Structural Battery Composites: A Review, Functional Composites and Structures

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Functional Composites
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TOPICAL REVIEW

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TOPICAL REVIEW

Structural battery composites: a review

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Abstract

This paper presents a comprehensive review of the state-of-the-art in structural battery composites research. Structural battery composites are a class of structural power composites aimed to provide mass-less energy storage for electrically powered structural systems. Structural battery composites are made from carbon fibres in a structural electrolyte matrix material. Neat carbon fibres are used as a structural negative electrode, exploiting their high mechanical properties, excellent lithium insertion capacity and high electrical conductivity. Lithium iron phosphate coated carbon fibres are used as the structural positive electrode. Here, the lithium iron phosphate is the electrochemically active substance and the fibres carry mechanical loads and conduct electrons. The surrounding structural electrolyte is lithium ion conductive and transfers mechanical loads between fibres. With these constituents, structural battery half-cells and full-cells are realised with a variety in device architecture. The paper also presents an overview of material modelling and characterisation performed to date. Particular reference is given to work performed in national and European research projects under the leadership of the authors, who are able to provide a unique insight into this emerging and exciting field of research.

1. Introduction

This paper addresses a new type of multifunctional lightweight composite material desired for its potential to reduce vehicle weight and ease future electrification across transport modes. We refer to these materials as *structural battery composites*. The paper reviews the current status of structural battery composites. Focus is on the activities performed during the last decade by an interdisciplinary team of researchers in Sweden set to realise structural battery composites from carbon fibre reinforced polymers (CFRP).

The need to develop greener, safer and more competitive road and air transport has been recognised as of critical societal and commercial importance. In Europe, the ERTRAC and EPoSS strategy paper[6] and Flightpath2050[7] have been used by the European Commission to define the Green car initiative and Green vehicle as well as the Clean Sky Joint Undertaking in European Union FP7 and H2020 research funding frameworks. For road transport electrification of urban mobility and transport has been highlighted as a most urgent research area. In addition, the Green vehicle initiative identify the need for advanced lightweight materials to realise future lightweight electric vehicle solutions. To this, Airbus has expressed a vision for an all-electric regional aircraft for 100 passengers by year 2050! The research on structural battery composites is conducted in this setting with ambition to pave the road for 'mass-less' energy storage in future vehicle structures. This will be achieved by realisation of multifunctional lightweight composite materials that simultaneously can carry mechanical loads and store electrical energy. Such materials will allow radical weight savings for future electric and hybrid vehicles,

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[6] ERTRAC and EPoSS European Roadmap Electrification of Road Transport, Version *2.0/November 2010*.

[7] http://ec.europa.eu/transport/air/doc/flightpath2050.pdf

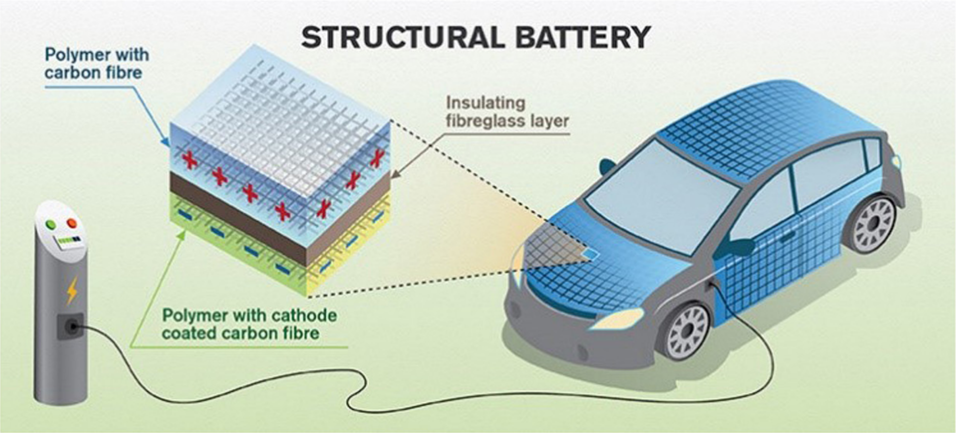

Figure 1. Carbon fibre can store energy in the body of a car[8].

contributing to highly energy-efficient and emission-free vehicles ranging from bicycles, city-movers, cars and boats to aircraft, as illustrated in figure 1.

Benefits of multifunctional engineering materials are numerous and diverse. The use of structural battery composites, for instance, goes well-beyond transportation applications. We foresee that they will be used in transportable electronics as mobile phones and lap-tops. However, concerning emissions of greenhouse gases transport applications are most interesting. While electrification of the automotive sector is already underway electrification of aircraft is less developed. This is partly due to the relatively moderate energy densities of available battery technologies. For example, Airbus' vision to make all-electric regional aircraft by 2050 is very challenging. Consider a 1 h flight with a 100-passenger aircraft. If Airbus were to simply replace the approximately 30 kg of kerosene per passenger used for such a flight with batteries, they would need batteries at a weight of approximately 1000 kg/passenger. Consequently, means to make secondary structures or interior parts from structural battery composites are very appealing to aircraft manufacturers.

Structural battery composites belong to a new class of multifunctional composites called structural power composites (Asp and Greenhalgh 2014). These comprise of structural composites with an inherent ability to store electrical energy, such as structural capacitors (Carlson *et al* 2010, O'Brien *et al* 2011), supercapacitors (Shirshova *et al* 2013) and batteries. The current paper addresses only structural battery composites.

In the literature there are two different approaches to achieve combined mechanical and electrical performance in structures. One approach is to add functionalities in to a component. This is done by, for instance, embedding thin-film batteries within composite laminates. These are essentially sandwich structures (Thomas and Qidwai 2004, Thomas *et al* 2005, Ladpli *et al* 2019). Such devices do not rely on multifunctional material constituents but is rather an assembly of components (i.e. the composite face sheets, foam core and thin-film battery). We refer to these as multifunctional structures. The second approach is to make a multifunctional material in which every constituent performs multiple functions (Asp and Greenhalgh 2014, Lendlein and Trask 2018). In such structural battery composites carbon fibres function as reinforcement and electrode and the polymer acts as matrix and electrolyte. This paper focuses solely on multifunctional materials, i.e. the structural battery composite materials.

The first attempts to make a structural battery composite material from multifunctional constituents were made at the US Army Research Labs (ARL) (Wetzel 2004, Snyder *et al* 2007). Wetzel and his team at ARL did pioneering work utilising different reinforcing materials as structural electrodes and a glass weave separator in a common polymer electrolyte matrix as they built the first structural battery composite material (Wong *et al* 2007). The resulting structural battery material demonstrated promising mechanical performance but did not work electrically due to poor electrical insulation. Following the attempts at ARL, Liu *et al* (2009) designed a structural battery composite with short-fibre reinforced electrodes and a solid polymer electrolyte matrix material. However, they were not able to manufacture the composite electrodes and failed to identify a sufficiently ion-conductive polymer electrolyte. For these reasons they used a robust gel electrolyte as matrix. This resulted in a working battery with very low mechanical properties. Inspired by these works, Ekstedt *et al* (2010) made a functioning laminated structural battery employing a gel electrolyte reinforced with a carbon fibre weave negative electrode, a glass weave separator and a lithium iron phosphate ($LiFePO_4$)/aluminium fibre weave positive electrode. The structural battery, containing lithiated carbon and $LiFePO_4$ showed an Open Cell Potential of 3.3 V.

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[8] www.chalmers.se/en/departments/ims/news/Pages/carbon-fibre-can-store-energy.aspx

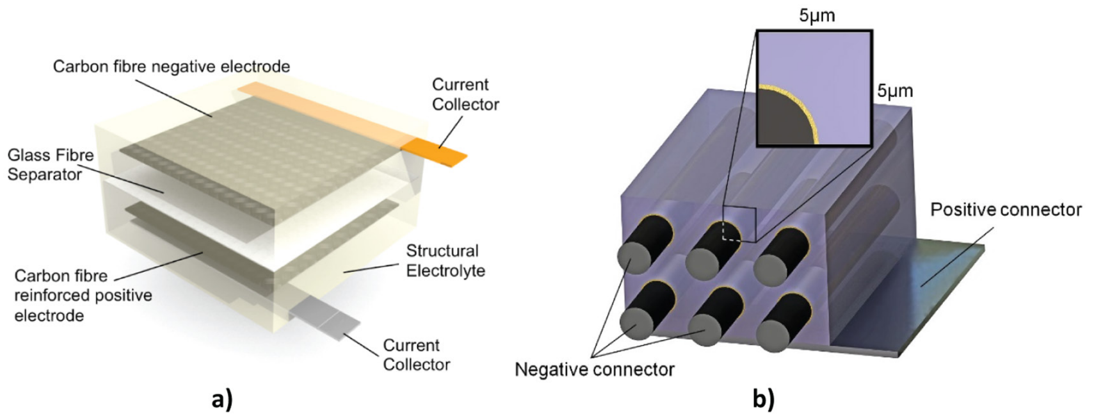

Figure 2. (a) Laminated and (b) 3D-fibre structural battery designs (the 3D-fibre battery illustration is from Carlson (2013)).

Early structural battery designs relied on a laminated structure where two electrode layers were separated by an electrically insulating (separator) layer. This design is the obvious choice as it is a common architectural feature for composite laminates and conventional batteries. However, the laminated design faces some substantial challenges that must be overcome in order to realise a working structural battery. As indicated above, one of these challenges is the ion-conductivity and mechanical properties of the combined electrolyte and matrix. Lack of highly ion-conductive and stiff polymer electrolyte systems will hamper realisation of structural battery composites. The second is the thickness of the separator layer. Thick separator layers will significantly reduce the power and energy density of the structural battery composite and internal resistances will cause high energy losses. Conventional batteries have a separator thickness of around 20–30 μm. This is much thinner than conventional composite plies, which typically are 100–200 μm in thickness. To overcome these challenges new structural battery electrolytes have been developed. This is reported in detail in chapter 3 of this paper. One approach for structural electrolyte development was first suggested by Snyder *et al* (2007) using block co-polymer electrolyte systems as composite matrix. These contain a cross-linking monomer as structural enhancer and an ethylene oxide (EO) rich monomer as diffusion enhancer of the polymer. An alternative approach used for structural supercapacitors is also considered for structural battery composites. Shirshova *et al* (2013) proposed a bi-continuous polymer/ionic liquid system. In such systems the hollow polymer structure transfers mechanical loads and the liquid diffuses ions in its hollow channels. With these approaches, structural electrolytes with stiffness of up to 1 GPa and ion-conductivity exceeding 10^{-4} S cm^{-1} have been realised. Given the multifunctional properties of the available structural electrolytes two different structural battery designs have evolved: the laminated and the 3D fibre structural battery architectures. These are schematically illustrated in figure 2, and further discussed in chapter 5.

In this paper the state of the art of structural battery composites is presented. The paper addresses carbon fibre reinforced structural battery composites and discusses the multifunctional constituents, battery cell and half-cell designs and performance as well as their modelling and characterisation.

2. Carbon fibres as negative electrodes in structural battery composites

To realise structural battery composites, the carbon fibres must perform two functions simultaneously: carry mechanical loads and act as an active battery electrode. In the latter the carbon fibres must be able to store ions in the micro-structure of the fibre itself. This is when ions (in our case lithium ions) are inserted into the fibre during charge and removed on discharge, in our case also denoted lithiation and delithiation, respectively. In a full battery cell, lithium ions are inserted or lithiated into the negative carbon electrode during charge and removed or delithiated during discharge.

There are a few key issues to be studied in relation to the fibre itself. Can carbon fibre be lithiated and store lithium as a battery electrode? What type of fibres are most favourable for storing lithium? How large are the electrochemical losses during charge and discharge? Does mechanical load affect the process of lithiation and delithiation? What happens to the fibre as lithium occupies space in the fibre micro-structure and does that affect the mechanical properties of the fibre?

2.1. Mechanical properties of carbon fibres

Carbon fibres have perhaps the best specific mechanical properties of all commercially used materials in high performance structures. There are two main classes of carbon fibres stemming from the pre-cursor from which they are made: pitch-based and polyacrylonitrile (PAN)-based fibres. Pitch-based fibres usually exhibit very high

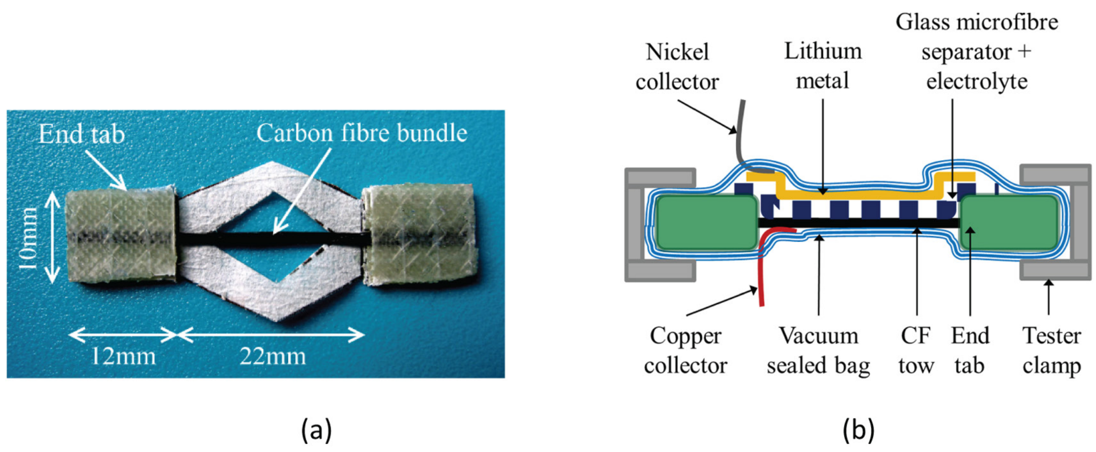

Figure 3. (a) Carbon fibre bundle with end-tabs and (b) schematic of electrochemical cell reprinted from (Jacques *et al* (2013b)), Copyright 2013, with permission from Elsevier.

stiffness (ranging up to around 800 GPa) but with moderate strength and with low failure strains in the order of less than 1%. They usually have a high graphitic content with large and oriented crystals. PAN-based fibres are the most commonly used fibres today and come in a wide range of qualities. By choice of precursor and process parameters, the microstructure can be varied considerably, especially with respect to crystallite size and orientation. Therefore, carbon fibres offer a wide range of mechanical and physical properties (Johnson 1987, Lavin 2001, Park 2015, Fredi *et al* 2018). Their modulus ranges from about 200–600 GPa and with strengths from around 3000–6000 MPa. The ones in the lower range of stiffness (and with high strength) are denoted HS (high strength) or HT (high tenacity) fibres. They have lower graphitic content and small and not very oriented crystals. The intermediate modulus (IM) fibres have a modulus of around 300–400 GPa, have both higher graphitic content and somewhat larger and more oriented crystals. There are also PAN-based ultra-high modulus fibres that are graphitised at very high temperatures, giving high graphitic content, high orientation and with a modulus over 500 GPa. Early work in Japan (Kanno *et al* 1989) and at ARL (Snyder *et al* 2009a) demonstrated PAN-fibres to be very good electrodes, whereas pitch-based carbon fibres were not. Extensive characterisation of contemporary carbon fibre performed by Kjell *et al* (2011, 2013) confirm high capacities of PAN-based carbon fibres, e.g. IMS65 and T800 fibres as discussed below.

2.2. Electrochemical properties of carbon fibres

A wide range of carbon fibres were tested electrochemically to study which types of fibres perform best as battery electrode (Kjell *et al* 2011, Fredi *et al* 2018). They ranged from Toray T300, which is a typical HT fibre, to Toho Tenax IMS65 and Toray M60J which are intermediate and high modulus fibres with moduli of 290 and 580 GPa, respectively. The carbon fibres were used either as received or with the sizing chemically removed before sample preparation.

In order to simplify the specimen preparation carbon fibre bundles were tabbed using two layers of standard glass fibre/epoxy pre-preg as seen in figure 3(a). The specimens were then placed in a cell with a microfibre separator and a lithium metal counter electrode. Current collectors were applied, liquid electrolyte was poured into the cell which was then vacuum sealed in a bag, see figure 3(b). This was all done inside a glovebox in an oxygen free and dry environment.

The cells were electrochemically cycled galvanostatically (using a constant current) between 0.002–1.5 V versus Li/Li^+. Different current rates were used ranging from 1 C (a constant current that gives a charge and discharge time of 1 h) down to C/10 (1/10:th of the current at 1 C giving charge and discharge times of approximately 10 h).

All the tested carbon fibres exhibited some electrochemical capacity. However, the high modulus fibres had quite low capacity which was explained by staging as the prevailing lithiation mechanism. Lithiation in graphite occurs via staging. However, for the high modulus fibre staging was partially obstructed by the turbostratic graphitic structure of the carbon fibre (Fredi *et al* 2018). As consequence, its larger graphite crystal size and higher crystal orientation could not be efficiently exploited for lithium insertion. Hence, the studied high modulus fibres demonstrated low electrochemical capacity. In contrast, two IM fibres showed excellent electrochemical capacity: Toho Tenax IMS65 and Toray T800. They are very similar mechanically (modulus 290 GPa and strength around 5500 MPa). The capacities were 130 (T800) and 177 (IMS65) mAh g^{-1} at 1 C, respectively (Kjell *et al* 2011). The intermediate modulus fibres were found to insert lithium with a mechanism like that found for disordered carbons. As consequence, the intermediate modulus fibres (IMS65 and T800) outperform high modulus fibres (M60J) as shown by Fredi *et al* (2018).

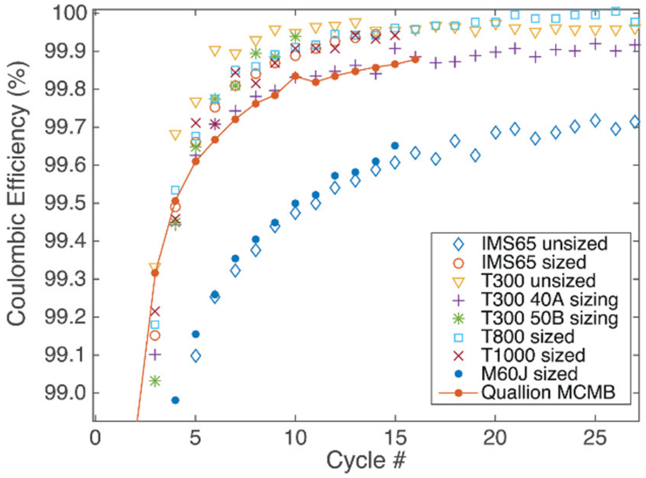

Figure 4. Coulombic efficiency as function of cycle number for various carbon fibre. Solid line is for a commercial MCMB (Quallion). Reproduced from (Hagberg *et al* (2016)). CC BY 4.0.

Some effect of the sizing was found but not for all fibre types. At low charge rates (C/10) the capacities were surprisingly high: 290 mAh g^{-1} for T800 and 357 mAh g^{-1} for IMS65, which is very close to the theoretical capacity of pure graphite, which is 372 mAh g^{-1}.

Kjell *et al* (2013) found that the high drop in capacity (about 50%) after the first charge cycle observed in the tests on fibre tows (capacity at first and tenth lithiation was 360 mAh g^{-1} and 177 mAh g^{-1}, respectively) was partly attributed to the bundle structure. In tests on single fibres the reversible capacity was reduced only by 25% compared to that at the first cycle. IMS65 single unsized fibres showed a reversible capacity of 250 mAh g^{-1}, whereas the capacity for the bundle was 177 mAh g^{-1}. Furthermore, a general lower influence of charge rate for single carbon fibres compared to that for carbon fibre tows was demonstrated. Consequently, Kjell *et al* (2013) found that a battery employing single fibre electrodes can be charged much faster than batteries made from carbon fibre tows. The tests on single fibres also showed that sized IMS65 fibres had the same electrochemical performance as unsized fibres.

In a later study by Hagberg *et al* (2016) the coulombic efficiency was measured using high precision coulometry. This relates to the electrochemical losses during charge and discharge cycles, which must be very low for a battery. This was done on several fibre types and compared with state-of-the-art graphitic battery electrodes from micro carbon meso beads (MCMB). The results are presented in figure 4.

As can be seen in figure 4 the coulombic efficiency is very high for some of the carbon fibres tested in this study, some even surpass that of state-of-the-art graphitic battery electrodes.

2.3. Effect of lithiation on mechanical properties

In a study by Jacques *et al* (2012) the effect on the mechanical properties of carbon fibres due to lithium insertion was studied. The same type of specimen as shown in figure 3 was used for tests on T800 and IMS65 fibre tows. They were cycled in a pouch cell (see figure 3(b)) at 1 C for up to 1000 cycles. The mechanical properties were measured after some intervals of cycling. Both the longitudinal stiffness and strength were measured. The result showed (in essence) that lithium insertion does not seem to affect the stiffness of the fibres, at least not during 1 C cycling up to 1000 cycles, as seen in figure 5(a). The tensile strength was reduced after lithiation (when the carbon fibre is charged) but some of it was regained as the fibres were delithiated (when the carbon fibre is discharged), see figure 5(b). This also indicates that some lithium is trapped inside the carbon fibres even after discharge.

In a later study (Jacques *et al* 2014) the same type of experiment was repeated but now the stiffness and strength was measured as function of capacity corresponding to the amount of lithium inserted into the fibre. It was found that the stiffness remained unchanged with capacity (lithium content). The strength decreased with capacity and with a drop of approximately 30% for the IMS65 fibre and approximately 20% for T800 at full capacity (above 300 mAh g^{-1}). At delithiated state the strength drop was approximately 15% for IMS65 and 5% for T800 at the slowest charge rate. Note that constant stiffness is not the same as constant modulus. In fact, as the fibre expands radially during lithium insertion the axial fibre modulus E_z is reduced (Jacques *et al* 2014). Work is currently in progress to determine the effect of lithiation on the radial and shear moduli of the fibres E_r and G_{zr} of IMS65, T800 and M60J carbon fibres (Duan *et al* 2019).

The opposite effect has also been investigated: the effect of mechanical loads on the electrochemical performance (Jacques *et al* 2011). In this case, no effect was found, and the carbon fibres had the same capacity whether there was a tensile strain applied to them or not during charge or discharge.

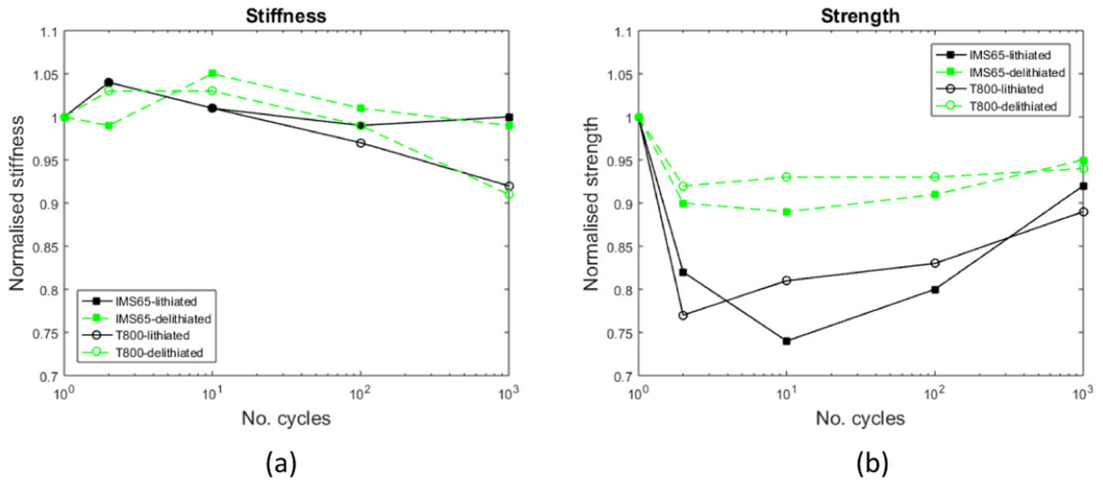

Figure 5. Stiffness (a) and strength (b) of carbon fibre after electrochemical cycling reprinted from (Jacques *et al* (2012)), Copyright 2012, with permission from Elsevier.

2.4. Fibre expansion during charge and discharge

In the electrochemical cell, lithium ions move from one electrode to the other during charge, and the other way during discharge. These ions are moving in and out of the carbon fibres, which leads to volume changes. It is well known that graphitic electrodes change its volume during lithiation. A study was performed by Jacques *et al* (2013b) to measure the axial expansion of carbon fibres when used as battery electrodes. A cell as in figure 3 was placed in a tensile testing machine and a constant strain was applied. Since the carbon fibre bundle was in a pouch bag, the bag material was first allowed to relax. The carbon fibre was then lithiated at various galvanostatic currents (constant current charge/discharge), leading to different charge rates. Since the fibres expand when lithiated the load applied decreased (the strain was constant). In a second step the fibre bundle was delithiated and the applied load increased again corresponding to a contraction. Since the fibres have the same stiffness in lithiated and delithiated states the load decrease/increase can be directly related to the expansion and contraction of the fibre. As anticipated, the expansion depends on the charge rate, or rather the amount of lithium inserted. The force changes during these experiments are shown in figure 6. As seen, the axial expansions and contractions (changes in load) were large for low charge rates (see e.g. curve for 0.3 C in figure 6) and small for high charge rates (6 C). The axial expansion strains were unexpectedly large, reaching almost 1% in the axial direction at low charge rates. It was also found that some of the expansion was irreversible, i.e. the fibres did not contract back to their original length after discharge. This further supports that some lithium is trapped in the micro-structure after discharge. The axial expansion was not linear with capacity (lithium content) but still quite close to linear. No reliable method to measure radial expansion of carbon fibres from lithium insertion has yet been developed and validated. Preliminary measurements using scanning electron microscopy (SEM) suggests much higher radial expansions, up to 5% (Jacques *et al* 2013b).

2.5. Piezo-electrochemical effect

Carbon fibres are good electrical conductors but do not exhibit a piezo-electrical response. However, when lithium ions are inserted into their micro-structure, they do become piezo-electric, or as a more appropriate term this is named the piezo-electrochemical effect. The experiments used to measure this effect was in principal the same as described above (Jacques *et al* 2013a). A tabbed carbon fibre bundle in a pouch cell (see figure 3) was placed in a tensile tester. It was electrochemically cycled and at various states-of-charge the charging/discharging was interrupted leaving the cell in an open circuit potential (no currents running so the cell is at its resting potential). A load was applied and the potential between the carbon fibre electrode and the counter electrode (in this case a lithium metal foil) was measured. Interestingly, a change in the electrical potential was found, for both IMS65 and T800 fibres. The response was a few mV for a strain of approximately 0.3%. The response was not constant with capacity but seemed to have a maximum around 50% state-of-charge. The piezo-electrochemical response was about the same at full charge (high capacity) and at full discharge (Jacques *et al* 2015). Again, this support the fact that lithium is being trapped in the structure even when the fibre is fully discharged.

2.6. Resistivity of carbon fibres

To accurately model the electrical processes in the structural battery the resistivity of the constituents must be known. Schutzeichel *et al* (2019) measured the resistance of a polymer electrolyte coated carbon fibres (PeCCF) unidirectional composite. A temperature independent ohmic longitudinal resistance of the PeCCF, lying at the lower end of semi-conductors, was found. This is beneficial, since Joule heating and resulting temperature

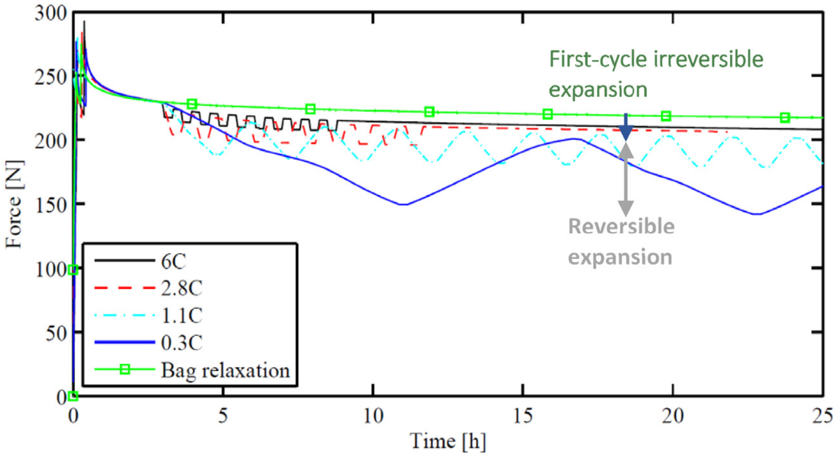

Figure 6. Load change measurements of IMS65 at various charge rates reprinted from (Jacques *et al* (2013b)), Copyright 2013, with permission from Elsevier.

predictions can be based on a temperature independent resistor. Temperature prediction is necessary to control/predict the mechanical behaviour of the compound. Furthermore, Schutzeichel and co-workers presented a method to compute the number of fibres in the composite. Once the total averaged resistance and the measurement length are determined the number of fibres in the composite can be calculated using the measured specific resistance. The specific resistance of the IMS65 fibre was determined to $1.35 \cdot 10^{-3}\ \Omega$ cm.

3. Structural battery electrolytes

One of the main challenges for a structural battery electrolyte (SBE) is to make a composite matrix that simultaneously allow for lithium ion conduction and a mechanical load transfer between the carbon fibres. These two properties in many cases counteract each other, i.e. either a high ion conductivity and low mechanical performance or vice versa is achieved. A second challenge is that the SBE also needs to interact with the fibres via a suitable interface, i.e. the constituents must be possible to combine. These challenges have been addressed in different ways as described in the following sections.

3.1. Homogenous systems

The first SBE approach described by Snyder *et al* (2007) utilized the well-established fact that poly(ethylene glycol) (PEG) can dissolve lithium ions as already described in the 1970s (Fenton *et al* 1973, Wright 1975). Using acrylate functional PEGs this formed thermoset structures with a structural integrity as well as allowing for a lithium ion transport (Nest *et al* 1988, Snyder *et al* 2007, 2009b, Wysocki *et al* 2010, Willgert *et al* 2011, 2012, 2013). Parameters such as lithium ion content, crosslink density, and presence of plasticizers have been shown to improve the multifunctional performance to some extent, but these SBE's still suffer from the trade-off between mechanical properties and ion conductivity (Ihrner and Johansson 2017). One approach to increase the ion conductivity for solid polymer electrolytes is to make so-called gel polymer electrolytes (GPEs) that are obtained by swelling a sparsely crosslinked thermoset polymers with a liquid electrolyte. This concept has been applied to a wide range of different polymers to give ion conductivities up to 10^{-2} S cm^{-1}, see e.g. review paper by Stephan (2006). The relationship between the two counteracting properties however to a large extent remains why new approaches had to be taken.

3.2. In-homogenous systems

The first approach that was pursued in this context was to start with a GPE that was reinforced to form a composite polymer electrolyte (CPE). Numerous reinforcements ranging from inorganic particles (see e.g. Croce (1998)) to nano-cellulose and chitosan (see e.g. Willgert *et al* (2014)) have been tested and shown that significant multifunctional improvement can be obtained. For example, a modulus of 400 MPa and a conductivity of $5\ 10^{-5}$ S cm^{-1} have been reported by Willgert *et al* 2014. The main problem for these systems arose when the SBEs should be combined with carbon fibres as electrodes, i.e. there was no feasible route to assemble these to a half-cell. It should however be noted that this concept of CPE can be applied in other contexts e.g. a structural membrane.

Hence, new routes to resolve the problem of improved multifunctional performance and processability had to be developed. One route towards this goal is to start with a homogeneous liquid that upon polymerization/solidification transforms to a heterogeneous material with two percolating phases, where one phase determines the ion transport and the other mechanical load transfer. The reason for the phase separation is that the solubility/miscibility of two components depend on the components' relative solubility parameters (Hansen 1967) and that the solubility parameters change when monomers transform to a polymer. By an appropriate choice of components with specific solubility parameters a system can be designed so that all components are totally miscible initially and totally immiscible after the monomers transformed to polymers. The initial liquid can then be infused into an electrode material and when polymerized form two intermingled continuous phases on a submicron scale. This technique is already well established and implemented into production in other areas where two counteracting properties are desired, e.g. in contact lenses (Seitz *et al* 2017). This concept has been demonstrated for several different systems (Shirshova *et al* 2013, 2014, Greenhalgh *et al* 2015, Yu *et al* 2016) to give a combination of relatively high modulus and conductivity. Ihrner *et al* (2017) utilized a combination very suitable for an SBE when combining a conventional liquid solvent/Li-salt electrolyte with a bis-phenol-A based methacrylate monomer that resembles a conventional vinyl ester resin often used for advanced composites. Fully phase separated SBE with two percolating intermingled phases was made utilizing photoinduced free radical polymerization of the monomer system. Using different monomer stiffness values in the range of 360–750 MPa were obtained while simultaneously having ion conductivities between 1–2 10^{-4} S cm^{-1}. The presence of two percolating phases was verified since the SBEs showed structural integrity at the same time as the liquid electrolyte could be fully extracted from the SBE. The utilized photopolymerization is however limited to systems that are transparent in the UV-range why a thermally induced polymerization route more recently was developed (Schneider *et al* 2019). This thermally induced polymerization process makes conventional production processes used for composite manufacturing possible as later described in section 4. An optimized curing scheme was 80 °C–90 °C for 30 min which yielded a monomer conversion more than 90%. Schneider *et al* (2019) further demonstrated that the thermal curing could be performed in presence of carbon fibres without any impact on the curing rate.

Looking more in detail into the thermally cured systems it reveals SBEs with porous morphology as seen in the SEM pictures in figure 7. All liquid electrolyte was removed soaking the SBE in water demonstrating that there was no detectable amount of electrolyte trapped in closed voids. Approximately 39 wt% of electrolyte was used with a retained high structural integrity of the SBE.

All thermally cured SBEs exhibit a similar morphology with structural features in the submicron range, i.e. pore sizes in the range of 50–200 nm. Mechanical analysis using DMA (dynamic mechanical analysis) revealed a broad glass transition temperature and a modulus at ambient temperature around 500 MPa. The ionic conductivities were for all samples around $2 \cdot 10^{-4}$ S cm^{-1}.

Recently He *et al* (2019) proposed an alternative structural battery electrolyte. They proposed a particle reinforced poly(ethylene oxide) (PEO) solid-state electrolyte. The composite polymer electrolyte was realised reinforcing PEO with $\mathrm{Li_{6.4}La_3Zr_{1.4}\,Nb_{0.6}O_{12}}$ (LLZNO) fillers. He and co-workers found that the incorporation of LLZNO fillers significantly improved the ionic conductivity of the composite electrolytes. The improvement was explained to be a combined result of increased amorphous region of PEO and pathways of ion transport. A wide electrochemical window of 5.2 V was demonstrated. The assembled solid $\mathrm{LiFePO_4}$/PEO-0.5LLZNO/Li batteries were shown to possess a discharge capacity of 153 mAh g^{-1} at the rate of 0.5 C and 60 °C.

4. Structural battery half-cells

In this section we describe some paths on the integration of carbon fibres and SBE to make structural battery half-cells, both negative and positive.

4.1. Laminated negative half-cells

In order make laminated carbon fibre structural negative half-cells, one needs to combine carbon fibres and the SBE described in section 3.2 into a load carrying composite lamina. This was recently done by Johannisson *et al* (2018) by using a standard vacuum infusion technique of a dry spread fibre bundle. The infusion was done on a glass plate with vacuum bag so the lamina could be cured through the glass plate after infusion using UV-light, see figure 8.

After curing the lamina was tested both electrochemically and mechanically, the latter both before and after electrochemical cycling. The results are very promising. The electrochemical capacity was around 230 mAh g^{-1} of carbon fibre after 10 galvanostatic charge/discharge cycles at C/10. Manufacturing of a single layer, and a very thin lamina, resulted in low fibre volume fraction, around 20% only. Still, the measured mechanical properties were quite good and are summarised in table 1.

As seen in table 1, the modulus in the fibre direction is moderate due to the low volume fraction. This value will increase with a higher fibre volume fraction. The transverse modulus and the estimated shear modulus on the other hand are not that much lower than for structural carbon composite laminas showing that the SBE has quite good mechanical properties. The strength values are preliminary but still quite promising. There was a

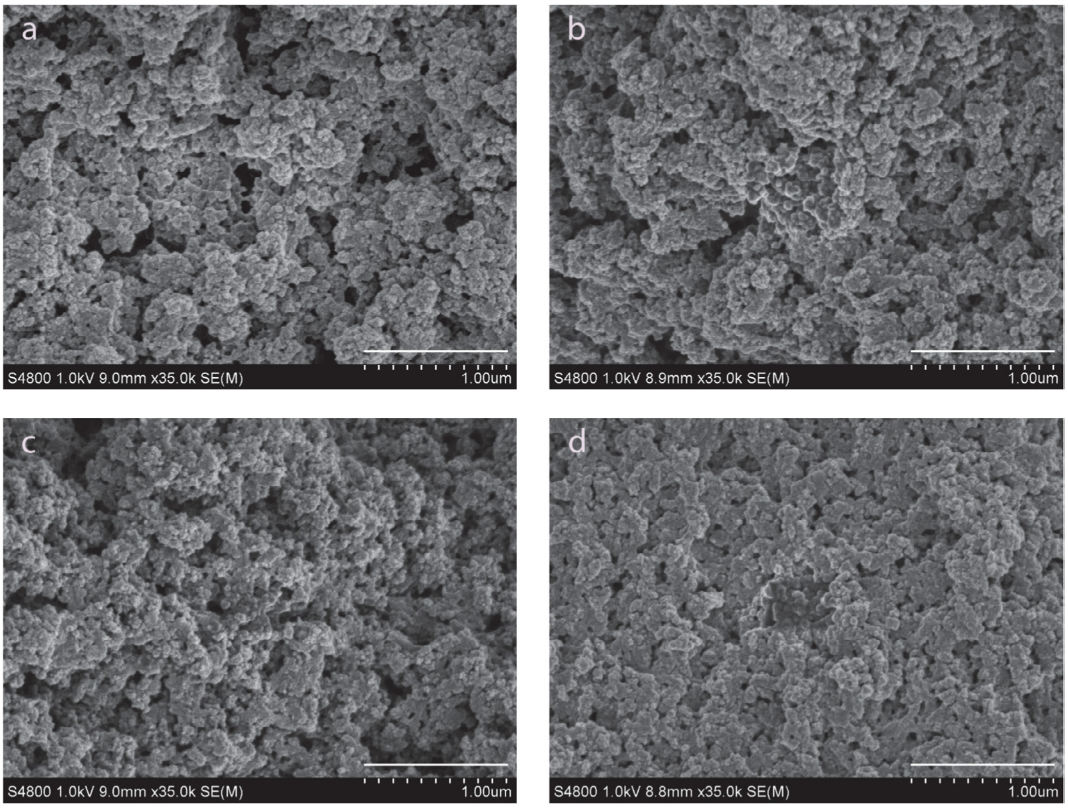

Figure 7. SEM images of SBEs cured at 70, 80, 90 °C ((a)–(c), respectively) in comparison to the corresponding system cured photoinduced polymerization (d) reprinted with permission from (Schneider *et al* (2019)). Copyright 2019 American Chemical Society.

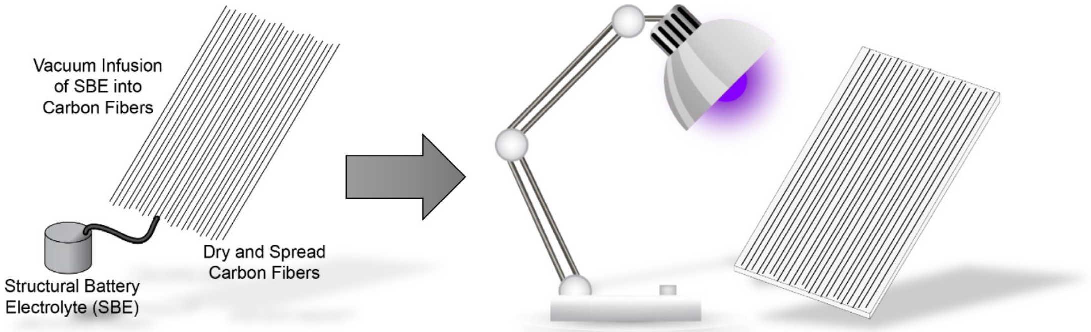

Figure 8. Schematic of infusion of a structural carbon fibre/SBE half-cell lamina.

Table 1. Mechanical properties of carbon fibre lamina half-cell reprinted from (from Johannisson *et al* (2018)), Copyright 2018, with permission from Elsevier.

Modulus and strength	Before cycling	After cycling
E_L (GPa)	52	52
E_T (GPa)	1.7	1.9
G_{LT} (GPa)	1.5	1.6
σ_L^t (MPa)	980	960
σ_T^t (MPa)	12	15
σ_L^c (MPa)	980	850
σ_T^c (MPa)	12	12
τ_{LT} (MPa)	13	14

worry that the volume changes in the fibres during charge and discharge (described in section 2.4) would potentially decrease the fibre to SBE adhesion. However, this does not seem to be the case as both the transverse and shear strengths remain unchanged, at least after 10 cycles. This was further supported by SEM imaging, which

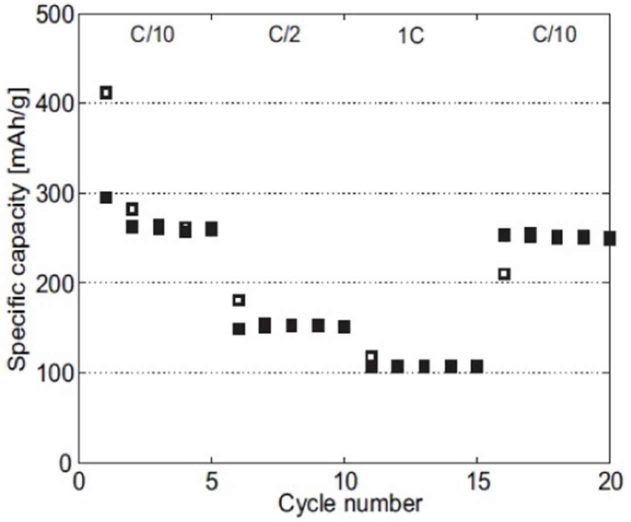

Figure 9. Lithiation (unfilled squares) and delithiation (filled squares) of electrocoated, reprinted from (Leijonmarck *et al* (2013)), Copyright 2013, with permission from Elsevier.

confirmed two things: the SBE phase separation was the same at the fibre/SBE interface as in bulk SBE, confirming ion transport through the interface, and fracture surfaces from before and after cycling looked essentially the same, both leaving SBE residue on the fibres indicating good adhesion.

Yu *et al* (2017) made laminated structural battery half-cells from thicker (0.21 mm) carbon fibre electrodes with T700 fibres employing an epoxy/ionic liquid structural electrolyte. The battery half-cells were made into coin cells and electrochemically cycled versus Li metal. A reversible capacity of approximately 15 mAh g^{-1} at best was reported by Yu *et al* (2017). Tensile stiffness data presented for the structural battery half-cells was for the fibre direction and hence dominated by the carbon fibres.

4.2. Micro-battery negative half-cells

Laminated structural battery composites, where the electrode layers are separated by an insulating material of a significant thickness, rely on highly ion conductive structural battery electrolytes. Current homogenous electrolytes (see section 3.1) require very small separator thickness (Asp and Greenhalgh 2014). For this reason, Asp and co-workers (Leijonmarck *et al* 2013, Carlson 2013) developed negative half-cells and battery full cells (alias the 3D-fibre battery) from individual polymer electrolyte coated carbon fibres, as illustrated in figure 2(b).

Leijonmarck *et al* (2013) developed a method to coat individual carbon fibres in a yarn with a homogeneous thin solid polymer electrolyte (SPE), see section 3.1. Approximately 500 nm thick methyl methacrylate coatings were applied onto the carbon fibres via an electro-polymerisation process in solution with a Li-triflate salt as supporting electrolyte. The Li-ion content in the solid polymer coating was enough to allow for the battery function. Under these conditions, being electrically insulating the SPE coating works as separator between fibres as well as between the fibres and any surrounding positive electrode material in a battery application. The electrochemical performance of a negative electrode from coated IMS65 fibres in a half-cell using lithium metal as counter electrode is depicted in figure 9. The coating does not deteriorate the cycling characteristics of the carbon fibre type used. At low currents (C/10) a specific capacity 250–260 mAh g^{-1} was measured and after the first two cycles and at high currents (1 C) up to 107 mAh g^{-1} was recorded. These results are in line with studies on unsized IMS65 carbon fibres used as negative electrodes by Kjell *et al* (2011).

4.3. Positive half-cells

To date only a limited number of studies on the design, manufacture and characterisation of the positive electrode have been published. Hagberg *et al* (2018) described a path to manufacture structural positive electrodes via electrophoretic deposition (EPD) of $LiFePO_4$ (LFP), carbon black and polyvinylidene fluoride (PVDF) onto carbon fibres, see figure 10. The carbon fibres simultaneously act as reinforcement and current collectors. Electrochemical characterisation demonstrated a specific capacity of up to 110 mAh g^{-1} with good rate performance and high coulombic efficiency. Furthermore, the cell was stable during cycling, with a capacity retention of approximately 50% after 1000 cycles. Mechanical tests on coated and uncoated carbon fibres demonstrated good adhesion between the coating and the fibre.

In a recent study, Bouton *et al* (2019) present an alternative method to coat the carbon fibres with LFP particles. The method is a repeated dip coating process to generate a layer-by-layer (LbL) assembly of LFP particles has been chosen. The process results in a few hundred nanometre thick coating layers. The dipped carbon fibres are

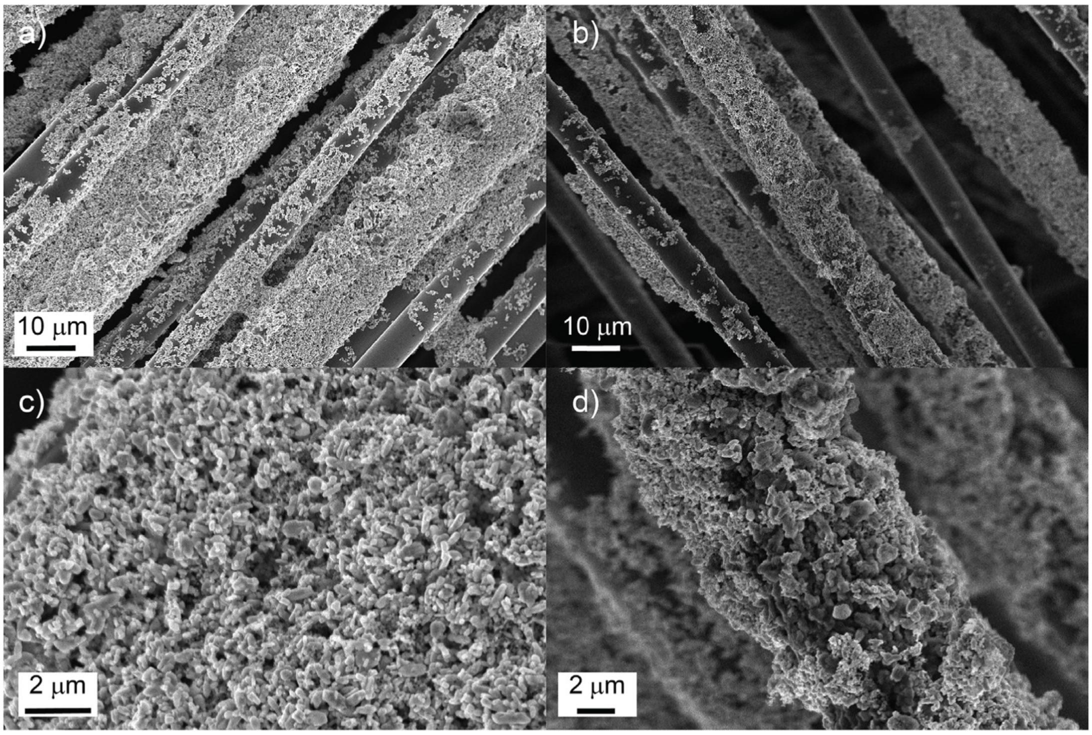

Figure 10. SEM images of LFP coated carbon fibres using electrophoretic deposition. (a) and (c) before cycling, (b) and (d) after cycling. Magnifications (a) 1000x, (b) 1000x, (c) 7000x, (d) 4000x. The active electrode material (LFP particles), conductive additive (carbon black) and binder (PVDF) were found to be well dispersed on the surface of the fibres (reprinted from (Hagberg *et al* (2018)), Copyright 2018, with permission from Elsevier).

heated up to 450 °C to carbonise the organic binders used in the solution. The LbL-electrodes showed a specific capacity close to 100 mAh g^{-1}. This is close to the capacity of LFP-coated aluminium foils, commonly used as commercial positive electrodes in conventional battery cells.

5. Architectures

The need for high energy and power density has immediate implication on the architectural designs of structural battery composites. In this section two material architectures for structural battery composites are presented: the laminated and the 3D-fibre battery designs schematically illustrated in figure 2. We start and describe the obvious laminate lay-out, separating the positive and negative electrodes with an electrically insulating, lithium ion conductive, separator layer (i.e. the traditional battery and composite laminate design). Following this, an approach making each carbon fibre into a negative electrode in a 3D-fibre battery design by coating carbon fibres with an approximately 500 nm thick SPE sheath is presented.

5.1. Laminated structural battery

As mentioned above the laminated structural battery is what first comes to mind to make a structural battery composite. Both conventional batteries and composites are laminated structures. However, there is an inherent problem to make laminated structural battery composites: the use of a solid polymer electrolyte/matrix material.

Wetzel (2004) and his team at ARL developed the first laminated structural battery composite material. ARL built a device using a metal mesh coated with a positive electrode material, a carbon fibre fabric acting as a negative electrode, a fibre glass separator layer, and a structural solid block co-polymer electrolyte matrix (Wong *et al* 2007). However, the composite did not work as a battery due to poor electrical insulation. Later, Ekstedt *et al* (2010) and Carlson (2013) were able to build working laminated structural battery composites. Carlson (2013) used an IMS65 carbon fibre woven fabric as the negative electrode, a bi-continuous multifunctional polymer electrolyte system with an ionic liquid, developed for structural supercapacitors by Imperial College London, on a $LiFePO_4$ coated metal foil positive electrode. The negative and positive electrode had copper and aluminium connectors, respectively.

A schematic illustration of a state-of-the-art laminated structural battery composite is presented in figure 2(a). The device consists of two electrodes. The negative electrode is made from carbon fibres. The positive electrode is made from carbon fibres coated with e.g. a lithium metal oxide or phosphate, such as $LiFePO_4$. The two electrodes are separated by an electrically insulating thin film, typically a polymer. The separator needs to be thin to reduce losses from ion currents in the battery. The three plies, i.e. the electrodes and the separator

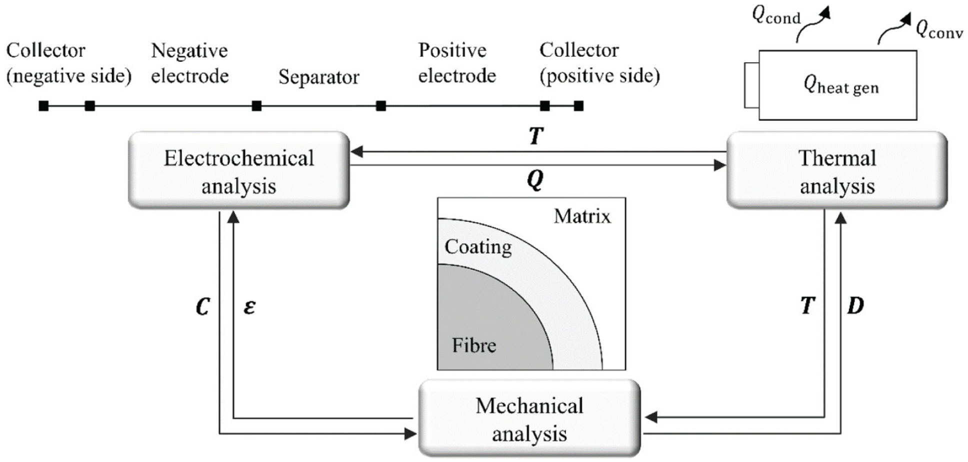

Figure 11. Schematic illustration of the coupled analysis in the multi-physics model, from Carlstedt *et al* (2019a).

are infused by a structural battery electrolyte, as those discussed in section 3. Bi-continuous structural battery electrolytes (Ihrner *et al* 2017, Schneider *et al* 2019) are preferred due their higher multifunctional performance compared to homogenous block co-polymer electrolytes. Finally, current collectors are introduced for electron currents. To date, no data are available for the laminated structural battery device.

5.2. 3D-fibre structural battery

Asp and co-workers (Carlson 2013, Asp *et al* 2016) prepared a semi-structural 3D carbon fibre battery cell, as schematically illustrated in figure 2(b). The battery was made from a tow of approximately one thousand IMS65 carbon fibres. The tow had been coated and dried according to the procedure described by Leijonmarck *et al* (2013). The coating was polymerised in a polytetrafluoroethylene (PTFE) rig from a solution containing monofunctional SR550 monomer from Sartomer, DMF and Li-triflate, leaving grafted polymer chains on the fibres. The battery was built under argon atmosphere where coated fibres made up a structure containing electrode material, current collector and a thin separator. The rig, containing the coated CFs, was attached to an aluminium sheet (current collector), in such a way that the coated CFs were in contact with the metal. A slurry consisting of $LiFePO_4$:Super-P carbon:PVDF 54:34:12 by weight in DMF was applied onto the CFs, where after the DMF was evaporated leaving a positive electrode introduced in contact with both the coated CFs and the aluminium current collector. The 3D-battery was electrochemically cycled at *C*/5. The capacity of this battery was at best 60% of the used $LiFePO_4$ (102 mAh g^{-1} compared to theoretically 170 mAh g^{-1}). Nevertheless, the result is promising for the concept of carbon fibre batteries. However, extensive work remains to up-scale the manufacturing process of this material. Currently, only stamp-sized 3D-batteries, from a few thousand carbon fibres can be made.

6. Modelling

To model the coupled mechanical, thermal and electrochemical processes that occur in a structural battery multi-physics models must be developed. Recently, Carlstedt *et al* (2019a) presented a coupled analysis for structural battery composites, see figure 11. The essential couplings between the physical domains are illustrated as arrows between boxes where $\boldsymbol{T}$ is the temperature field, $\boldsymbol{C}$ is the lithium concentration in the active electrode materials, ε are the mechanical strains, $\boldsymbol{Q}$ is the generated heat due electrochemical cycling and $\boldsymbol{D}$ is the energy dissipation.

To estimate the electrical performance, related to the battery functionality of this material, electrochemical analysis needs to be performed. This analysis allows estimation of the electrical performance of the material related to electrical power, current, etc. During electrochemical cycling heat is generated or absorbed ($\boldsymbol{Q}$) and the lithium concentration in the active electrode materials ($\boldsymbol{C}$) is altered. It has been shown that the volume and elastic properties of electrode materials such as carbon fibres and positive electrode particles are affected by lithium-ion concentration (Qi *et al* 2010, Jacques *et al* 2013b). This means that the battery functionality is coupled with the thermal and mechanical analysis. Moreover, mechanical strains (ε) will affect the electrochemical process, e.g. mechanical fracture of constituents, etc and energy dissipation ($\boldsymbol{D}$) will generate heat. Finally, the heat distribution inside the material and exchange with the surroundings ($\boldsymbol{T}$) will affect both the mechanical and electrical properties of the material. Hence, to estimate the complete performance of the material with respect to the different physical phenomena refined modelling techniques that allow for two-way data exchange between the different physical models are required.

6.1. Constituent-level analysis

6.1.1. *Effect of galvanostatic cycling on 3D-fibre battery constitutive properties.*

Given that the electrode materials undergo significant volume change during electrochemical cycling the elastic properties of the composite may be affected by the state of charge (SOC). Substantial volume change will effectively result in a variation of fibre volume fraction in the composite for different SOC. In addition, the constituents may undergo change in their elastic properties. In fact, since the carbon fibre tow stiffness is constant for different levels of lithiation the fibres longitudinal modulus must be reduced. Jacques *et al* (2013b) indicated that the diameter increases approximately five percent as it becomes fully lithiated. A change in fibre moduli (i.e. E_z, E_r and G_{zr}) may significantly change the composite's mechanical properties. Carlstedt *et al* (2019b) developed an analytical model to investigate how the elastic properties of the 3D-fibre structural battery composite are affected by changes in volume and stiffness of the constituents associated with a change in SOC. Carlstedt *et al* (2019b) present results from a parametric study performed to analyse how different design parameters such as volume fraction of active materials, stiffness of constituents, type of positive electrode material, etc affect the moduli of the structural battery composite for extremes in SOC. Indeed, the composite's transverse elastic properties E_{T} and G_{TT} and the in-plane shear modulus G_{LT} were found to be highly affected by the SOC for critical configurations, while the longitudinal stiffness E_{L} was not. Fibre volume fraction and properties of the coating (stiffness and Poisson's ratio) were identified as critical design parameters. For configurations with a high fibre volume fraction ($V_{\mathrm{f}} \geqslant 0.6$) and Young's modulus of the solid polymer electrolyte coating of 1 GPa or higher a change in transverse stiffness E_{T} up to 30% from electrochemical cycling was predicted. This shows that the effects of SOC on the elastic properties may be crucial to consider when designing a 3D structural battery.

In addition to the volume changes heat will be generated during electrochemical cycling. Potentially, the generated heat will additionally affect the elastic properties of the structural battery composite as well as alter the internal stress state. To date, only one study has been reported which considers diffusion induced stresses in combination with heat generation from ion and electron currents in a structural battery (Carlstedt and Asp 2019). Note that heat generated in a structural battery is generally higher than in conventional batteries due to the lower ion and electron conductivities in the SBE and carbon fibre than in their liquid electrolyte and copper connector counterparts.

Carlstedt and Asp (2019) present predictions of the internal stress state in a 3D-fibre structural battery composite due to a combination of heat generation and volume change of electrode materials occurring during electrochemical cycling. The results show that heat generation during electrochemical cycling must be accounted for when evaluating the internal stress state. For a C-rate of 1 and large lamina dimensions a temperature increase of approximately 35 °C is predicted at full charge. Such temperature increase will significantly affect the internal stress state and change the elastic properties of the structural battery composite.

6.1.2. *Damage analysis*

A multi-scale physics based electrochemical model to describe the 3D-fibre battery containing a coupled system of nonlinear partial differential equations (PDE) was proposed by Xu *et al* (2018a, 2018b). The PDEs describe the spatial and time-dependent variation in Li concentration in the constituents. At the beginning of charging, all available Li is stored in the active material of the matrix. As the charging progresses via electrochemical reactions, lithium ions are transported to the fibre, starting at the coating/matrix interface, see figure 2(b). The difference in Li concentration at the centre of the fibre and the fibre surface is negligible. However, due to low ionic conductivity in the matrix, there is a Li concentration gradient arising in the matrix during charging. This concentration gradient is equalized closer to full charge. At full charge, the Li distribution in both fibre and matrix is uniform.

The insertion expansion-induced stresses in the transversely isotropic carbon fibres are calculated through a thermo-mechanical analogy (Xu *et al* 2018a, 2018b, Xu and Varna 2019, Carlstedt and Asp 2019). Figure 12 shows the stress distributions in the fibre (F), coating (C) and matrix (M) at four instants in time at 1 C-charging in absence of mechanical loads. The initial state, corresponding to completely delithiated fibre and fully lithiated matrix, is assumed stress free.

As shown in figure 12, all stresses are highest at full charge. Consequently, damage is most likely to form at full charge. The high tensile hoop stress, σ_θ, built up during charging indicates that radial cracks may form at the coating/matrix interface. Radial cracks are important as they may contribute to debonding, and even failure, under mechanical in-service loading. Furthermore, such debonding is likely to impair electrochemical performance since Li-ion transport is hindered.

Xu and Varna (2019) used linear elastic fracture mechanics to study propagation of radial matrix cracks and subsequent debond at the coating/matrix interface in a 3D-fibre structural battery composite (figure 13). The load case considered was pure electrochemical loading (1 C charging), i.e. only battery operation with no mechanical load. Crack propagation was studied by calculating energy release rate (ERR) in 2D FEM models assuming square packing of fibres with a radial crack introduced at the SBE/matrix interface.

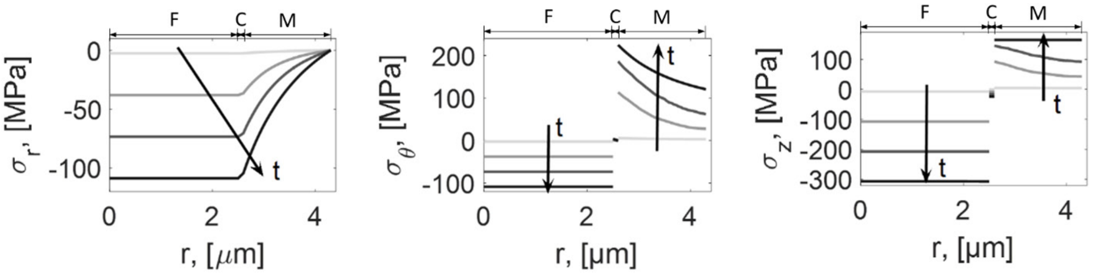

Figure 12. Stress distribution in the constituents during charging. (a) Radial stress. (b) Hoop stress. (c) Axial stress. Radial position, *r*, is zero at the fibre centre Reproduced with permission from (Xu and Varna (2019)).

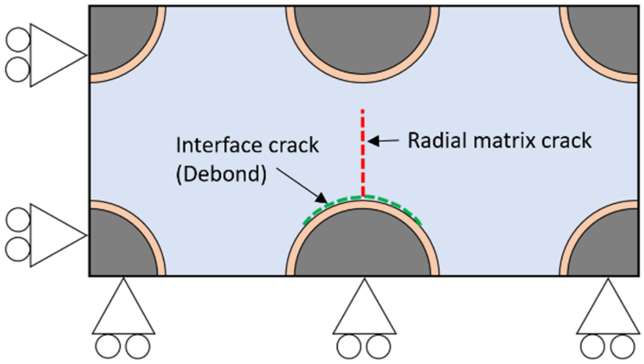

Figure 13. Model geometry and boundary conditions for crack analysis in a 3D-fibre structural battery composite material.

For an electrochemically balanced 3D-fibre structural battery ($V_f = 0.34$), results show that crack growth is unstable. The crack is found to grow towards its closest neighbour. For an undamaged composite, there is no risk for debonding to occur. However, in the presence of a radial crack, large tensile radial and shear stresses at the coating/matrix interface may cause debonding. Results for ERR show that debond crack growth occurs in Mode II. Consequently, the formed interface crack is expected to remain closed, with the constituents remaining in contact through sliding. This may be beneficial for the electrochemical performance of the structural battery.

6.1.3. *Effect of mechanical stress state on ion conductivity of structural polymer electrolytes*

In a very recent study Grazioli *et al* (2019) proposed a coupled electrochemical-mechanical model for analysis of solid polymer electrolytes. They studied the effect of an applied mechanical load on the performance of the solid polymer electrolyte. They found the predicted conductivity of the solid polymer electrolyte to change considerably when a mechanical contribution was included in the analysis. Thus, stresses and deformations need to be accounted for in the design of structural batteries as they are expected to experience high mechanical stresses from in-service loads.

6.2. Laminate level analysis

As discussed above, the carbon fibres in the negative electrode expand and contract during charge and discharge. In a laminated battery there are also positive electrodes, for example carbon fibres coated with an active material (e.g. $LiFePO_4$ particles). These particles will also expand and contract during charge and discharge. Thus, both the negative and positive electrodes will expand and contract, however, not the same amount and not the same in the fibre direction as in the transverse direction.

An analytical model was developed for the prediction of the shape change of an arbitrary laminated structural battery laminate by Dionisi *et al* (2017). The aim of this model was to be able to make lay-ups that would minimise overall deformations of the battery laminate by alternating the stacking sequence, and possibly also by finding elaborate schemes for charging and discharging the cells individually. The aim was also to predict the free edge stresses which could potentially lead to delaminations in the laminate. The model was based on an extension of classical lamination theory and leads to closed form solutions of deformation, strains and stresses, although the stress solution eventually must be solved numerically.

In terms of overall deformation of the laminated structural battery composites one must of course rely on symmetric stacking sequences in order to avoid bending (ensuring a zero $\boldsymbol{B}$-matrix). Still, it is not possible to avoid any in-plane deformation although they can be smaller or larger depending on the stacking. Regarding

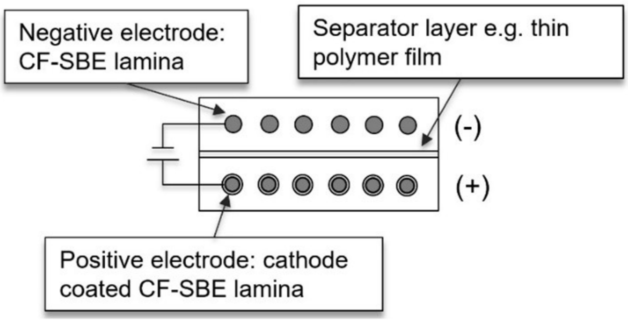

Figure 14. Schematic representation of a laminated battery cell.

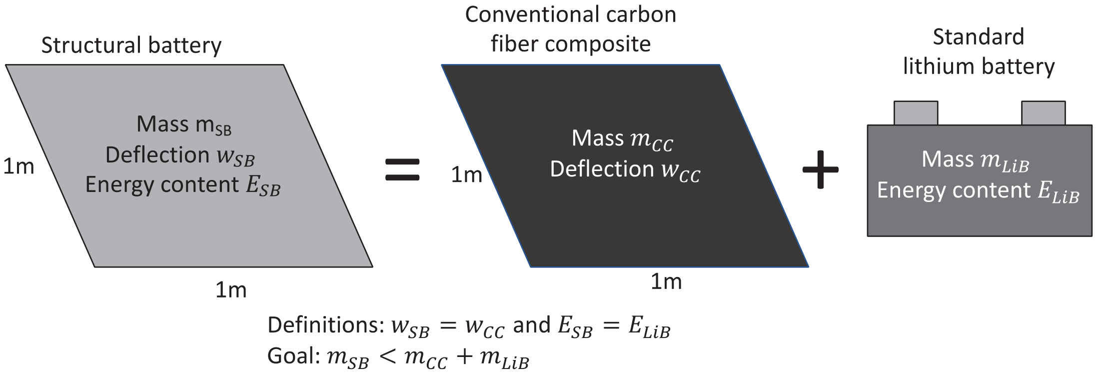

Figure 15. Schematic representation of the modelling of multifunctional performance for a stacked structural battery laminate reproduced from (Johannisson *et al* (2019)). © IOP Publishing Ltd. CC BY 3.0.

Table 2. Mass saving potential for structural battery composites (data from Johannisson *et al* (2019)).

Application	Today's design	Weight saving potential 'target' (in %)
Aircraft interior panel	Glass fibre composite sandwich structure	67
Car roof in steel	Steel 0.8 mm	62
Hull of electric ferry	Carbon fibre composite sandwich structure	11
Lap-top casing	Aluminium	38

interlaminar stresses it was seen that it is beneficial to place the negative electrodes on the outside of the laminate and keeping the positive electrodes closer to the middle surface, at least for simple cross-ply laminates.

6.3. Conceptual battery design and multifunctional performance

The laminated battery cell is a material that can potentially save significant weight on a systems level compared to separate components for structural and battery functions. As schematic representation of a laminated battery cell is illustrated in figure 14.

In this set-up the negative electrode lamina consists of structural battery electrolyte (SBE) developed by Ihrner *et al* (2017) reinforced with carbon fibres. The positive electrode lamina also consists of SBE but is instead reinforced with lithium-based electrode material (e.g. $LiFePO_4$) coated carbon fibres. A separator layer is also needed between the two laminae to avoid short-circuit. The mechanical and electrical performance of the laminated structural battery composites can be assessed by estimating the in-plane elastic properties of the laminate using Classical Laminate Theory and the specific capacity and energy density of the complete component.

Based on these performance metrics it is possible to estimate the overall multifunctional performance of the laminated battery cell and to compare the performance with that of monofunctional based systems. To identify the key parameters to improve multifunctionality and their impact on the performance, the framework has been extended by refining the modelling of the mechanical and electrical performance and defining parametric variables for the design parameters of the battery cell. To identify possible weight reductions the performance of the structural battery is compared with the performance of the separate structural and battery components.

Here, a structural battery is modelled with the dimensions 1 m × 1 m, built up by stacked positive and negative electrodes with SBE. The structural battery has a known mass m_{SB} and energy storage E_{SB}, see figure 15. This structural battery is then loaded with a distributed pressure and simply supported boundary conditions which results in a deflection at its midpoint (w_{SB}) to find a single stiffness metric for the laminate. For comparison a state-of-the-art carbon fibre and epoxy composite plate is modelled to have the same deflection due to the same load (same global stiffness), as well as a standard lithium ion battery with the same energy content as the structural battery. Then one can find the mass of the two monofunctional systems m_{CC} and m_{LiB} that performs like the multifunctional structural battery. The goal of the modelling is to find parameters which maximize the mass saving potential of the structural battery with respect to the separate structure and battery.

Potential mass saving offered by replacing conventional carbon fibre composite and standard lithium ion battery with a structural battery can be estimated for any application. In a recent study Johannisson *et al* (2019) employed this model and compared weight savings for several scenarios. The benchmark model uses targeted performance metrics (conductivity, thicknesses, rate dependence, capacities, etc) for the structural battery. Mass saving estimates were presented for several applications; a composite aircraft interior panel, a car roof made from steel, a carbon fibre composite boat hull and an aluminium lap-top casing. The mass saving by using a multifunctional structural battery, compared to a monofunctional structural material and a separate lithium ion battery is given by the mass saving in percent by:

$$\mathrm{MS}_{\%} = \frac{m_{CC} + m_{LiB} - m_{SB}}{m_{CC} + m_{LiB}} \cdot 100.$$

The results are very promising leading the mass saving potential estimates given in table 2.

7. Concluding remarks

The structural battery composite is a novel lightweight material. Not only is the density of the composite material constituents low, these constituents also demonstrate exceptional ability to perform two vital functions for lightweighting. Firstly, carbon fibres are stiff and strong, with unprecedented specific mechanical properties. Carbon fibres also have exceptional electrochemical characteristics, with very high specific capacity and coulombic efficiency. Secondly, structural battery electrolytes are now available that can efficiently transfer mechanical loads and transport Li-ions. As consequence, numerous structural battery composite architectures can now be realised. Having said that, there is still little research performed on how to design and manufacture positive structural electrodes. Similarly, only a few attempts to make structural battery full cells have been reported to date. This is partly due to the lack of structural positive electrodes. Finally, new modelling approaches, considering the coupled thermomechanical-electrochemical problem are being developed. Multifunctional performance analyses with such tools rely on an extensive data base, which is currently lacking.

In this paper we provide an overview of research performed on structural battery composites to date. Substantial work has been performed. However, significant work on the structural battery full cell and its manufacture and validation remains before structural battery composites offer a solution for mass-less energy storage in future electric road, air and sea transport applications. To date, no show-stoppers for the realisation of structural battery composites have been identified. Nevertheless, issues related to up-scaling of the manufacture process, power management and connectivity as well as recycling are still to be addressed (Asp and Greenhalgh 2015).

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