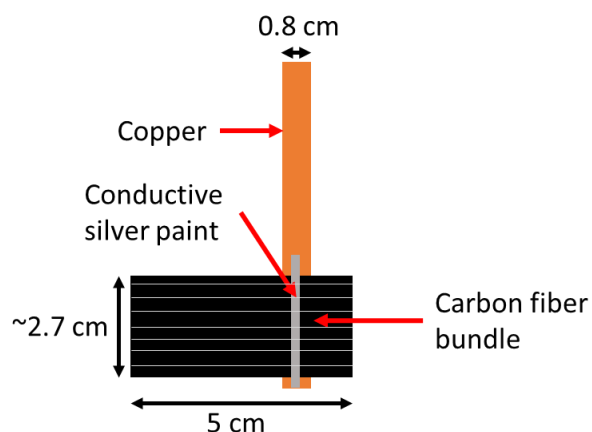


Figure 15. Anode assembly and sizing

Cathode

LFP coated foils were cut to match the size of the CF electrodes (2.8 cm × 5 cm). For fixing the Al current collectors with the cathodes, they were taped using scotch tapes on back (Al side) of the cathode foils and folded around the cathodes a little for more reliable integration (Figure 16).

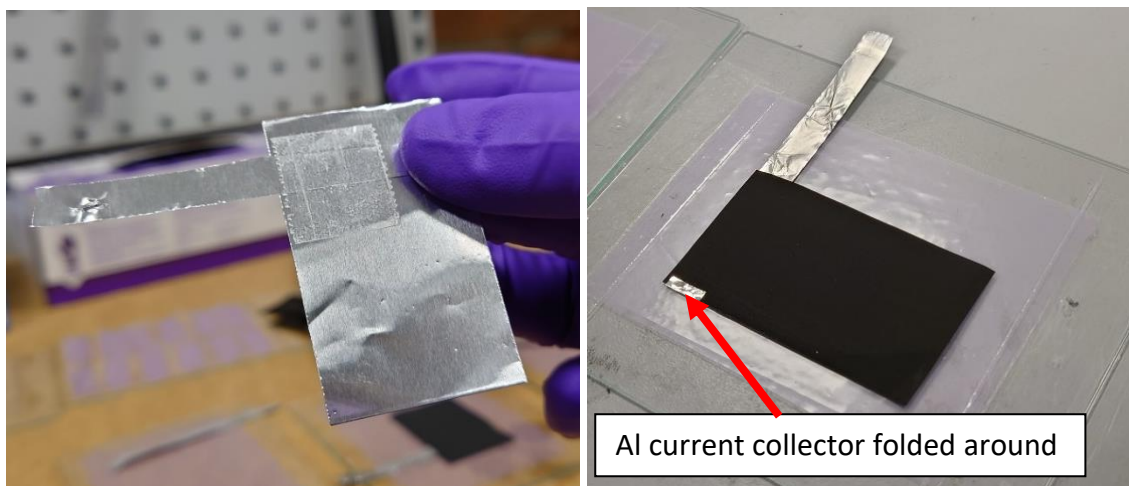


Figure 16. Methods used for securing current collectors with cathode foils. Scotch taped current collector with LFP foil back side (left), and folded edge of current collector around the LFP foil for extra anchoring (right).

The schematic diagram of the LFP anode is given in Figure 17. It shows the sizing of the cathode and its assembly used for all the cells manufactured in this study.

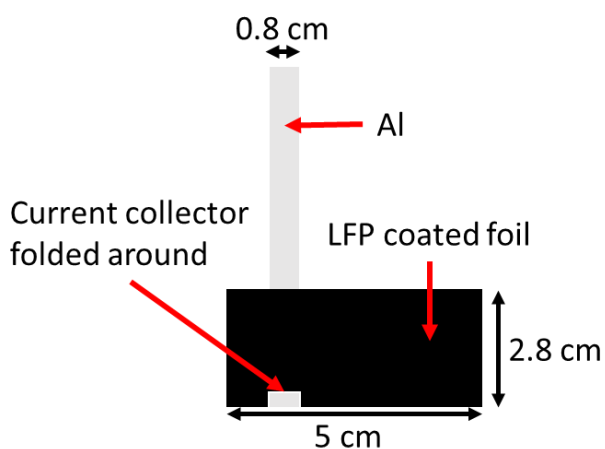


Figure 17. Cathode assembly and sizing

3.3.2. Vacuum infusion

The general infusion technique was used as means of adding SBE resin into the cell assembly. All vacuum bagged dry cell assembly had 4 mm silicone tubes as inlet and outlet with clamps as seen in Figure 18. This simple system can be sealed using the clamps on both sides once the syringe with SBE was integrated.

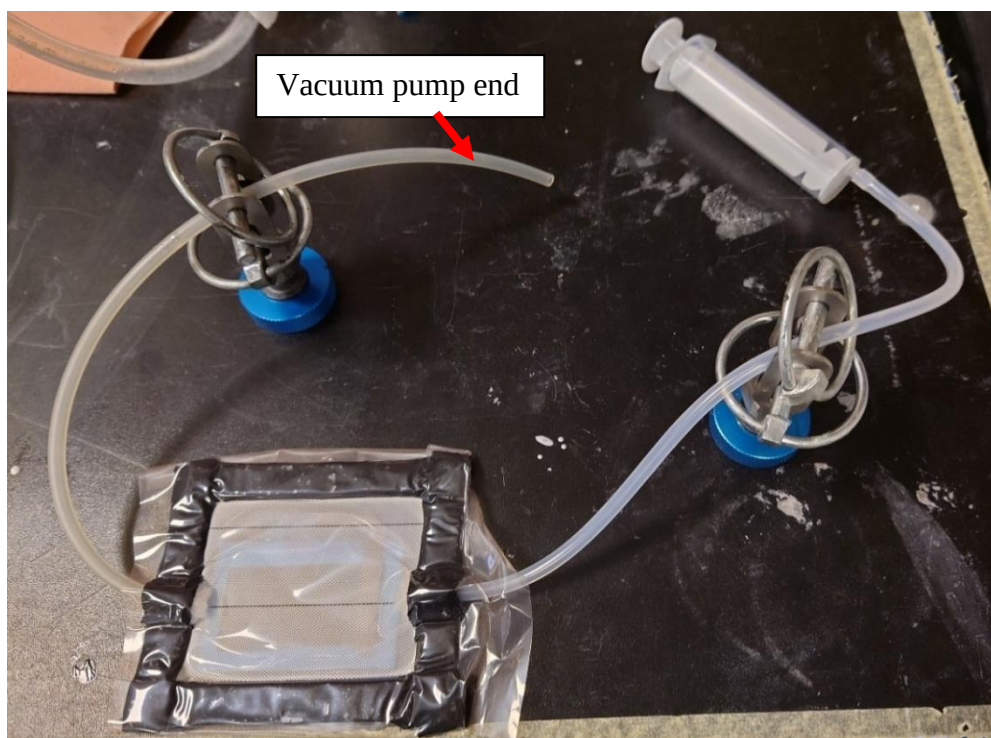


Figure 18. Sealed Vacuum infusion setup.

Infusion of SBE solution was carried out at -1 bar and -0.5 bar pressures, the latter giving superior results though the infusion time was much longer (approx. 25 min per cell). Pressure was regulated using a vacuum valve (Figure 19). Once the pressure was set, clamps were opened (outlet first) slowly to start the infusion process. At the end of infusion, clamps were closed (inlet first), the system is detached from the pump and left in oven for curing.

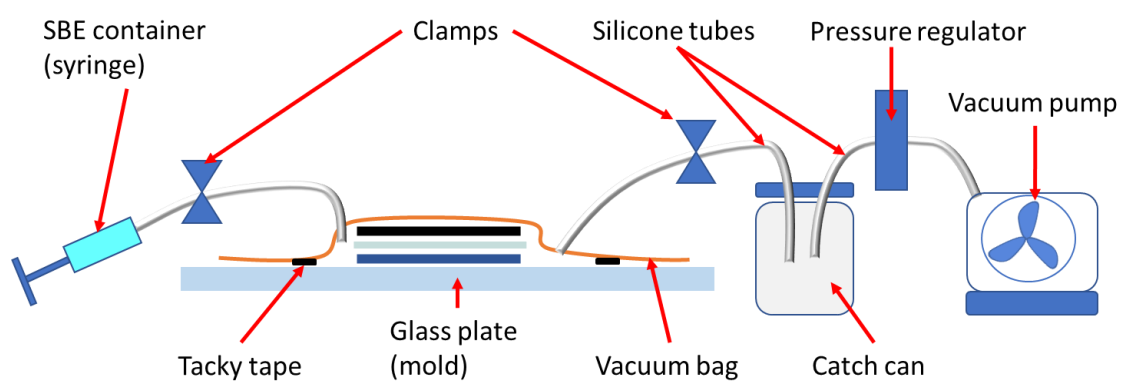


Figure 19. Schematic diagram of the full infusion setup.

3.3.3. Cell sealing

Sealing of cured cells was done inside a glovebox. The full setup shown from Figure 18 are transferred from the oven to the glovebox after the curing process where the cell is debagged from the vacuum bag and transferred into a pouch bag as seen in Figure 20.

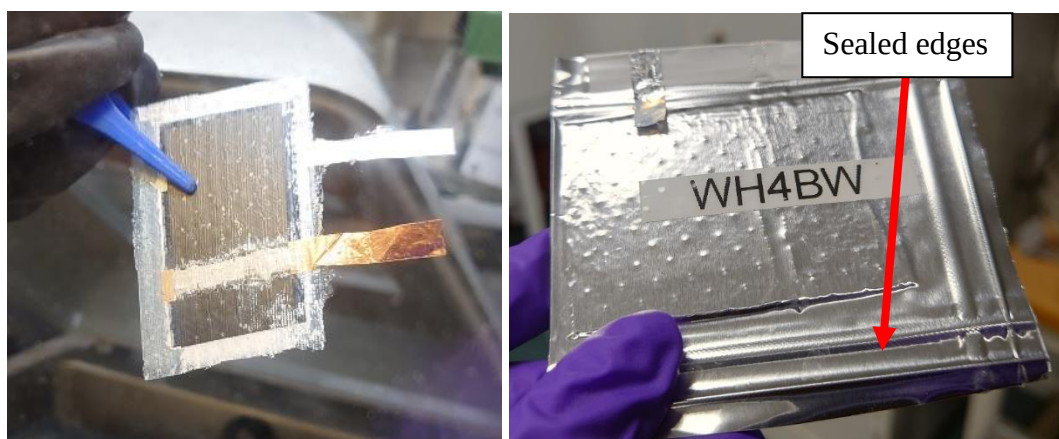


Figure 20. Cured full cell (left). Cell sealed in pouch bag (right).

Pouch bag preparation is described in Figure 21 below. Electrical insulating tapes are used to cover the cut parts (edges) where the Al layer from pouch bag might be exposed. A hot melt tape ensures proper sealing around the current collectors. After placing the cured cell, pouch bag is folded and sealed from all sides (Figure 20).

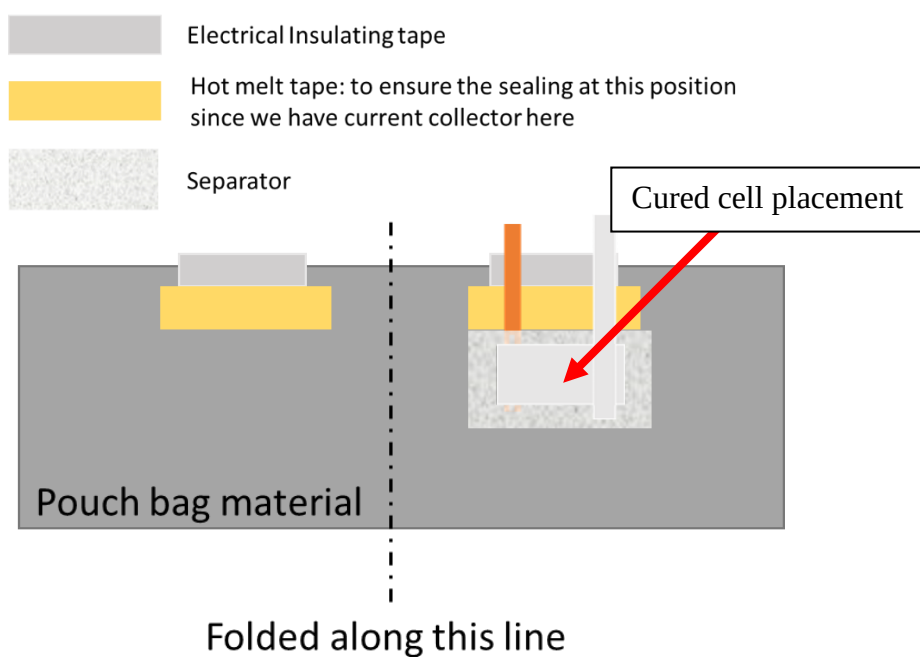


Figure 21. Pouch bag preparation.

3.4. Electrochemical characterization

The structural battery cells are first conditioned by charging and discharging at constant 0.300 mA current. This galvanostatic charge and discharge (GCD) was repeated at 0.567 mA for 5 cycles and the resulting specific capacity and energy density were calculated for each cell. The cycles of the GCD were kept within 2.00 V and 3.55 V and each cycle had a resting time for 2 hours. All of these tests were done using Neware CT-4008-5V10mA-164 battery cycler.

The choice of the current for the charging cycles was based on the theoretical C rate which is calculated from the area capacity rated for the commercially available positive electrode (1 mAh/cm²). Detailed calculations are provided in appendix A.5

For testing the coulombic efficiency of the cells, one cell was exposed to a long term cycling exceeding 170 cycles of GCD at a rate of 1 C.

Electrochemical impedance spectroscopy (EIS) was also used to test the power capability of the battery pack. Since the battery pack is assumed to be nonlinear system, it would change the battery impedance when the frequency is altered. To compare if the impedance of the cells measured earlier in Xu et al. [7] was different to this thesis, same technique of measuring the impedance at different frequencies for each cell was used.

3.5. Multicell Demonstrator

A multicell demonstrator using 6 cells was manufactured at Volvo Cars' Concept Center. Individual cells were arranged and enclosed in a number of electrically insulating layers of composite materials to form a single laminate (structural battery). A sketch of the laminate can be found in appendix A.1 showing the arrangement of the cells. The laminate had individual connection points for the cells so that they could be connected outside in any desired configuration. Schematic diagram of the layers applied for the laminate can be found in Figure 22.



Figure 22. Structural battery demonstrator laminate layers.

The layups were vacuum infused at -1 bar with the epoxy-hardener mix (10:3 respective mass ratio) and cured initially at room temperature overnight then heated up to 50°C in steps for 8 h. Current collectors from the cells were taped with release plastic caps to avoid epoxy contact as seen Figure 23.

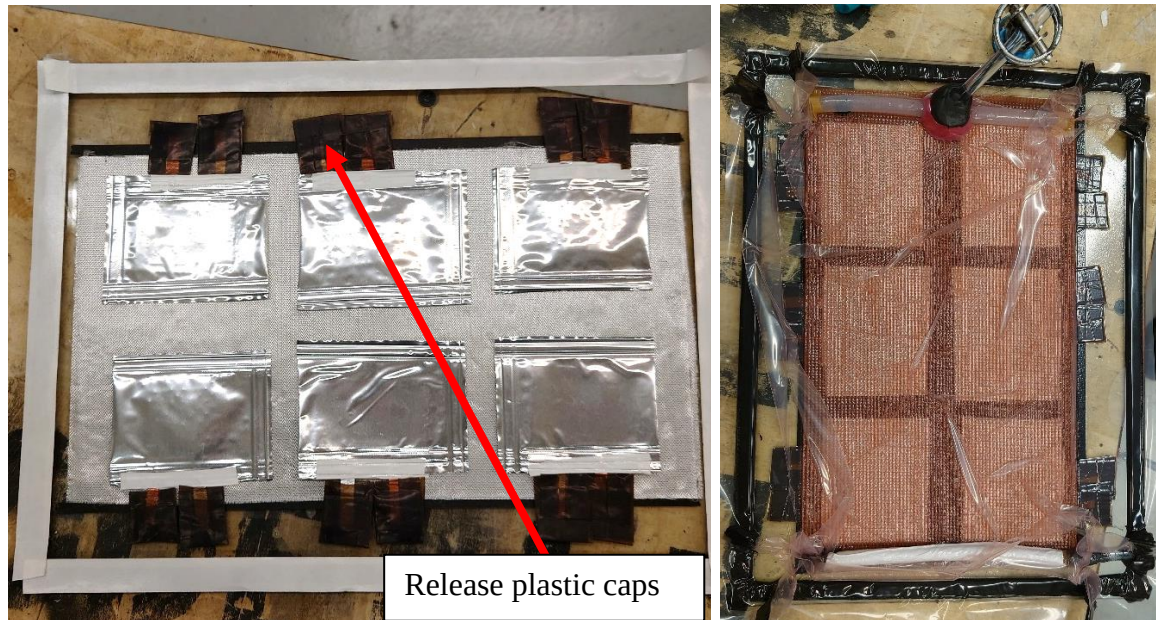


Figure 23. Arrangement of cells inside the layups (left). Vacuum infusion of the laminate (right).

The resulting multicell demonstrator is presented in Figure 24 below. The glass fiber only side of the laminate is see-through showing the cells within the panel.

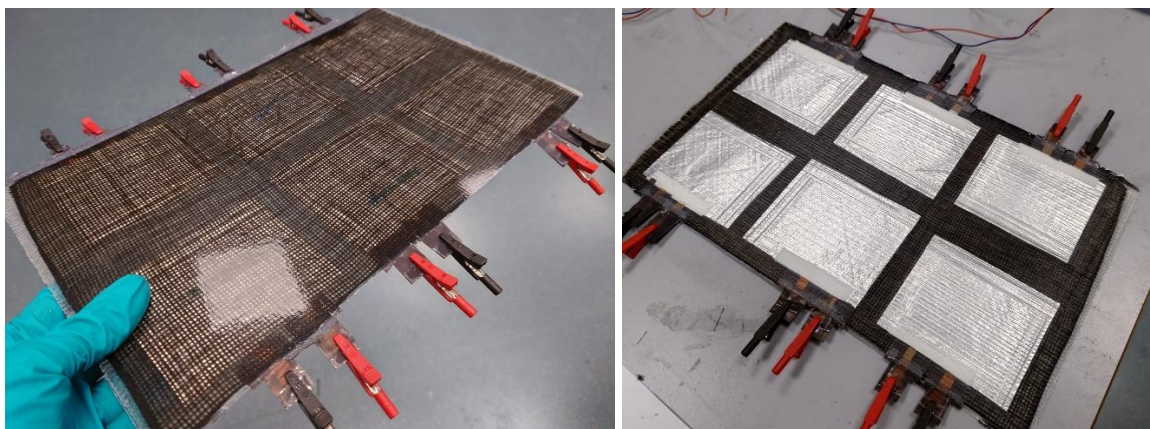


Figure 24. Structural Battery multicell demonstrator showing the opposite sides of the laminate.

3.6. Manufacturing time

Table 3 shows the manufacturing time taken for one cell on a laboratory scale and a comparison to the estimated manufacturing time if the cells were to be produced on a large commercial scale. Time stated in the table takes current Li-ion cell manufacturing techniques into account and assumes similar techniques to be applied for manufacturing the composite structural battery. From start to finish, a composite structural cell is completed within 10 days in a lab. With current industrial cell manufacturing technologies, the manufacturing time for composite structural batteries can be greatly reduced in the sizing and assembly steps [10], it also will eliminate the variation in performance among the cells by reducing manual processing such as sizing and assembly. For composite battery manufacturing, extra steps such as vacuum checking, infusion, and curing, the time should be similar as they do not depend on automation but rather are time dependent processes. Infusion of structural battery can be done using conventional infusion equipment with the exception of SBE solution making which could be prepared in an inert environment and fed using ducts directly. Also, the cells need to be more robust for commercial use and therefore need to be conditioned properly for a longer period. The number of conditioning cycles can be interpreted based on the results shown in Figure 32.

As research on these structural batteries are continuously improving and changing, industrialization or automation of manufacturing processes can be costly [10] at current research stage as with changes to structural battery technology will also involve altering manufacturing methods.

Table 3. Lab scale manufacturing time compared with commercial level manufacturing of composite cells

Tasks	Duration for lab-scale manufacturing (approx.)	Duration for commercial level manufacturing
Cutting and dimensioning of cell components	1 h 30 min	80 to 150 m/min [10]
Cell assembly	1 h	Depends on cell design [10]
Vacuum checking	1 h	1 h
Vacuum drying	15 h	12 to 30 h [10]
Infusion and curing	4 h	< 4 h
Cell conditioning	8 days	1.5 to 3 weeks [10]

4. Results & Discussions

4.1. Separators for full cells

Cell performances based on different separator materials were investigated in the beginning of the thesis: Whatman GF/A, GF plain weave, and Freudenberg (FR). Cells based on the Freudenberg separator showed unpredictable performance with a low success rate. It was concluded that the knowledge on its application in composite batteries was limited and therefore was discarded in order to work towards the objectives for this thesis. GF plain weave and Whatman GF/A separators were used for benchmarking the new infusion manufacturing process. Whatman separator was chosen for the cells in the demonstrator.

As shown in the SEM image below, voids in the carbon fibers were observed on a failed cell with FR separator. It was believed that the low vacuum pressure (-1 bar) was causing the voids mainly due to the boiling of the SBE. Due to this extreme low pressure, the spreading happened too quickly leaving less time for the SBE to penetrate the FR separator. This could be because of the low porosity of the separator. Whatman separator, on the other hand, showed better SBE spread with little or no void in the carbon fibers as observed in Figure 25.

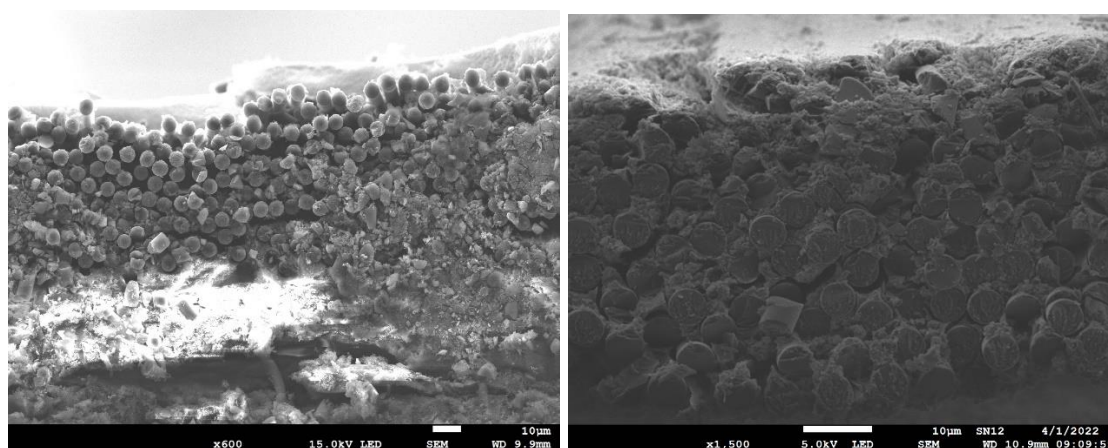


Figure 25. SEM samples at -1 bar pressure. A failed cell with voids resulting in loose fibers (left). Working cell with Whatman GF/A separator (right).

After manufacturing more than a dozen cells using FR separator, the maximum energy density and area capacity obtained at 0.63 mA (theoretical 0.05 C rate) is presented in Figure 26, where they are compared with a cell using Whatman GF/A separator manufactured and tested in the same conditions. The area capacity of a FR cell is much less compared to Whatman GF/A cell. However, the energy density is higher for FR cell since the thickness is much less compared to the Whatman G/F A as seen in Figure 27, resulting in a much lighter cell (mass of 0.031 g/cm² for FR cells compared to 0.064 g/cm² for Whatman GF/A cells). This is an indication that we can increase the performance of the SB cells using lighter and more porous separator.

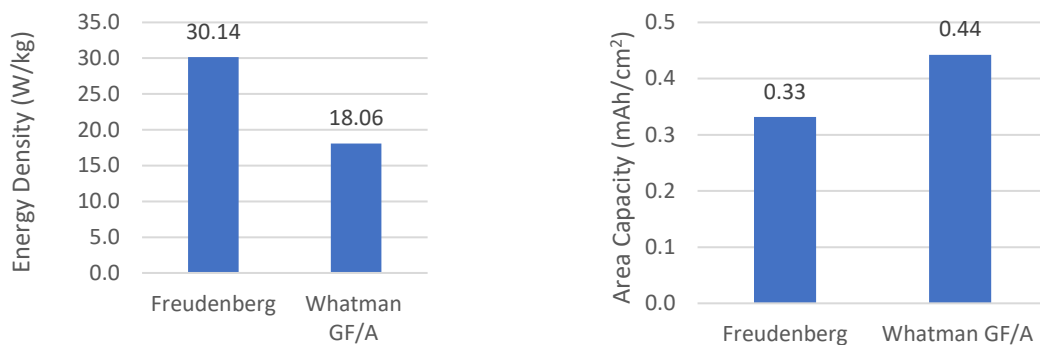


Figure 26. Energy densities [W/kg] (left) and area capacities [mAh/cm²] (right) of Freudenberg and Whatman GF/A cells at 0.63 mA discharge current (theoretical 0.05 C). Cells infused at -1 bar pressure.

The vacuum pressure was then **changed to -0.5 bar** which gave much better electrochemical results explained in section 4.3. Cells with FR separator were also tested at -0.5 bar, but they all failed. Further tests on cells with FR separator were stopped at this point and GF plain weave were tested instead along with Whatman separator.

The cells using FR had a higher failure rate compared to the cells with Whatman G/F separator. This is most likely because the separator is extremely fragile, and some fibers maybe came in contact with the positive electrode causing a short circuit.

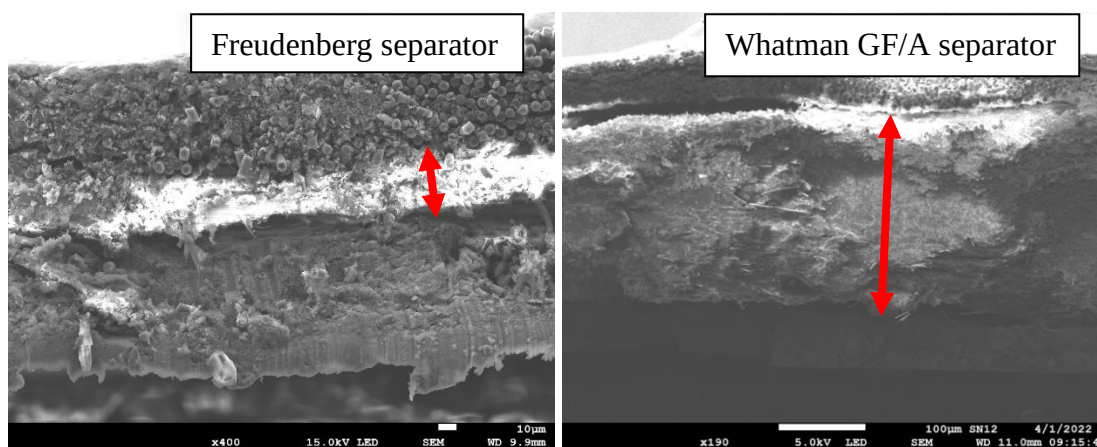


Figure 27. Thickness comparison of cells with Freudenberg (left) and Whatman GF/A separators (right).

4.2. Effect of vacuum pressure on SBE composition

FTIR spectroscopy was done on two samples of SBE to check if the vacuum pressure has any boiling off effect on any components. If the chemical composition of the SBE was altered, the FTIR spectroscopy of the sample after applying vacuum pressure would have shown different peaks or different absorbance. As observed in Figure 28, the two samples had the exact same major peaks proving that the vacuum pressure does not affect the composition of the SBE. The composition of the samples from the spectrum was not studied in this thesis due to the time limitation.

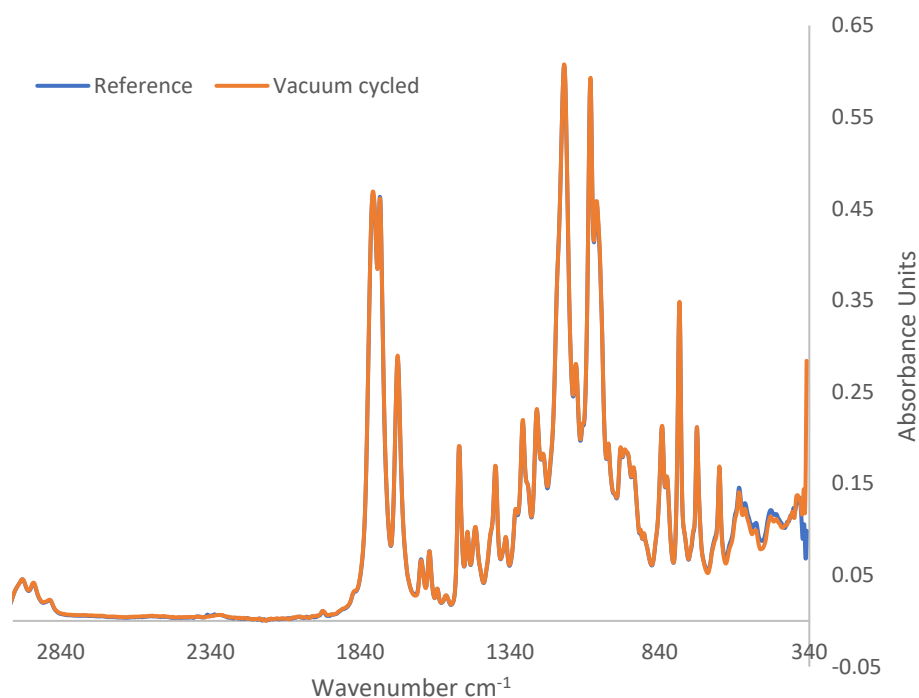


Figure 28. FTIR spectrum of SBE samples

However, we do see that the reference spectrum does slightly differ when the wavenumber is within $340 - 690 \text{ cm}^{-1}$ which is also known as the far-IR region. The spectroscopy of the reference sample was rerun to understand the spectrum better. Figure 29 shows the wavenumbers where the spectrums do not overlap. From $440 - 620 \text{ cm}^{-1}$, the absorbance units were observed to reduce with every sample. This could be due to the side reactions of the SBE as it was exposed to air and moisture during the procedure. Since far-IR spectrum usually is considered to be weak [11], the noisy peaks at $340 - 390 \text{ cm}^{-1}$ were not investigated.

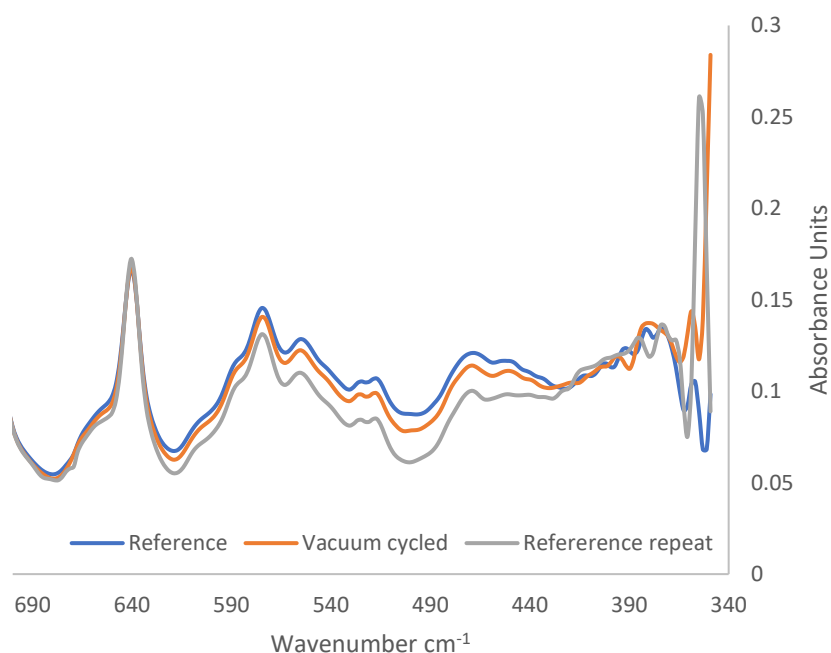


Figure 29. FTIR spectra with repeat result

4.3. Electrochemical performance

The electrochemical properties of the cells infused at -0.5 bar with Whatman GF/A and GF plain weave separators are given in Table 4. It can be observed that the energy densities of Whatman GF/A and GF plain weave are 30 Wh kg⁻¹ and 41.2 Wh kg⁻¹ respectively for a discharge time of 20 hours (0.05 C). The change of infusion pressure from -1 bar to -0.5 bar enhanced the performance of the Whatman GF/A cells significantly. The mass value of cells used to calculate these figures includes all cell components (electrodes, separator, and current collectors). Comparing this to state-of-the-art cells manufactured by Asp et al., 2021 [2], the energy densities of the cells were increased significantly when the vacuum infusion technique was used to manufacture the cells. This increase can be explained by the pressure compacting the layers and the spread of SBE evenly throughout the electrodes for a better composite structure.

Table 4. Electrochemical data for structural batteries using Whatman GF/A and GF plain weave separators at a discharge C rate of 0.05 C (20 h discharge time). Cells infused at -0.5 bar pressure.

	Whatman GF/A	GF plain weave
Energy density (Wh/kg)	30.0	41.2
Area capacity (mAh/cm ²)	0.66	0.70
High SOC voltage (V)	3.22	3.23
Low SOC voltage (V)	2.62	2.64
Mass (g/cm ²)	0.064	0.042

The difference between the energy densities of the cells manufactured using the vacuum infusion technique and the cells manufactured earlier in Asp et al., 2021 [2] are better shown in Figure 30. Due to the shortage of time, the GF plain weave could not be repeated at all test points. The Whatman GF/A cells from this thesis showed higher capacities than the GF plain weave (Asp et al., 2021 [2]) at low C rates. As it can be observed, after a certain C rate increase, the performance decreases for all cells which could be the lack of ion extraction at higher current. The cells manufactured using the vacuum infusion technique showed almost identical capacities (Whatman GF/A 1 and 2. Also the Demo cells in section 4.4) so we can conclude that this technique produces cells with repeatable performance.

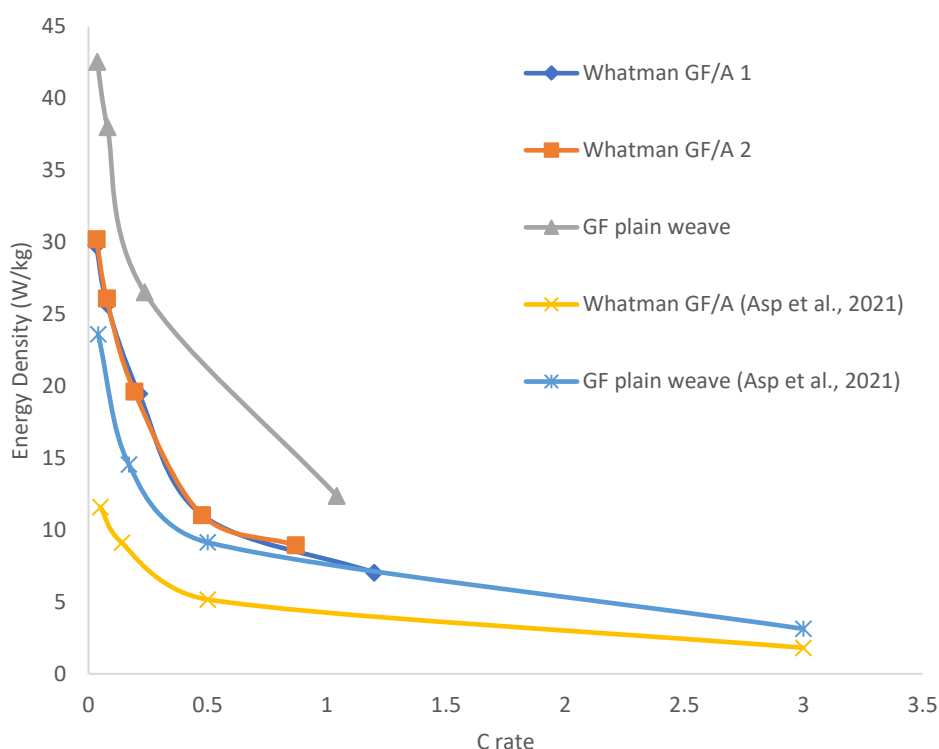


Figure 30. Comparison of energy densities at different C rates

Energy density of the Whatman GF/A cells at different theoretical C rates is given in Figure 31. This graph does not report the actual C rate since the choice of different currents was based on the calculations as mentioned in appendix A.5. Even after going through 20+ cycles, the cell is observed to have negligible fade of capacity. The drop in capacity at theoretical 0.05 C (0.63 mA current) is observed to be around 2 Wh kg⁻¹. When higher current was used to achieve a theoretical 0.4 C (5 mA current) rate, the cell does not perform at all (fully charged and discharged within 10 seconds) which could be due to the limitations created by the low ion conductivity of the SBE and the low electron conductivity of the carbon fibers [9].

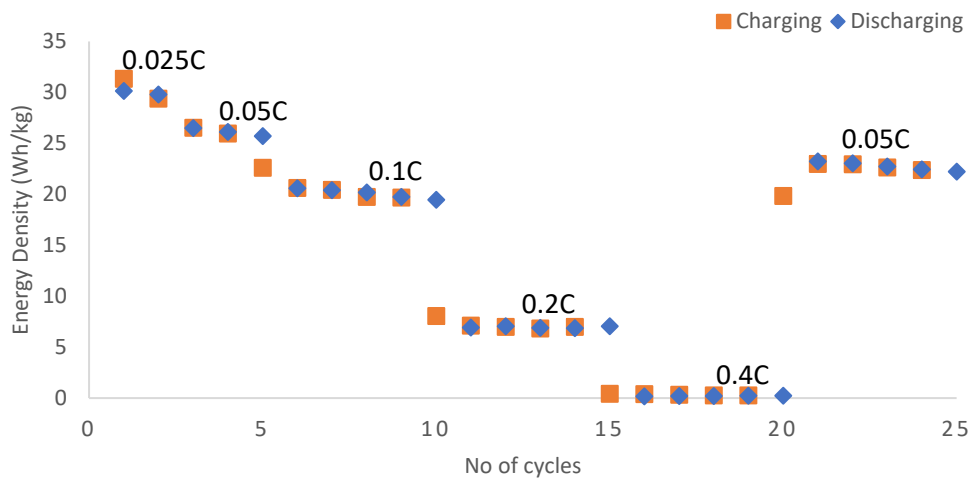


Figure 31. Whatman cell energy density at different theoretical C rates

Figure 32 shows the capacity retention during galvanostatic cycling at 1.6 mA. The cell was first pre-conditioned and tested so it went through 35 cycles before the capacity retention test. The energy density dropped from 10.3 Wh kg⁻¹ to 5.2 Wh kg⁻¹ within 160 cycles. It is observed to become stabilized after 140 cycles and retained capacity on repeated cycles afterwards. From both Figure 31 and Figure 32, it is evident that the difference in charge- discharge energy density is negligible for each cycle which shows that these structural batteries have high coulombic efficiency.

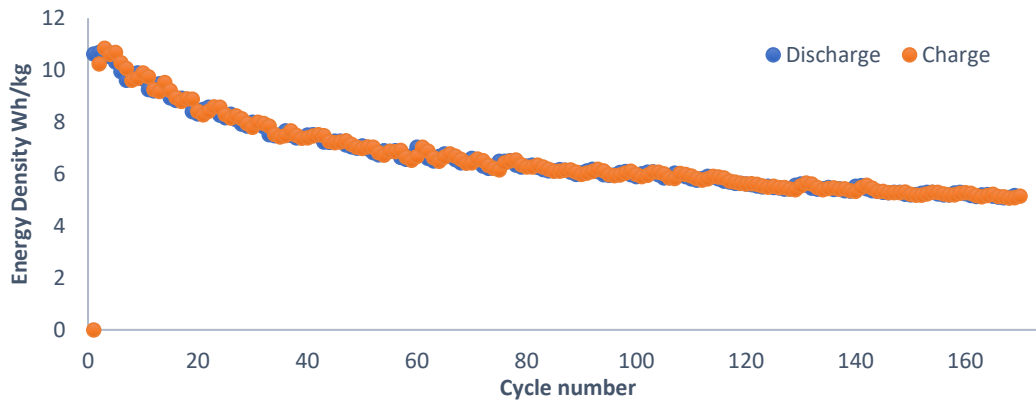


Figure 32. Capacity retention test results for the Whatman GF/A cell at 1 C.

4.4. Results of demo cells

Cells having an energy density of more than 24 Wh kg⁻¹ (more than double capacity compared to previous benchmark composite cells with Whatman GF/A by Asp et al., 2021 [2]) were considered as high performing. These high performing cells had a deviation of ± 1.4 Wh kg⁻¹ with an average energy density of 26.2 Wh kg⁻¹ when discharged at 0.567 mA (theoretical C rate of 0.05 C). This variation can be due to manual handling (sizing and assembly) of the cells which might be greatly reduced by process automation. The energy densities of all cells manufactured with Whatman GF/A separators are given in Figure 33. Demo cells with missing data in the figure indicates that the cell did not reach infusion stage due to handling errors or equipment failure such as cracking of mold or vacuum bag leakage.

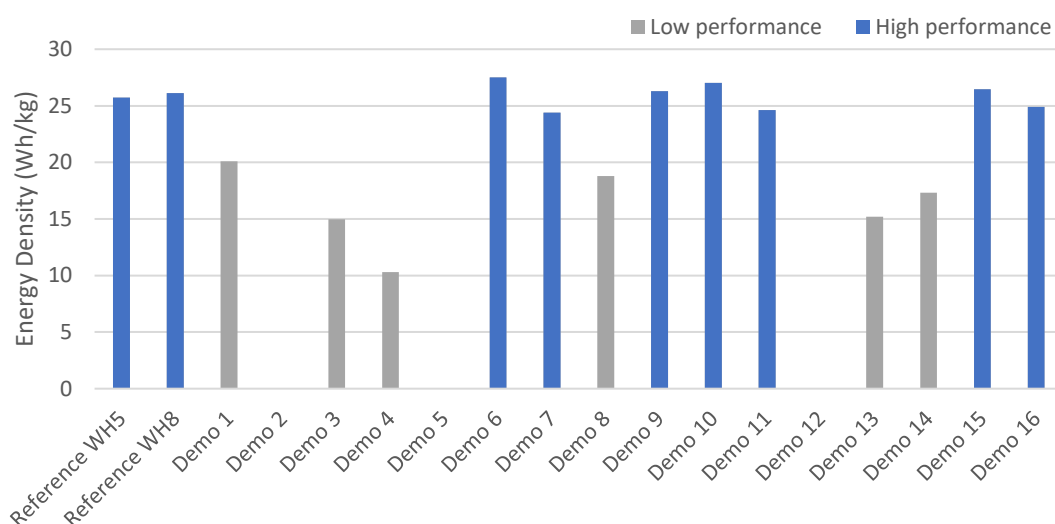


Figure 33. Energy densities of all demo cells used for the multicell laminate demonstrator and reference cells at a discharge rate of 0.05C (theoretical C rate).

The Nyquist plots in Figure 34 shows trend of the surface impedance of the composite cells at high state-of-charge (high SOC). The cells demonstrated a similar behavior as reported in conventional battery research. Considering a surface area of 11.3 cm², the measured surface impedance of the composite structural battery pack ranges from 283.5 Ωcm² to 1530.9 Ωcm². When compared with previous studies on state-of-the-art composite structural batteries done by Xu et al. [9], the differences in resistance between the manufactured cells was greatly reduced (3 to 5 times reduction) by implementing this new vacuum infusion manufacturing technique for the cells. Interestingly, the charge transfer resistance was also decreased (226.8 Ωcm² compared to 360.0 Ωcm² [9]) even though the same SBE chemical composition was used. This could be due to the even wetting of cell components by vacuum infusion of SBE solution compared to manual wetting method of Asp et al. [2] and Xu et al. [9]. The disturbance observed at the lower frequencies were not

investigated in this thesis. Surface impedance test at low SOC was not completed due to time limitation of this research.

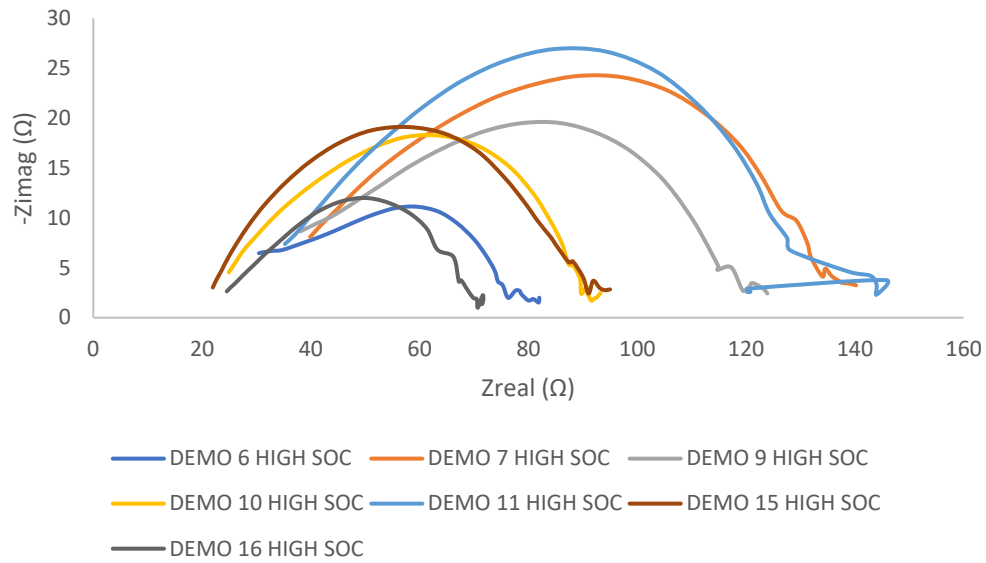


Figure 34. Nyquist plot of all the demo cells used for the multicell laminate demonstrator.

4.5. Battery pack performance

The design of the laminate battery pack allowed the connection of cells to be done separately outside the laminate. This provided with the flexibility to connect and test the six demo cells in any configuration. As a result, the cells could be charged separately and discharged in a configured circuit as seen in Figure 35, eliminating the need of a battery management system (BMS) for this thesis work. Within the limited time available, the battery pack was tested for three configurations: 1s6p and 2s3p the results of which are summarized in Table 5.

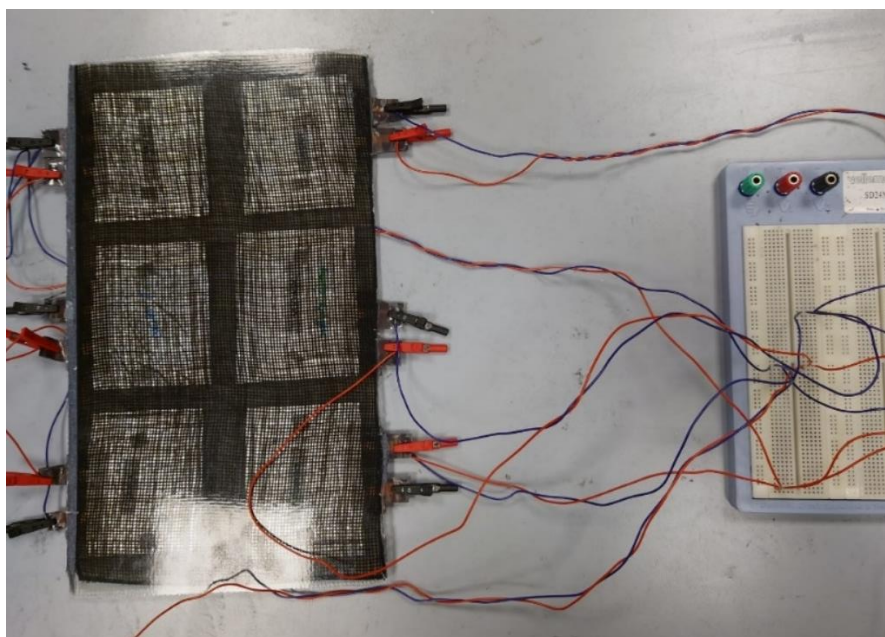


Figure 35. Battery pack testing setup.

For the electrochemical testing of the battery pack in 1s6p, a C rate of 0.5C at 9.6 mA ($1.6 \text{ mA} \times 6$) current was considered with an expected discharge time of 2 hr. Capacity, energy, and power were then calculated using the actual recorded current, voltage, and discharge times. Similarly, testing for 2s3p configuration was also carried out. The testing results are referred to as ‘Experimental’ in Table 5.

Table 5. Structural battery pack performance in different configuration.

Current (mA)	Voltage (V)	C rate	Capacity (Ah)	Energy (Wh)	Power (W)	Runtime	Comments
1s6p							
9.6	2.9	0.5C	0.0202	0.0585	0.0278	2 h	Theoretical
9.6	2.9	0.3C	0.0288	0.0835	0.0278	3 h	Experimental
57.6	2.9	6C	0.0096	0.0278	0.1670	10 min	Estimated
2s3p							
4.8	5.8	0.5C	0.0101	0.0585	0.0278	2 h	Theoretical
4.8	5.8	0.3C	0.0144	0.0835	0.0278	3 h	Experimental
28.3	5.8	6C	0.0048	0.0278	0.1670	10 min	Estimated

Recorded data for a single cell was used for theoretical calculation of the battery pack performance referred to as ‘Theoretical’ rows of the table. ‘Experimental’ rows are direct measurements from the configuration circuit of the battery pack. ‘Estimated’ row calculations are based on a single cell performance with an estimation that the battery pack will behave in a similar way to a single cell, for instance, at 6 C discharge rate, the battery pack is assumed to run for 10 minutes. This estimation for high currents is done because the equipment used for the battery pack testing are rated for low current applications and

also no cells were tested at this high C rate to avoid risks of damaging the cells since only a limited number was produced. A comparison in Table 5 between the experimental and theoretical, i.e., calculates, rows of 1s6p configuration shows that the 1s6p configuration was supposed to run for 2 h at 9.6 mA but experimentally recorded discharge time for the battery configuration was 3 h. This is because the cells in the battery pack were relatively new and therefore underwent a smaller number of cycles prior to this test and as a result, had higher capacity. Same procedure was followed for the other configuration testing, estimation, and theoretical calculation.

5. Conclusion

It has been demonstrated that the proposed vacuum infusion method for composite cell manufacturing is superior to any previous method used to manufacture structural battery composites. The infusion at a pressure of -0.5 bar resulted in 100% success rate of manufacturing cells without failure. Furthermore, 70% of the cells was high-performing, with an energy density in excess of 24 Wh kg⁻¹ and low variability (with a deviation of ± 1.4 Wh kg⁻¹). Cells with Whatman GF/A and glass fiber plain weave separators exhibited energy densities of 30.0 Wh kg⁻¹ and 41.2 Wh kg⁻¹ respectively at 0.05 C (approximately 20 h of discharge time) which is a breakthrough when compared to all previous research mentioned in the literature. Moreover, the surface impedance and charge resistance of the cells have been reduced significantly (3 to 5 times) by adopting this manufacturing technique compared to previous studies.

Based on experience from this research, the main limitation was the size of transfer chamber of glovebox. Cell size, manufacturing quantity, time, and complexity all depends on this. Moreover, LFP Al foils posed post curing issues such as delamination of LFP particles from the foil and therefore an alternative cathode such as LFP coated CF might be promising. In future research, cured cell sealing techniques other than pouch bags should be explored. It is also recommended to research on CF weave with embedded Cu fibers in between which might have potential mechanical benefits and remove the issues with loose, flying fibers making the manufacturing of cells more robust, simpler, and quicker. The principal implication of this methodology has the possibility to be applied on a large-scale production system although more studies should be done beforehand.

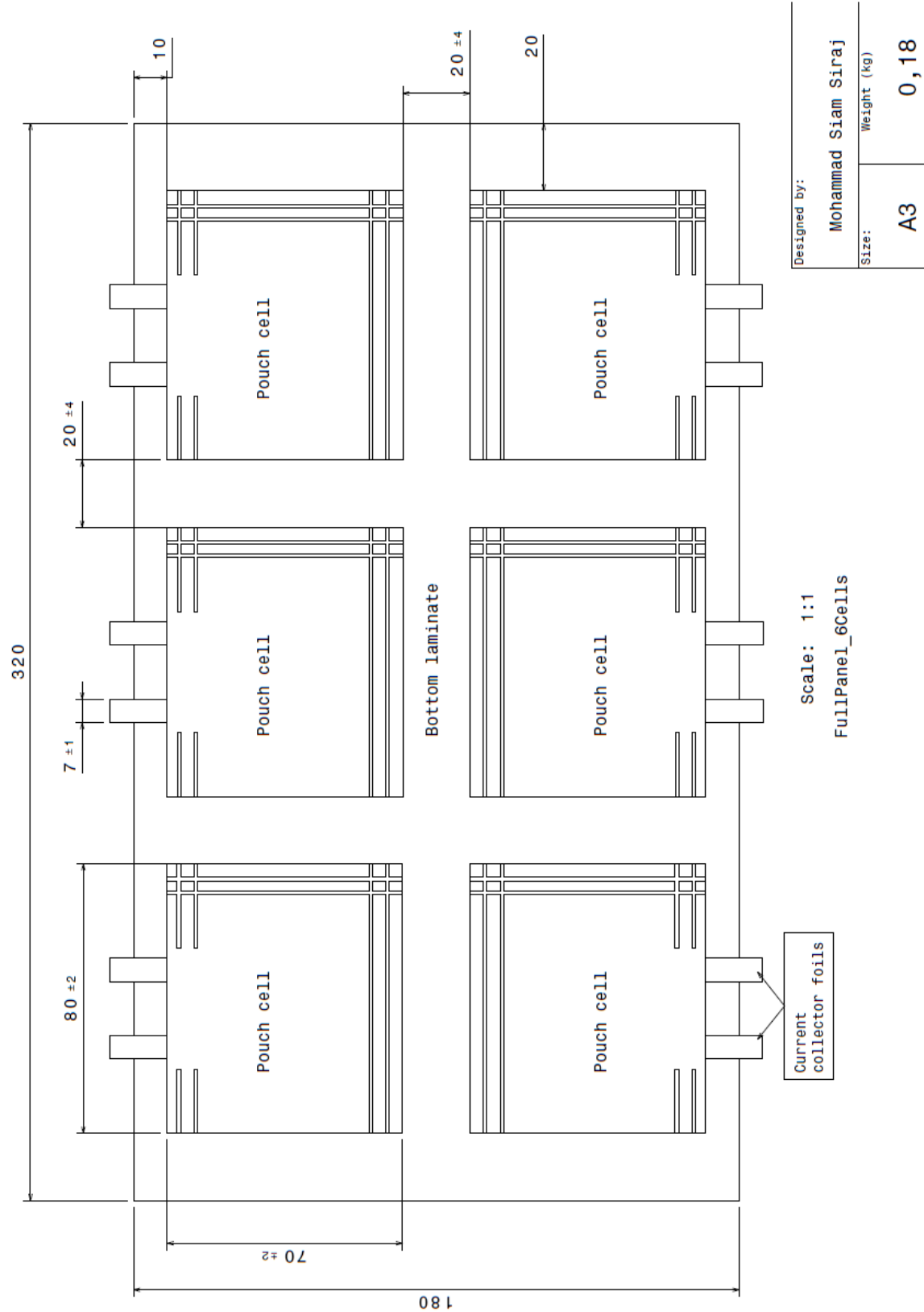
6. References

- [1] L. E. Asp, S. Leijonmarck, T. Carlson and G. Lindbergh, "Realisation of structural battery composite materials," in *20th International Conference on Composite Materials*, Copenhagen, 19-24th July, 2015.
- [2] L. E. Asp, K. Bouton, D. Carlstedt, S. Duan, R. Harnden, W. Johannisson, M. Johansen, M. K. G. Johansson, G. Lindbergh, F. Liu, K. Peuvot, L. M. Schneider, J. Xu and D. Zenkert, "A Structural Battery and its Multifunctional Performance," *Advanced Energy and Sustainability Research*, vol. 2, no. 3, pp. 1-9, 2000093, 2021.
- [3] D. Deng, "Li-ion batteries: basics, progress, and challenges," *Energy Science & Engineering*, vol. 3, no. 5, pp. 385-418, 2015.
- [4] W. Johannisson, "Exploring structural carbon fiber composites for mass-less energy and actuation," Doctoral Thesis, KTH Royal Institute of Technology, Stockholm, 2020.
- [5] L. E. Asp, M. Johansson, G. Lindbergh, J. Xu and D. Zenkert, "Structural battery composites: a review," *Functional Composites and Structures*, vol. 1, no. 4, 2019.
- [6] L. M. Schneider, N. Ihrner, D. Zenkert and M. Johansson, "Bicontinuous Electrolytes via Thermally Initiated Polymerization for Structural Lithium Ion Batteries," *Applied Energy Materials*, no. 2, pp. 4362-4369, 2019.
- [7] J. Xu, W. Johannisson, M. Johansen, F. Liu, D. Zenkert, G. Lindbergh and L. E. Asp, "Characterization of the adhesive properties between structural battery electrolytes and carbon fibers," *Composites Science and Technology*, vol. 188, 2020.
- [8] R. Harnden, "An investigation into ion-inserted carbon fibres for structural energy storage, shape-morphing, energy harvesting & strain-sensing," Doctoral Thesis, KTH Royal Institute of Technology, Stockholm, Sweden, 2021.
- [9] J. Xu, Z. Geng, M. Johansen, D. Carlstedt, S. Duan, T. Thiringer, F. Liu and L. E. Asp, "A multicell structural battery composite laminate," *EcoMat*, vol. 4, no. 3, 2022.
- [10] Y. Liu, R. Zhang, J. Wang and Y. Wang, "Current and future lithium-ion battery manufacturing," *iScience*, vol. 102332, no. 24, 2021.
- [11] N. Jaggi and D. R. Vij, "Fourier Transform Infrared Spectroscopy," in *Handbook of Applied Solid State Spectroscopy*, Boston, Springer, 2006, p. 411–450.
- [12] S. Duan, *Full cell instruction*, Gothenburg: Chalmers University of Technology, 2021.
- [13] E. Jacques, M. H. Kjell, D. Zenkert, G. Lindbergh, M. Behm and M. Willgert, "Impact of electrochemical cycling on the tensile properties of carbon fibres for structural lithium-ion composite batteries," *Composites Science and Technology*, vol. 72, pp. 792-798, 2012.

- [14] E. Jacques, M. H. Kjell, D. Zenkert, G. Lindbergh and M. Behm, "Expansion of carbon fibres induced by lithium intercalation for structural electrode applications," *SciVerse ScienceDirect*, vol. 59, pp. 246-254, 2013.
- [15] L. E. Asp and E. S. Greenhalgh, "Structural power composites," *Composites Science and Technology*, vol. 101, pp. 41-61, 2014.
- [16] G. Fredi, S. Jeschke, A. Boulaoued, J. Wallenstein, M. Rashidi, F. Liu, R. Harnden, D. Zenkert, . J. Hagberg, G. Lindbergh, J. Patrik, L. Stievano and L. E. Asp, "Graphitic microstructure and performance of carbon fibre Li-ion structural battery electrodes," *Multifunctional Materials*, vol. 015003, no. 1, 2018.
- [17] D. Carlstedt and L. E. Asp, "Performance analysis framework for structural battery composites in electric vehicles," *Composite Part B*, vol. 107822, no. 186, 2020.
- [18] L. E. Asp and E. S. Greenhalgh, "Multifunctional structural battery and supercapacitor composites," in *Multifunctionality of Polymer Composites*, Elsevier Inc., 2015, pp. 619-661.
- [19] D. Carlstedt, F. Rittweger, K. Runesson, A. M. Navarro-Suárez, J. Xu, S. Duan, F. Larsson, K.-R. Riemschneider and L. E. Asp, "Experimental and computational characterization of carbon fibre based structural battery electrode laminae," *Composites Science and Technology*, vol. 109283, no. 220, 2022.
- [20] S. N. Nguyen, A. Millereux, A. Pouyat, E. S. Greenhalgh, M. S. Shaffer, A. R. Kucernak and P. Linde, "Conceptual Multifunctional Design, Feasibility and Requirements for Structural Power in Aircraft Cabins," *Journal of Aircraft*, vol. 58, no. 3, May-June 2021.

Appendices

A.1 Laminate sketch



A.2 Manufacturing equipment

The following equipment were used in the manufacturing of the cells:

- Separate scissors; one for general purpose, and two separate scissors for cutting LFP and CF electrode materials.
- Separate scalpels; one for general purpose, one for debagging the cells, and two separate scissors for cutting LFP and CF electrode materials.
- Separate tweezers. one for general purpose, two for debagging the cells, and two separate scissors for cutting LFP and CF electrode materials.
- Electrical insulating tape
- Scotch tape
- Hot melt tape
- Pouch bag
- Vacuum bagging material
- Infusion mesh
- Breather fabric
- 4mm outer diameter silicone tubes
- Tacky tape
- Plain release plastic
- Perforated release plastic
- Peel ply
- Vacuum pump
- Catch can
- Clamps

A.3 Electrolyte recipe

Electrolyte recipe:

- EC: 6.5 g
- PC: 6.5 g
- LiTFS: 0.94 g
- LiBoB: 0.78g

A.4 SBE recipe

SBE recipe for infusing one cell (if no breather or infusion mesh is used while vacuum bagging):

- Monomer: 3 g
- Electrolyte: 3 g
- Initiator: 0.03 g (1 wt % of monomer)

A.5 Theoretical C rate calculation

Rated capacity of LFP foil: 1 mAh/ cm²

Area of active material: 11.34 cm²

Based on rated capacity, theoretical C rate of 1 C should require 1 mA of current.

For example,

A theoretical 0.05 C rate would require the following current:

$$\text{Current} = \text{rated capacity} \times \text{area of active material} \times \text{theoretical C rate}$$

$$\text{Current} = 1 \text{ mAh/cm}^2 \times 11.34 \text{ cm}^2 \times 0.05 \text{ C} = 0.567 \text{ mA}$$



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