

THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY IN SOLID AND
STRUCTURAL MECHANICS

Computational modelling of structural battery composites

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Cover:

Schematic illustration of a structural battery composite specimen and the coupled thermo-electro-chemo-mechanical problem for which a computational framework is developed in this thesis.

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ABSTRACT

Batteries and surrounding structures (e.g. battery modules and packs) in electrical vehicles and devices are often designed in a way that prevents the electro-chemically active part of the battery cells from being exposed to mechanical loads during operation/service. This means that the energy storage capability is added as a monofunctional addition to the system (i.e. it only provides one functionality, storing energy). Hence, one of the main drawbacks of the existing technology is its energy storage to weight ratio, in terms of the complete system. A viable route to improve this ratio is to develop energy storage solutions with the ability to sustain mechanical loads. Indeed, by adding this additional functionality, such solutions offer significant system mass and volume savings and allow for innovative future design of electric vehicles and devices.

The structural battery composite material is made from carbon fibre reinforced structural battery electrolyte (SBE), and exploits the multifunctional capability of the material constituents to facilitate electrical energy storage in structural components. Due to its inherent multifunctionality, the physical phenomena occurring within the material during operation will interact. Further, due to the fact that the studied material is intended to perform multiple functions some of the couplings between the physical processes are expected to be more pronounced, and critical to design, as compared to conventional batteries. Hence, to accurately predict and evaluate the combined performance of structural batteries, coupled multiphysics models are needed.

In this thesis, a computational modelling framework to predict the coupled thermo-electro-chemo-mechanical performance of structural batteries is developed. The framework is utilized to study the essential couplings between the physical processes and numerical predictions are compared favourably with experimental data. It is shown that two-way coupling between the electro-chemical and mechanical processes is important to account for when evaluating the combined electro-chemo-mechanical performance of structural batteries. Further, it is shown that the convective contribution to the mass flux of ions in the SBE, as well as the thermal effects during operations are crucial to consider when evaluating the combined performance. Moreover, the framework is extended to study an electro-chemically driven actuator and sensor utilizing carbon fibre-SBE electrodes. Finally, in addition to the modelling work a laminated structural battery with unprecedented multifunctional (i.e. combined mechanical and electro-chemical) performance is manufactured and characterized, featuring an energy density of 24 Wh kg^{-1} and an elastic modulus of 25 GPa and tensile strength exceeding 300 MPa.

Keywords: Li-ion batteries, Carbon fibre composites, Multifunctional materials, Thermo-electro-chemo-mechanical coupling, Finite Element Analysis (FEA)

"Curiosity is the starting point for great science."
– Philip Kotler –

PREFACE

The work in this thesis was carried out from April 2017 to March 2022 at the Division of Material and Computational Mechanics, Department of Industrial and Materials Science, Chalmers University of Technology. The research was financially supported by the European Union, Clean Sky Joint Undertaking 2, Horizon 2020, Grant Agreement no. 738085, USAF, contracts FA9550-17-1-0338 and FA8655-21-1-7038, Swedish National Space Agency, Grant Agreement no. 2020-00256, and the Swedish Research Council (VR), Grant Agreement no. 2020-05057.

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Gothenburg, March 2022
David Carlstedt

THESIS

This thesis consists of an extended summary and the following appended papers:

- Paper A** L.E. Asp, K. Bouton, **D. Carlstedt**, S. Duan, R. Harnden, W. Johannisson, M. Johansen, M. Johansson, G. Lindbergh, F. Liu, K. Peuvot, L.M. Schneider, J. Xu, D. Zenkert. A structural battery and its multifunctional performance. *Advanced Energy and Sustainability Research*, Vol. 2, 2021, 2000093.
- Paper B** **D. Carlstedt**, K. Runesson, F. Larsson, J. Xu and L.E. Asp. Electrochemo-mechanically coupled computational modelling of structural batteries. *Multifunctional Materials*, Vol. 3, 2020, 045002.
- Paper C** **D. Carlstedt**, K. Runesson, F. Larsson, V. Tu, R. Jänicke and L.E. Asp. Computational modelling of structural batteries accounting for stress-assisted convection in the electrolyte. *International Journal of Solids and Structures*, Vol. 238, 2022, 111343.
- Paper D** **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 electrodes. *Composites Science and Technology*, Vol. 220, 2022, 109283.
- Paper E** **D. Carlstedt**, K. Runesson, F. Larsson and L.E. Asp. On the coupled thermo-electro-chemo-mechanical performance of structural batteries with emphasis on the thermal effects. *Submitted for publication*.
- Paper F** **D. Carlstedt**, K. Runesson, F. Larsson, R. Jänicke and L.E. Asp. Hierarchical modelling of a structural sensor-actuator comprising beam action and electro-chemical-mechanical interactions. *Manuscript*.

The appended papers in Part II of the thesis were prepared in collaboration with the co-authors. In **Paper A** the author of this thesis was responsible for the major progress of the work associated with manufacturing and electro-chemical characterization, including writing the original drafts for these sections. In **Paper B-F** the approach and methodology originate from discussions among the authors. The majority of the work was conducted by the thesis author, e.g. took part in formulating the theory, led the planning and writing of the papers, and carried out the numerical implementation and simulations. Further, the experimental studies in **Paper D** were planned and partly executed by the author.

In Part III of the thesis a corrigendum related to equation (3) in **Paper B** is presented. The correction was discovered after publishing. However, the correct expressions were used in the simulations. Hence, the results and conclusions in this paper are unaffected by this error.

Other publications related to thesis:

1. W. Johannisson, N. Ihrner, D. Zenkert, M. Johansson, **D. Carlstedt**, L.E. Asp, F. Sieland, Multifunctional performance of a carbon fiber UD lamina electrode for structural batteries. *Composites Science and Technology*, Vol. 168, 2018, pp. 81-87.
2. **D. Carlstedt**, E. Marklund, L.E. Asp, Effect of state-of-charge on elastic properties of 3D structural batteries. *Composites Science and Technology*, Vol. 169, 2019, pp. 26-33.
3. **D. Carlstedt**, L.E. Asp, Thermal and diffusion induced stresses in a structural battery under galvanostatic cycling. *Composites Science and Technology*, Vol. 179, 2019, pp. 69-78.
4. **D. Carlstedt**, L.E. Asp, Performance analysis framework for structural battery composites in electric vehicles. *Composite Part B*, Vol. 186, 2020, 107822.
5. W. Johannisson, **D. Carlstedt**, A. Nasiri, C. Buggisch, P. Linde, D. Zenkert, L.E. Asp, G. Lindbergh, B. Fiedler, A screen-printing method for manufacturing of current collectors for structural batteries. *Multifunctional Materials*, Vol. 4, 2021, 035002.
6. S. Duan, A.H.S. Iyer, **D. Carlstedt**, F. Rittweger, A. Sharits, C. Maddox, K.R. Riemschneider, D. Mollenhauer, F. Liu and L.E. Asp, Effect of lithiation on the elastic moduli of carbon fibres. *Carbon*, Vol. 185, 2021, pp. 234-241.
7. J. Xu , Z. Geng, M Johansen, **D. Carlstedt**, S. Duan, T. Thiringer, F. Liu, L.E. Asp. A multicell structural battery composite laminate. *EcoMat*, 2022, e12180.
8. R. Harnden, **D. Carlstedt**, D. Zenkert, G. Lindbergh, Multifunctional carbon fibre composite: a structural, energy harvesting, strain-sensing material. *Submitted for publication*.

Conference papers:

- **D. Carlstedt**, W. Johannisson, D. Zenkert, P. Linde and L.E. Asp. Conceptual design framework for laminated structural battery composites. In: Proceedings of the ECCM18, 2018, Athens, Greece.
- **D. Carlstedt**, J. Xu, K. Runesson, F. Larsson and L.E. Asp. Unit cells for multiphysics modelling of structural battery composites. In: Proceedings of the ICCM22, 2019, Melbourne, Australia.

Additional publications by the author:

- M. Alves, **D. Carlstedt**, F. Ohlsson, L.E. Asp and S. Pimenta. Ultra-thin and stiff randomly-oriented discontinuous composites. In: Proceedings of ASC34, 2019, Atlanta, US.
- M. Alves, **D. Carlstedt**, F. Ohlsson, L.E. Asp and S. Pimenta. Ultra-strong and stiff randomly-oriented discontinuous composites: closing the gap to quasi-isotropic continuous-fibre laminates. *Composite Part A*, Vol. 132, 2020, 105826.

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Part I

Extended Summary

1 Introduction

1.1 Background

In recent years, there has been a growing interest in energy storage solutions which can enable pathways for energy efficient and sustainable transportation. Today, one of the dominating solutions for energy storage in electrical vehicles and devices are lithium-ion (Li-ion) batteries [1–3]. These batteries and surrounding structures (e.g. battery modules and packs) are often designed in a way that prevents the electro-chemically active part of the batteries from being exposed to mechanical loads during operation/service. This means that the energy storage capability is added as a monofunctional addition to the system (i.e. it only provides one functionality, storing energy). Hence, one of the main drawbacks of the existing technology is its energy storage to weight ratio [1, 2, 4, 5], in terms of the complete system. A viable route to improve this ratio is to develop energy storage solutions with the ability to sustain mechanical loads [6–24], i.e. to provide an additional functionality (mechanical integrity). Indeed, combining these functionalities, such solutions offer significant system mass and volume savings [6, 25–28] and allow for innovative future design of electric vehicles [25] and devices (e.g. laptops and phones) [28], as well as autonomous robotics [20], unmanned aerial vehicles [13, 23] and satellites.

Energy storage solutions in the form of batteries with ability to sustain mechanical loads are often referred to as structural batteries. The basic concept of the structural battery is illustrated in Figure 1.1, which shows the application to an electric vehicle. The basic idea is to allow the structural system to store energy while simultaneously providing mechanical integrity. It should be noted that in the literature, two types of structural battery concepts are often reported [26]: (i) *multifunctional materials* and (ii) *multifunctional structures*. The difference between multifunctional materials and multifunctional structures is that in the former the constituents perform multiple functions, which means that the material itself is multifunctional, while in a multifunctional structure monofunctional subcomponents are combined. An example of the latter solution is embedding thin-film batteries within structural components [29–34].

In this thesis, focus is on multifunctional materials, in particular material solutions based on carbon fibre reinforced polymers, which exploit the multifunctional capability of the material constituents to facilitate electrical energy storage in structural components. Further, the focus has been on the so-called *laminated structural battery* architecture. The internal structure of a conventional Li-ion battery and the laminated structural battery are shown schematically in Figure 1.2 for comparison. The main differences between the structural battery and a conventional Li-ion battery design are essentially threefold: (i) The active electrode materials are fibres instead of particles; (ii) An electrolyte, referred to as Structural Battery Electrolyte (SBE), which enables ion transport between the

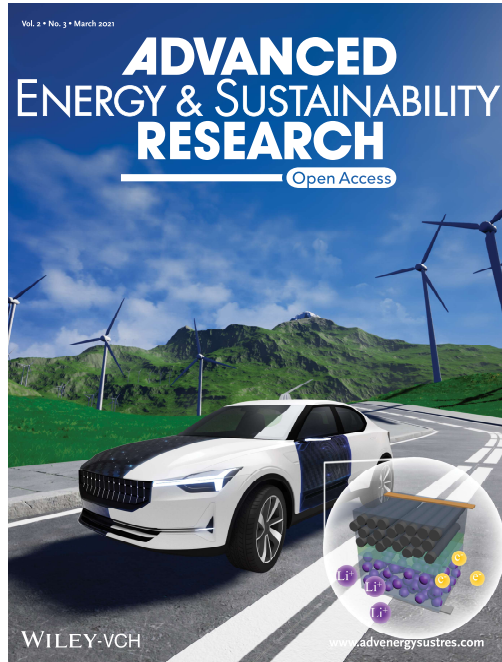


Figure 1.1: The structural battery composite concept: a multifunctional composite material capable of simultaneously carrying mechanical load and storing energy (i.e. work as a battery). This image is a reprint of the back cover page for **Paper A**, [16].

electrodes while distributing mechanical loads is used instead of liquid electrolyte; (iii) The carbon fibres work as current collectors. In this design the individual laminae correspond to different components in a battery cell (electrodes, separator, etc.). When these laminae are stacked into a laminate, the resulting material provides mechanical properties that are similar to those of conventional fibre reinforced polymer composites that are commonly used in structural applications. The laminated design concept was first proposed by Wetzel and co-workers [6] at the US Army Research Laboratory (ARL) and the first attempt to make such a material was done by Wong et al. [8]. This attempt, however, resulted in a material with decent mechanical performance but with no electro-chemical functionality. Further, this material concept was later demonstrated by Liu et al. [9], Ekstedt et al. [10], and Carlson [11]. However, the combined electro-chemical and mechanical (i.e. multifunctional) performance remained inferior to comparable monofunctional systems when this project started. Hence, the material concept had not been demonstrated as a competitive alternative to conventional design concepts utilizing monofunctional components at that time.

Given the fact that the structural battery composite material is relatively new, significant challenges are identified from scientific as well as from engineering points of view. One of the main challenges, which remained to be investigated, was the materials combined performance during operation. Due to its inherent multifunctionality, the

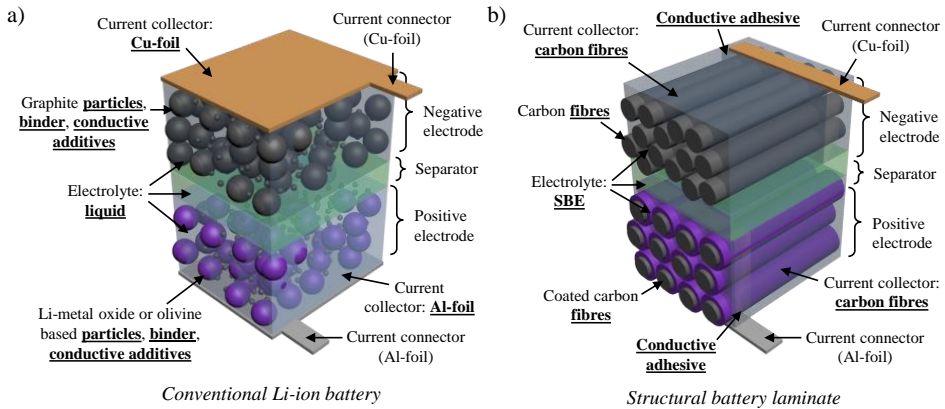


Figure 1.2: Schematic illustration of (a) a conventional Li-ion battery and (b) the laminated structural battery. The bold underlined text represents the main differences between the two types of battery cells. SBE = Structural Battery Electrolyte.

physical phenomena occurring within the structural battery material during operation will interact. A schematic illustration of the coupled analysis is presented in Figure 1.3. As for conventional batteries (cf. [35]), the physics involved is associated with the electro-chemical, mechanical and thermal processes (boxes in Figure 1.3). Further, the different fields are coupled (arrows in Figure 1.3) which leads to a highly complex multiphysics problem with a high number of unknown dependent variables, e.g.: chemical potentials (μ_α), concentrations (c_α), electric potential (φ), displacements (\mathbf{u}), pore pressure (p), and temperature (θ). Moreover, due to the fact that the studied material is intended to perform multiple functions some of the essential couplings are expected to be more pronounced, and critical to design, as compared with conventional batteries. Hence, to accurately predict and evaluate the combined thermo-electro-chemo-mechanical performance of structural batteries, we needed coupled multiphysics models. Such models were lacking at the start of this project.

The current research is part of a large research initiative on structural batteries with the overall aim to advance the maturity of this material. This research initiative includes a series of collaborating projects such as the SORCERER [36] project, as well as projects funded by USAF [37], Swedish National Space Agency [38] and Swedish Research Council (VR) [39]. These projects build upon, and are continuations of, earlier work done within e.g. the STORAGE [40] and KOMBATT [41] projects.

1.2 Aim, scope and limitations

The main focus of this thesis is to develop computational modelling tools to predict the coupled thermo-electro-chemo-mechanical performance of structural batteries. Further, the thesis aims at performing initial experimental studies for model validation and, as part of the large research team within the research initiative, develop and characterize

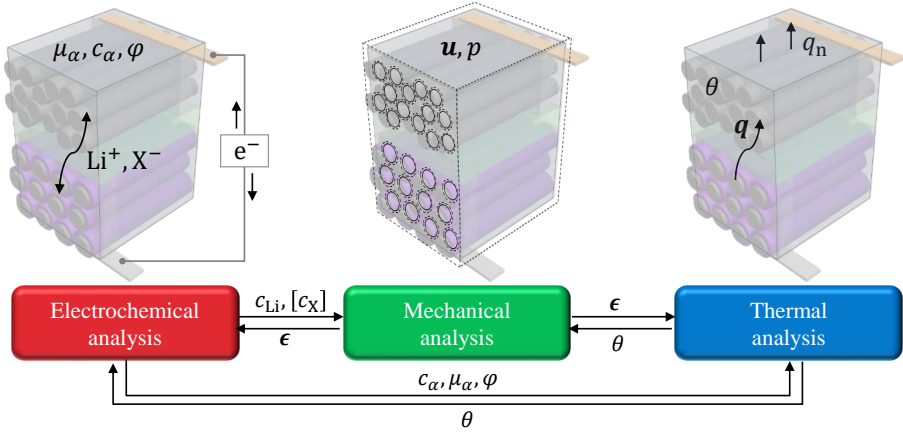


Figure 1.3: Schematic illustration of the coupled analysis in the multiphysics model for structural battery composites. The introduced notation is defined in Sections 3 and 4.

next generation structural batteries.

The aims for this thesis are summarized as follows.

- Manufacture and characterize material design solutions for *next generation* structural batteries. In **Paper A**, a novel material design is proposed, manufactured and characterized with respect to its combined electro-chemical and mechanical (multifunctional) performance.
- Develop a computational modelling framework to predict the coupled thermo-electro-chemo-mechanical performance of structural batteries. The original formulation of the framework is described in **Paper B**, and extended to account for seepage of the liquid phase of the SBE in **Paper C**, and thermal effects in **Paper E**.
- Perform initial experimental validation of the developed framework. Experimental studies on carbon fibre-SBE vs. Li-metal half-cells are performed and numerical predictions (utilizing the developed framework) are compared with experimental data in **Paper D**.
- Extend the modelling framework to study additional functionalities of composite materials made from stacked carbon fibre-SBE electrode laminae. In **Paper F** the numerical framework is extended and utilized to predict the coupled performance of an electro-chemically driven actuator and strain-sensing material made from carbon fibre-SBE electrodes. It should be noted that from a theoretical perspective, replacing the reference (Li-metal) electrode with the additional fibre electrode results in a model description similar to a so-called *full-cell*.

The experimental work in **Paper A** is limited to structural battery full-cell designs utilizing carbon fibres against conventional Lithium Iron Phosphate (LFP) foils (as positive electrode), see Section 2.2. Moreover, experiments in **Paper D** are performed on carbon fibre-SBE vs. Li-metal half-cells and the comparison with numerical predictions is limited to the electro-chemical performance. Only basic electro-chemical (e.g. energy density) and mechanical (e.g. stiffness and strain to failure) characteristics are measured.

The theory in **Papers B-F** is limited to small strain kinematics. With respect to mechanical properties, the material response of the constituents are assumed to be linear (i.e. linear elastic material response). With respect to the electro-chemical response on the other hand, nonlinearities are considered. Self-weight and any piezoelectric effects are ignored. The material representation is resolved at *micrometre scale*, i.e. the carbon fibres are discretized while effective (homogenized) properties of the SBE are utilized. Further, the model description is limited to idealized two-dimensional material representations. It should be noted that simplistic constitutive relations and idealized material representations are utilized due to the main focus of the modelling work being "proof-of-concept".

Finally, in **Paper F** the electro-chemically driven actuator and strain-sensing composite material made from stacked carbon fibre-SBE electrodes was chosen as the "full-cell" design to study in this thesis. The coated carbon fibre positive electrode design (cf. Figure 1.2b) was not studied due to its low maturity, uncertainty in design, lack of material data and complex structure (e.g. the involved spatial scales).

2 Material architecture and applications

2.1 Carbon fibre-SBE electrode

A schematic illustration of a carbon fibre-SBE electrode (i.e. the negative electrode in Figure 1.2b) is presented in Figure 2.1a. The utilized SBE consists of two phases: a solid phase, which corresponds to a mechanically robust porous polymer network, and a liquid phase, which contains liquid electrolyte with Li-salt (Figure 2.1b). The liquid phase in the porous polymer network enables ion transport between the electrodes, while the solid phase makes it possible to distribute mechanical loads. In Figure 2.1c the hierarchical structure of the carbon fibre-SBE electrode is shown to illustrate the different length scales.

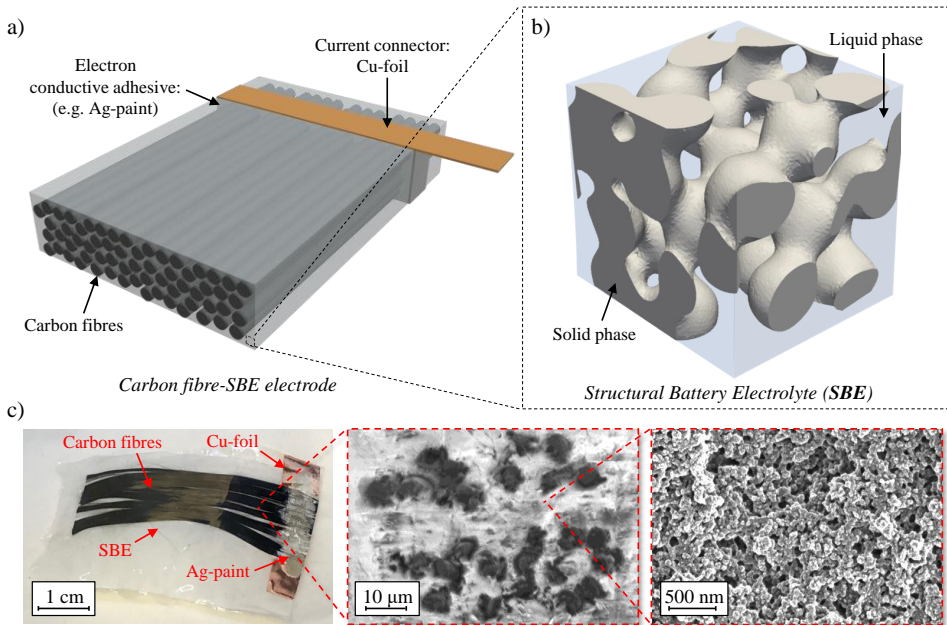


Figure 2.1: (a) Schematic illustration of the carbon fibre-SBE electrode. (b) Numerically generated porous bi-continuous nano-structure, from Tu et al. [42], representing an idealized fine-scale geometry of the Structural Battery Electrolyte (SBE). (c) Hierarchical structure: Photo of electrode (length scale: cm), SEM image of electrode cross-section (length scale: μm), SEM image of SBE (length scale: nm).

This type of electrode laminae can be used in laminated structural batteries or in electro-chemically driven structural actuators [43] and energy harvesting and strain sensing composite materials [44–46] (Figure 2.2). The battery application is studied in **Paper A-E** while the actuator and sensor applications are studied in **Paper F**. The particular material designs and applications studied in this thesis are discussed in the following.

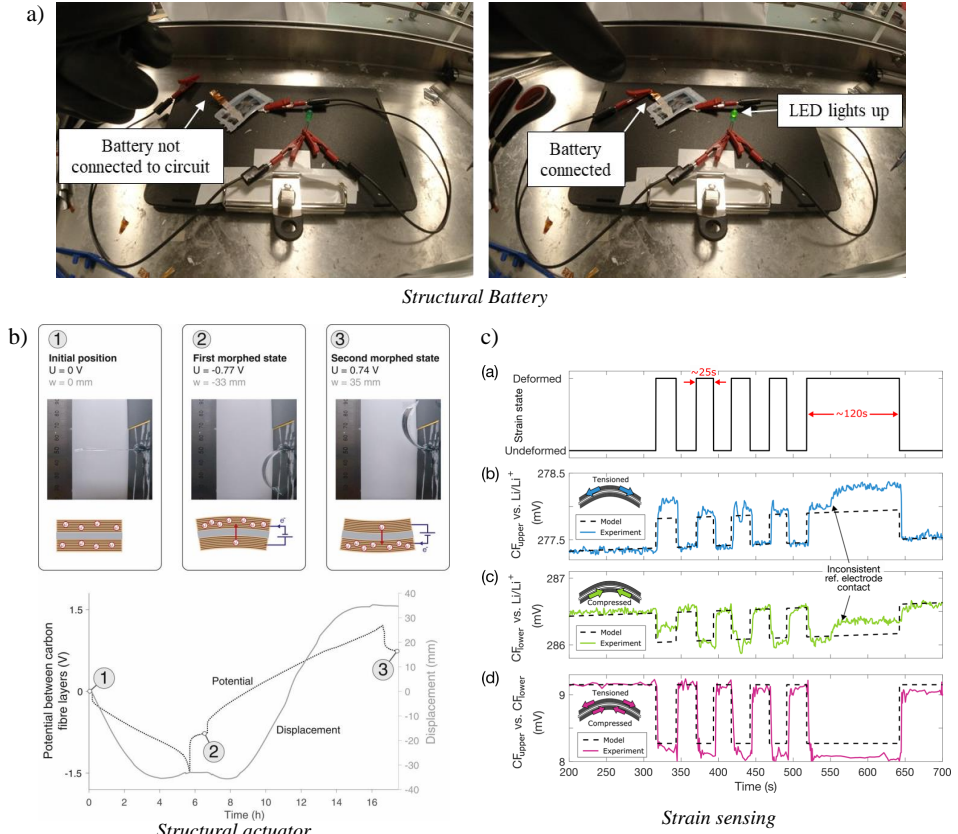


Figure 2.2: Carbon fibre-SBE electrodes utilized in different applications: (a) Structural battery (images from **Paper A**, [16]), (b) structural actuator (image from [43]), and in (c) structural energy harvesting and strain sensing material (image from [47]).

2.2 Laminated structural battery

The conceptual microstructure of the laminated structural battery is illustrated in Figure 1.2b. In this design, the carbon fibres in the negative electrode function as combined electrode material, current collector and reinforcement while in the positive electrode the carbon fibres are coated with lithium metal oxide or olivine based particles, e.g. LiFePO_4 , binder and conductive additives (see e.g. [48–50]). Hence, in the positive electrode the carbon fibres function as current collectors and reinforcement. The two electrodes are separated by a separator layer which assures that the electrode materials in the two electrodes do not come in contact (i.e. to avoid short-circuit). The fibres in the negative electrode and the particles in the coating in the positive electrode act as active electrode materials (hosts for the lithium).

As mentioned above, one of the main differences between the structural battery and a conventional Li-ion battery design is that continuous fibres are used instead of particles. This means that there is no need for a polymeric binder, nor conductive additives, in the carbon fibre-SBE electrode (i.e. the upper surface of the fibre electrode in Figure 1.2a does not need to be fully covered by metal foil). It should, however, be noted that the electronic conductivity of carbon fibres [51] is about three orders of magnitude lower than that of copper. Hence, the ohmic losses in the studied material will be significantly larger and will limit the electrode dimensions. Further, conductive adhesive is used in **Paper A** and **Paper D** to adhere the carbon fibres to the metal foil current connectors allowing for efficient transfer of electronic current between the fibres and the foil (cf. Figures 2.1a and c). In [52] (Paper 5 in the list of publications not included in the thesis), we demonstrate how screen-printing technology can be used to print current connectors along the fibres. This does not resolve the issue of the low conductivity, but by utilizing a certain printing pattern the electron transport along the fibres can be reduced/limited. Furthermore, it is noted that reported specific capacities and diffusion coefficients for certain types of carbon fibres are similar to those of commercially available graphite electrode materials [51, 53–55]. Hence, in combinations with their high specific mechanical properties, carbon fibres are well suited for multifunctional applications.

The laminated structural battery architecture allows for the individual laminae to be stacked in different configurations, as often done in conventional carbon fibre reinforced polymer (CFRP) composite laminates. This offers further tailoring of the design to provide improved multi-axial mechanical performance. As the fibres and connectors in the laminae in Figure 1.2b are electrically conductive, they need to be insulated from the surroundings (neighbouring plies). The structural battery composite is also sensitive to moisture and oxygen (like ordinary Li-ion batteries). In **Paper A** and **Paper D**, the battery cells are vacuum heat sealed in pouch bag’s to allow the cells to be tested outside the glovebox (photos in Figure 2.2a are taken from inside the glovebox).

The material design proposed in **Paper A** is illustrated schematically in Figure 2.3a. This corresponds to the design utilized for the structural battery shown in Figure 2.2a. In this design, the positive electrode was made from commercially available LFP-foil due to the low maturity of the coated carbon fibre solution in Figure 1.2b (cf. [48, 49]). The negative electrode was made from ultrathin spread tow UD tapes of T800SC-12k-50C PAN-based carbon fibres kindly provided by Oxeon AB. Further, two types of separators were used: (i) one made from Whatman micro glass fibre (GF) filter and (ii) one made from a stack of two $0^\circ/90^\circ$ woven GF fabrics (denoted GF plain weave). The layup was impregnated and cured with an SBE solution as described in [16], developed by Ihrner et al. [56] and Schneider et al. [57].

In **Paper B-E**, the theoretical analysis and experimental investigation were simplified by using the similarity of the two electrodes in the laminated architecture: both the negative and positive electrodes consist of fibres embedded in a SBE-based matrix material. For this reason, we studied the negative half-cell schematically illustrated in Figure 2.3b. Compared with the so-called full-cell in Figure 2.3a, the positive electrode is merely replaced with Li-metal (working as combined counter and reference electrode). This corresponds to the battery cell that have been studied experimentally in [12] (Paper 1 in the list of publications not included in the thesis) and in [56, 57].

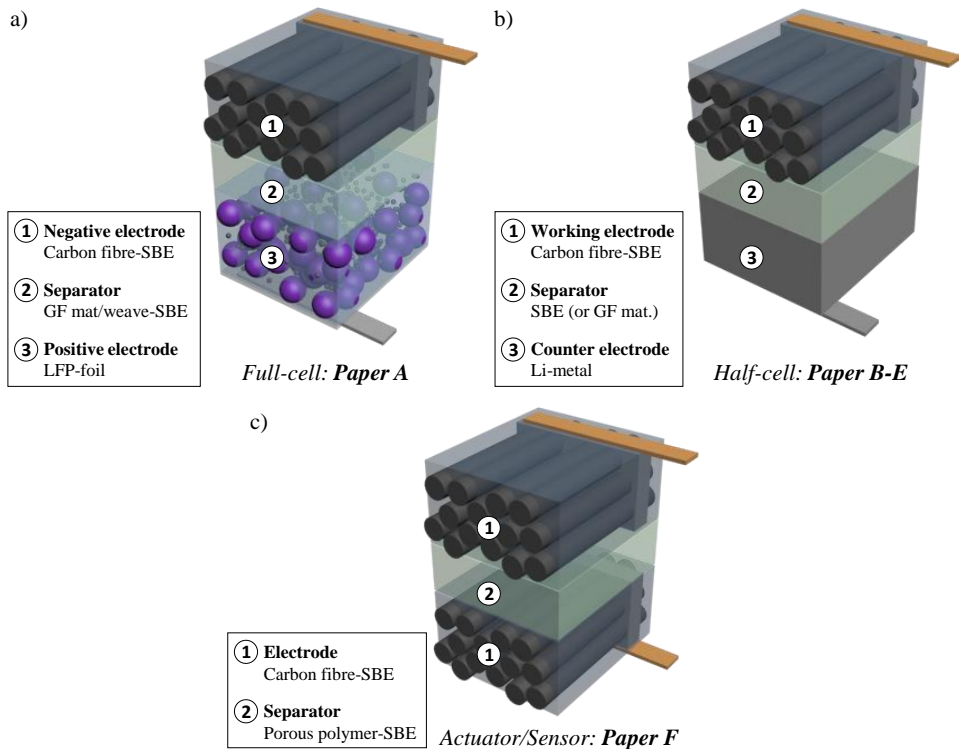


Figure 2.3: Schematic illustrations of: (a) Full-cell material design proposed and characterized in **Paper A**. (b) Negative half-cell studied in **Paper B-E**. (c) Electro-chemically driven structural actuator and strain sensing composite material studied in **Paper F**.

2.2.1 Multifunctional performance

Since the structural battery composite material provides multiple functions, the mechanical and electro-chemical performance need to be considered simultaneously. In **Paper A**, we evaluated the mechanical and electro-chemical (i.e. multifunctional) performance in terms of the materials elastic moduli and energy density, respectively. These performance metrics are measured by means of tensile tests and repeated galvanostatic cycling, respectively, as described in the paper. It should be noted that this is in line with performance metrics utilized by others in the scientific community to evaluate the multifunctional performance of structural batteries, see e.g. [58, 59]. Figure 2.4, shows the elastic modulus and cell level energy density of reported structural battery composites. In this figure, *Current study* refers to **Paper A** and the image is a reprint from the paper, i.e. [16]. This graph also illustrates the significant advance in combined mechanical and electro-chemical performance of the developed technology in **Paper A**. The references in Figure 2.4 are: (i) "Meng et al. [27]" refers to [60]; (ii) "Thakur and Dong [26]" refers to [61]; (iii) "Moyer et al. [24]" refers to [15]; (iv) "Liu et al. [17]" refers to [9].

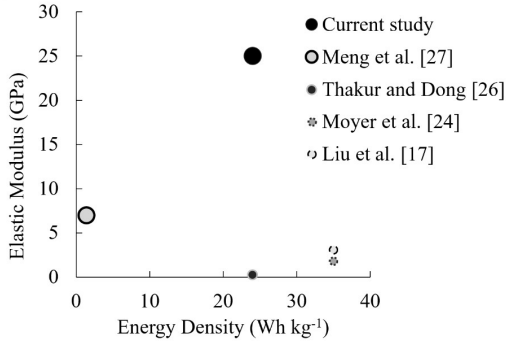


Figure 2.4: (a) Graph showing the elastic modulus and cell level energy density of reported structural battery composites, numbered by their references. *Current study* refers to **Paper A** and the image is a reprint from this paper, i.e. [16].

2.3 Actuator and sensor application

The conceptual material design of the composite actuator and sensor studied in **Paper F** is schematically illustrated in Figure 2.3c. The cross-sectional design is composed of two (partly) lithiated carbon fibre-SBE electrodes on either side of a separator. This design corresponds to the material which has been studied experimentally in [43, 46]. The actuation and sensor functionalities are achieved by the interplay between electro-chemistry and mechanics, in particular the ability of carbon fibres to (de)insert Li-ions repeatedly and the coupling between the chemical potential of Li in the fibres and mechanical stress state (see Section 3).

2.4 3D-battery (or Micro-battery)

Even though this thesis is limited to the study of the laminated structural battery, the so-called *3D-battery* or *Micro-battery* architecture is studied in e.g. [62, 63], i.e. Paper 2 and 3 in the list of publications not included in the thesis. In these studies, semi-analytical models are developed and the analysis are limited to one-way coupling (i.e. electro-chemical analysis provides input to the mechanical analysis). In the 3D-battery design the individual carbon fibres are coated with a thin homogeneous polymer electrolyte coating and work as individual battery cells in the composite laminae. This design was first developed by Asp and co-workers [11, 26, 64].

3 Coupled processes in structural batteries

Due to its inherent multifunctionality, the physical phenomena occurring within the structural battery material during operation will interact (illustrated in Figure 1.3). The coupled processes in the studied material are discussed in some detail in this section.

3.1 Electro-chemical cycling (battery function)

During electro-chemical cycling (i.e. charging or discharging the battery) electrical energy is converted to chemical energy (or vice versa) via redox reactions [65]. These redox reactions are reduction and oxidation processes that occur on the surfaces of the electrode materials during charge/discharge. Accompanying these reactions is the transport of

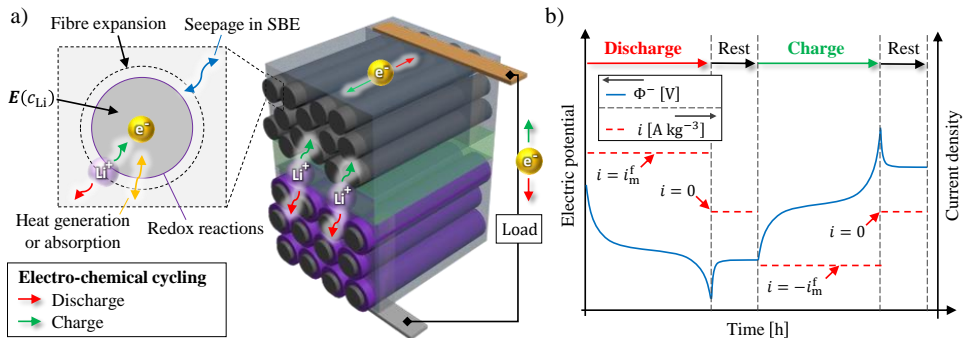


Figure 3.1: (a) Schematic illustration of the coupled processes during electro-chemical cycling. (b) Electric potential and applied current density versus time, illustrating the charge/discharge conditions (battery function).

charge species, i.e. electrons and ions moving between the electrodes (Figure 3.1a). The electrons are driven by the electric potential difference between the electrodes (migration) while the ions are transported via gradients in the chemical potential, or concentration (diffusion), and electric potential (migration). In addition, the mechanical stress field in the fibres (stress driven diffusion) and pore pressure gradients in the SBE (convection) will influence the ion transport.

3.1.1 Volume changes

The redox reactions and flow of charged species cause the lithium concentration in the active electrode materials to change and generate or absorb heat (Figure 3.1a). The change in lithium concentration in the electrode material and the change in temperature will induce mechanical strains. To describe the relation between the physical quantities

related to these phenomena we utilize the following constitutive relations

$$\boldsymbol{\epsilon}^{\text{ch}}(c_{\text{Li}}) = \boldsymbol{\alpha}^{\text{ch}} [c_{\text{Li}} - c_{\text{Li},0}] \quad (3.1a)$$

$$\boldsymbol{\epsilon}^{\text{th}}(\theta) = \boldsymbol{\alpha}^{\text{th}} [\theta - \theta_0] \quad (3.1b)$$

In Eqs. (3.1a)-(3.1b), $\boldsymbol{\epsilon}^{\text{ch}}$ and $\boldsymbol{\epsilon}^{\text{th}}$ are the so-called insertion and thermal (free) strains, c_{Li} is the lithium concentration in the fibre and θ is the (absolute) temperature. The reference/initial values $c_{\text{Li},0}$ and θ_0 denote the state of a pristine material, i.e. the state at which no chemical or thermal strains are present in the material, respectively. Further, $\boldsymbol{\alpha}^{\text{ch}}$ and $\boldsymbol{\alpha}^{\text{th}}$ are second order tensors containing the coefficients of the lithium insertion and thermal induced expansion, respectively, and are defined as¹

$$\boldsymbol{\alpha}^{\text{ch}} = \alpha_{\perp}^{\text{ch}} [\mathbf{E}_1 + \mathbf{E}_2] + \alpha_{\parallel}^{\text{ch}} \mathbf{E}_3 \quad (3.2a)$$

$$\boldsymbol{\alpha}^{\text{th}} = \alpha_{\perp}^{\text{th}} [\mathbf{E}_1 + \mathbf{E}_2] + \alpha_{\parallel}^{\text{th}} \mathbf{E}_3 \quad (3.2b)$$

where $\alpha_{\perp}^{\text{ch}}$, $\alpha_{\perp}^{\text{th}}$ and $\alpha_{\parallel}^{\text{ch}}$, $\alpha_{\parallel}^{\text{th}}$ are the transverse and longitudinal expansions coefficients associated with lithium insertion and thermal expansion, respectively. It should be noted that Eqs. (3.2a)-(3.2b) are defined for a transversely isotropic material (as the carbon fibres). However, the same definition is valid for an isotropic material where $\alpha_{\perp} = \alpha_{\parallel}$.

Electrode materials in conventional Li-ion batteries, such as graphite, experience a volume expansion of approximately 10% with lithiation [66, 67], while electrode materials such as silicon and tin expands almost 300% [68]. With respect to carbon fibres, Jacques et al. [69] measured a reversible longitudinal fibre expansion in liquid electrolyte in the range of 0.8% and an area expansion of around 9% for a (dis)charge current of 37.2 A kg⁻¹ of fibre (equivalent C-rate ≈ 0.1). For comparison, Duan et al. [70] (Paper 6 in the list of publications not included in the thesis), recently reported a longitudinal fibre expansion of 0.6% and an area expansion of 13.6% measured with the same (dis)charge current. Further, in **Paper D** ([71]), we show that the longitudinal expansion of a carbon fibre-SBE electrode (i.e. fibres embedded in SBE) is approximately 0.2% lower compared with the "free" fibre expansion in liquid electrolyte. It should be noted that the free longitudinal expansion of the fibre is less than 1%. Such expansion is normally accepted for conventional polymers systems in carbon fibre reinforced polymers [72]. However, the cross-section area expansion is remarkable and will induce residual stresses during electro-chemical operation. Moreover, in case of large temperature variations, e.g. associated with the electro-chemical processes or external temperature variations, large thermal strains may be present inside the material. Hence, these volume changes must be considered in the design to avoid damage or performance degradation during operation.

3.1.2 Concentration dependent elastic moduli

The elastic properties of battery electrode materials, such as carbon fibres, graphite and positive electrode particles, are affected by the lithium concentration, [66, 67, 70, 73, 74]. For example, Jacques et al. [73] performed tensile tests on carbon fibre yarns and found that the stiffness of the fibres is unaffected by the lithium concentration (or

¹ $\mathbf{E}_i := \mathbf{e}_i \otimes \mathbf{e}_i$ is the i:th base dyad.

degree of lithiation). Moreover, Duan et al. [70] reported measurements for the lithium concentration dependent elastic moduli of carbon fibres. The transverse modulus of the fibres was found to increase more than 100% when fully lithiated. In contrast, its longitudinal modulus was slightly decreased. Hence, we may express the elasticity tensor, that is pertinent to transverse isotropy (i.e. defined by five independent parameters), as function of the lithium concentration $\mathbf{E}(c_{Li})$. Since measurements have shown that it is mainly the transverse elastic modulus of the carbon fibre that is significantly influenced by lithiation [70], Lamé's first parameter in the plane perpendicular to the fibre direction ($L_{\perp}(c_{Li})$) is considered to be the only elastic modulus that depends on the lithium concentration in **Paper E** (to simplify the problem).

3.1.3 Thermal effects

In addition to the coupling effects described above, the thermal processes will also be coupled with the mechanical and electro-chemical response. For example, heat will be generated (or absorbed) inside the battery cell during electro-chemical cycling, e.g. due to resistive heating (related to the flow of charged species) and redox reactions. This will alter the temperature dependent properties and induce thermal expansion/shrinkage of the constituents (cf. Eq. (3.1b)). Due to the fact that the reported transport properties of the constituents in the structural battery are inferior to those of the conventional batteries, these effects are expected to be more pronounced as compared to conventional batteries. For example, the electrical conductivity of carbon fibres [51] is about three orders of magnitude lower than that of copper, which is commonly used as current collectors in conventional graphite electrodes. Moreover, the ion conductivity of the SBE is in the order of 10^{-4} S cm $^{-1}$ [56, 57], whereas the conductivity of conventional liquid electrolytes is significantly higher, typically in the order of 10^{-2} S cm $^{-1}$ [75] at room temperature. Further, the electro-chemical performance of conventional batteries [76–78], as well as the mechanical properties of conventional carbon fibre reinforced polymers (as construction material), are known to be highly affected by temperature [79]. Hence, in combination with potential variation in external temperature, the temperature inside the material will vary during operation, which will affect the electro-chemical and mechanical processes. In **Paper E** we add the thermal energy balance as an additional governing equation and study the thermal effects on the combined performance.

3.2 Applied mechanical load (mechanical function)

Compared with conventional batteries, the active materials (in terms of the battery functionality) in the structural battery are intended to carry mechanical loads. Hence, the expected stress and strain level within the active materials will be larger as compared with a conventional battery. This means that coupling effects between the mechanical and electro-chemical response are expected to be more pronounced.

3.2.1 Coupling between chemical potential and mechanical stress

Electrode materials exhibit a voltage-strain coupling, sometimes referred to as the Piezo-Electro-Chemical-Transducer (PECT) effect, which can be seen from a change in the electric potential caused by a change in the stress state in the host material [44, 46, 80–83]. With respect to carbon fibres, experimental studies by Jacques et al. [44] and Harnden et al. [45, 46] (the latter being Paper 8 in the list of publications not included in the thesis) have demonstrated a noticeable PECT effect in carbon fibres exposed to tensile mechanical loads in the fibre direction. In [46], the PECT effect is demonstrated for bending of a cantilever beam consisting of two (partly lithiated) carbon-SBE electrodes sandwiched between a polymer separator. Moreover, an analytical closed form solution is derived² for the PECT effect and the predicted and measured coupling effect are found to agree remarkably well. This coupling effect is also predicted in **Paper B** where we demonstrate that the shift in the electric potential caused by an applied mechanical strain can be estimated from the coupling between the chemical potential of the lithiated fibres and the mechanical stress state. This particular coupling can be illustrated as follows.

The chemical potential of Li in the fibres can be expressed as

$$\mu_{\text{Li}} = \frac{\partial \psi}{\partial c_{\text{Li}}} = \mu_{\text{Li}}^{\text{ch}} + \mu_{\text{Li}}^{\text{me-th-ch}} \quad (3.3)$$

where ψ is the free energy. In Eq. (3.3), $\mu_{\text{Li}}^{\text{ch}}$ and $\mu_{\text{Li}}^{\text{me-th-ch}}$ refer to the stress-independent and stress-dependent part of the chemical potential, respectively, and are defined as follows

$$\mu_{\text{Li}}^{\text{ch}}(\boldsymbol{\epsilon}, c_{\text{Li}}, \theta) = \mu_{\text{Li}}^0 - R\theta_0 \log\left(\frac{\tilde{c}_{\text{Li},0}}{1 - \tilde{c}_{\text{Li},0}}\right) + R\theta \log\left(\frac{\tilde{c}_{\text{Li}}}{1 - \tilde{c}_{\text{Li}}}\right) \quad (3.4a)$$

$$\begin{aligned} \mu_{\text{Li}}^{\text{me-th-ch}}(\boldsymbol{\epsilon}, c_{\text{Li}}, \theta) &= -\rho^{-1} \boldsymbol{\alpha}^{\text{ch}} : \boldsymbol{\sigma}(\boldsymbol{\epsilon}, c_{\text{Li}}, \theta) \\ &+ \rho^{-1} \frac{1}{2} [\boldsymbol{\epsilon} - \boldsymbol{\epsilon}^{\text{ch}}(c_{\text{Li}}) - \boldsymbol{\epsilon}^{\text{th}}(\theta)] : \frac{\partial \mathbf{E}(c_{\text{Li}})}{\partial c_{\text{Li}}} : [\boldsymbol{\epsilon} - \boldsymbol{\epsilon}^{\text{ch}}(c_{\text{Li}}) - \boldsymbol{\epsilon}^{\text{th}}(\theta)] \end{aligned} \quad (3.4b)$$

Here, μ_{Li}^0 is a reference chemical potential and R is the universal gas constant. We have also introduced the normalized mass concentration of Li as $\tilde{c}_{\text{Li}} = \frac{c_{\text{Li}}}{c_{\text{Li,max}}}$, where $c_{\text{Li,max}}$ is the assumed maximum lithium concentration in the fibres. Further, $\boldsymbol{\sigma}$ is the (symmetric) stress tensor and ρ is the bulk density. It should be noted that the second term in Eq. (3.4b) is neglected in **Paper B-D** and **Paper F**, while it is included in **Paper E**.

Now, since the electric potential of the battery cell is linked to the chemical potentials of the active electrode materials³, it is possible to measure the change in electric potential caused by a change in mechanical stress state. Hence, the PECT effect is what enables the sensing mode for the composite laminate studied in **Paper F**.

Further, the assumed constitutive relation for the mass flux of the (neutralized) Li-ion in the fibre is defined as

$$\mathbf{j}_{\text{Li}} = -M_{\text{Li}} \cdot \nabla \mu_{\text{Li}} - FM_{\text{Li}} \cdot \nabla \varphi \quad (3.5)$$

²The derivation of the analytical closed form in [46] was made by the thesis author.

³More information on how the chemical potentials of the electrode materials are related to the electric potential of the battery cell is available in Supporting information to [46] (Paper 8 not included in thesis).

where \mathbf{M}_{Li} is the mobility tensor in terms of the Li-ions in the fibre, F is Faraday's constant and φ is the electric potential. It should be noted that in **Paper B-F**, we assume that the electric potential is uniform (and equal) in all fibres, $\varphi(\mathbf{x}, t) = \Phi^-(t)$, for $\mathbf{x} \in \Omega_f = \cup_i^{N_{\text{fibres}}} \Omega_{f,i}$; consequently, the electric field is zero within the fibre domain. From Eq. (3.5) it is clear that the ion flux is directly influenced by the stress state due to the fact that the diffusion driven contribution of the flux is associated with the gradient of the chemical potential.

3.2.2 Stress-driven convection

Due to the porous structure of the SBE (cf. Figure 2.1b), applied mechanical loads are expected to induce pore pressure gradients, which will drive a convective contribution to the mass transport of the liquid electrolyte (which facilitates the ion transport). In **Paper C** we add the governing equation for the flow of the liquid phase of the SBE, for which the pore pressure (denoted p) is added as an additional independent variable. This allows us to evaluate the effect of the stress-driven convection (seepage) of the liquid phase in the SBE on the electro-chemo-mechanical performance. For example, the constitutive relation for the fluid mass flux ($\hat{\mathbf{w}}$) is described by (Darcy's law)

$$\hat{\mathbf{w}} = -\mathbf{K} \cdot \nabla p \quad (3.6)$$

where \mathbf{K} is the permeability (hydraulic conductivity) tensor and p is the pore pressure. Hence, by accounting for the seepage we may define the stress ($\boldsymbol{\sigma}$) and ion flux of species $\alpha=\text{Li,X}$ (\mathbf{j}_α) as follows

$$\boldsymbol{\sigma} = \mathbf{E} : \boldsymbol{\epsilon}[\mathbf{u}] - \beta p \mathbf{I} \quad (3.7a)$$

$$\mathbf{j}_\alpha = -\mathbf{M}_\alpha \cdot \nabla \mu_\alpha - z_\alpha F \mathbf{M}_\alpha \cdot \nabla \varphi + c_\alpha \hat{\mathbf{w}} \quad (3.7b)$$

where Eq. (3.7a) expresses the so-called "effective stress" principle of porous media; $\boldsymbol{\sigma}' = \mathbf{E} : \boldsymbol{\epsilon}[\mathbf{u}]$ is the effective stress, where $\mathbf{E} = L\mathbf{I} \otimes \mathbf{I} + 2G\mathbf{I}^{\text{sym}}$ (expressed in terms of Lamé's parameters) is the standard isotropic elasticity tensor. Moreover, $\boldsymbol{\epsilon}[\mathbf{u}]$ is the (small) strain tensor expressed as a linear operator of the displacement field \mathbf{u} and β is the so-called Biot coefficient. Finally, z_α is the valence number (i.e. +1 for Li and -1 for X) and c_α is the concentration of species $\alpha = \text{Li,X}$. It should be noted that in comparison with the definition for the ion flux in Eq. (3.5), the convective contribution (related to the seepage) is added as an additional transport term in Eq. (3.7b).

From Eqs. (3.7a)-(3.7b) it is clear that the seepage is directly linked to the stress state (mechanical process) and the ion flux in the electrolyte (electro-chemical process). Hence, the processes are coupled.

3.3 Actuator and sensor functionalities

The actuator and sensor functionalities are schematically illustrated in Figure 3.2. For simplicity, we consider a cantilever beam structure with zero normal force. The "actuator mode" is shown in Figure 3.2a. This mode is enabled via the lithium insertion induced

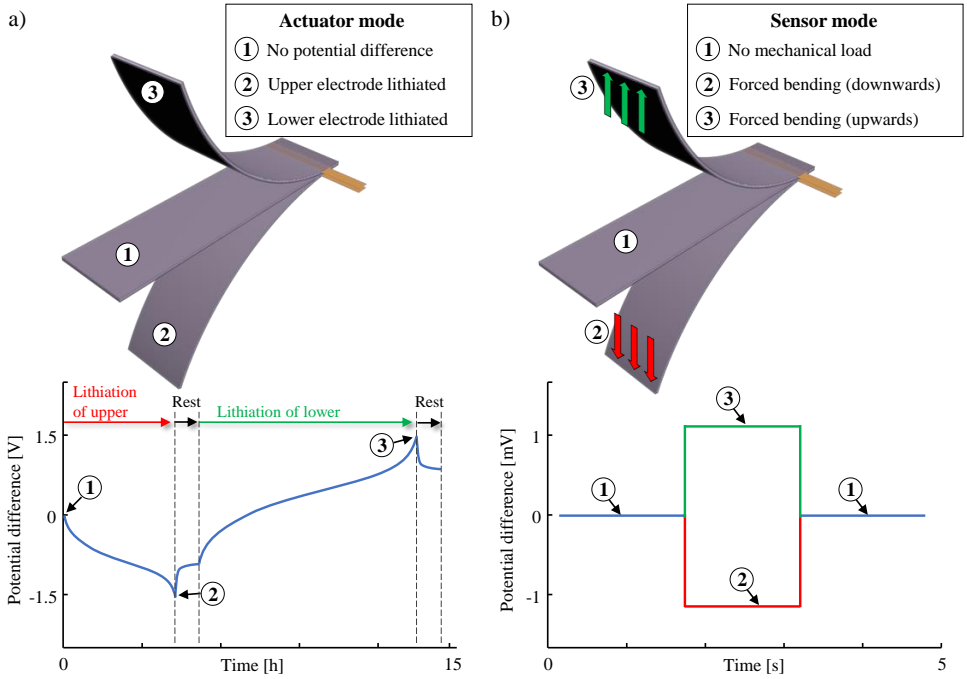


Figure 3.2: (a) "Actuator mode": Structural deflection. States: (1) No potential difference between electrodes; (2) Fibres in upper electrode lithiated (lower delithiated); (3) Fibres in lower electrode lithiated (upper delithiated) (b) "Sensor mode": Change in electric potentials. States: (1) No mechanical load; (2) Applied bending (downwards); (3) Applied bending (upwards). Figure from **Paper F**.

expansion of the carbon fibres (cf. Eq. (3.1a)), in particular the longitudinal expansion $\alpha_{\parallel}^{\text{ch}}$. For this mode three states are defined in the figure: (1) No potential difference between electrodes, i.e. the degree of lithiation, or fibre expansion, is the same in the two electrodes; (2) The fibres in upper electrode are lithiated, i.e. expanded, while the lower electrode is delithiated; (3) The fibres in lower electrode are lithiated while the fibres in the upper are delithiated. Hence, by moving the Li-ions from one electrode to the other (equivalent to charging or discharging a battery), the fibres expand in one layer while they contract in the other. This causes the potential difference between the electrodes to vary and the laminate to bend as illustrated in Figure 3.2a, i.e. enables the actuator functionality.

The "sensor mode" is shown in Figure 3.2b. The sensor functionality is enabled via the effect of mechanical stress on the chemical potential, in particular linked to the definition of $\mu_{\text{Li}}^{\text{me-th-ch}}$ (cf. Eq. (3.4b)). Hence, the strain sensing functionality is enabled through the possibility of measuring the change in electric potentials of the fibre electrodes and correlate that with the equivalent strain (or stress) state within the material, e.g. due to forced bending (Figure 3.2b). The three states shown in Figure 3.2b are: (1) No

mechanical load applied; (2) Forced bending (downwards); (3) Forced bending (upwards). It should be noted that in Figure 3.2b the potential difference between the electrodes is assumed to be zero (i.e. the electrodes have the same degree of lithiation) when no mechanical load is applied.

4 Computational modelling of structural batteries

4.1 Literature study

4.1.1 Conventional Li-ion batteries

Structural batteries have much in common with conventional Li-ion batteries. Among the wealth of literature related to modelling of batteries one should note important seminal contributions by Newman and co-workers [84–87]. This modelling framework (sometimes referred to as the *Newman model*) has been developed continuously over the years and provides today a foundation for much of the so-called physics based battery models.

In recent years there have been many studies aiming to assure that the modelling assumptions are consistent with the laws of mechanics and thermodynamics (which is not always guaranteed using the previously mentioned framework). In this context, there are numerous examples of relevant studies which have helped to advance the understanding and which are relevant also for structural batteries. Further, several review articles regarding electro-chemo-mechanical modelling of batteries are available in the literature, see e.g. [35, 88, 89]. Moreover, there are numerous examples of studies evaluating the electro-chemo-mechanical coupling effects in conventional batteries (utilizing liquid or solid-state electrolytes). For example, Grazioli et al. [90, 91] have developed a coupled numerical electro-chemo-mechanical model for solid polymer electrolytes and an analytical model for a uniform planar cell operating under galvanostatic conditions with and without externally induced deformations. Purkayastha and McMeeking [92] have developed an integrated 2D model to study the mechanical stress in storage particles as a function of material properties, and Ganser et al. [93] have developed a finite strain electro-chemo-mechanical theory for ion transport with application to binary solid electrolytes. Wan and Ciucci [94] have developed a general electro-chemo-mechanical model for studying the electro-chemical stability and mechanical stresses in solid-state-batteries, and Bucci et al. [95], have developed a continuum-level theory which couples the transport of multiple charged species with the deformation of a solid medium. In [95], the developed framework is used to e.g. simulate the behaviour of a thin-film with embedded continuous fibres (constrained by the underlying substrate). Bucci et al. [96], have also studied how the mechanical stress state affects the electro-chemical capacity of batteries. In particular, they show how the stress-induced voltage shift depends on the chemically induced expansion of the active material and on the constraint-induced stress.

In terms of dealing with the multiple length scales involved, Salvadori et al. [97–100] have developed multi-scale and computational homogenization approaches for modelling conventional Li-ion battery cells (utilizing liquid electrolyte). Moreover, Wu and Lu [101, 102], have developed a multi-scale mechanical-electro-chemical model which enables fully coupled mechanics and electro-chemistry at both particle and electrode levels. Further, Gupta and Gudmundson [103] have developed a multi-scale model for simulation of electro-chemically induced stresses on multiple scales (active particles, electrode layers,

and battery level) in conventional Li-ion batteries. In the work by Zhuo et al. [104] a nested finite element (FE2) multiscale framework is presented in which microscale simulations are employed to comprehensively characterize an anisotropic medium (macroscale). Finally, one should also note the variety of approaches for modelling the coupled thermo-electro-chemo-mechanical problem in conventional Li-ion batteries, see e.g. [105, 106].

Hence, one may favourably learn from the existing work and apply the same conceptual modelling approaches to structural batteries while keeping in mind that the main differences are: (i) Fibres (with anisotropic properties) are used as active electrode material in the negative electrode and as current collectors in both electrodes; (ii) A porous structural battery electrolyte (SBE) is used (instead of conventional liquid or solid-state electrolyte).

4.1.2 Fibre based structural batteries

In the context of fibre based structural batteries, Pupurs and Varna [107] studied the diffusion-induced stresses in carbon fibres due to lithium concentration gradients associated with electro-chemical cycling. Further, Xu et al. [108, 109] have studied how the internal stresses in structural 3D-batteries are affected by volume changes of the constituents. In [63] (Paper 2 in the list of publications not included in the thesis), the thesis author and co-authors study the effect of state of charge (i.e. degree of lithiation) on the elastic properties of structural 3D-batteries. Moreover, Dionisi et al. [110] and Johannisson et al. [43] considered a strongly simplified geometry and a simplified electro-chemical process in order to adopt an analytical and a numerical model for predicting deformations and stresses in laminated structural batteries and an electro-chemical actuator, respectively, due to cyclic volume change of the active materials. Further, Yin et al. [18] have studied (numerically and experimentally) carbon fibre electrode half-cells using conventional liquid electrolyte and modified carbon fibres. In [18, 108, 109, 111], the computational frameworks are based on the battery modelling scheme originally developed by Newman and co-workers [84–87] that presumes one-way coupling between the electro-chemical and mechanical processes.

With respect to the thermal interaction on the performance of structural batteries, only limited evaluation has been conducted. For example, Schutzeichel et al. [112, 113] studied, both experimentally and numerically, the thermal behaviour of multifunctional composite materials made from polymer electrolyte coated carbon fibres. Moreover, in [62] (Paper 3 in the list of publications not included in the thesis) a semi-analytical framework was developed to predict the mechanical consequences (thermal and diffusion induced stresses) from electro-chemical cycling. In this study, the electro-chemical and thermal analysis were strongly simplified, and the 3D-battery design was considered.

In the context of theoretical modelling of structural battery electrolytes, Tu et al. [42] have studied their bifunctional performance while assuming linear constitutive relations for stiffness and ionic conductivity. Moreover, Pejman et al. [114] have developed a design optimization tool for structural batteries. Further, Goudarzi et al. [115] recently presented an efficient computational approach for modelling and simulation of three-dimensional fibrous battery electrodes. Zhuo et al. [116] have also developed a computational model to evaluate the effective conductivity and capacity of the same type of fibre based battery electrodes. In these studies, the fibres in the battery electrodes are assumed to be randomly

orientated in three-dimensions.

In summary, despite the wealth of literature regarding computational modelling of batteries, there was a lack of a general computational framework for studying the fully coupled thermo-electro-chemo-mechanical response of carbon fibre based structural batteries (and electro-chemically driven actuators and sensors).

4.2 Computational modelling framework

The developed computational modelling framework is based on the governing balance equations associated with the respective physical domains and constitutive relations describing the the essential couplings (cf. Figure 1.3). The framework is thermodynamically consistent, i.e. the constitutive relations for (the energetic parts of) mechanical stresses, electro-chemical potentials and the electric flux density are derivable from a volume-specific free energy density for the bulk materials.¹ In other words, the formulation obeys the first and second law of thermodynamics (related to the energy and entropy of the system). Further, the highly anisotropic behaviour of the fibres in the longitudinal and radial directions (transverse isotropy) is considered.

The complete formulation of the galvanostatic/potentiostatic problem is defined in the context of the time-incremental weak format (based on the strong format). The system of equations is then solved using the commercial finite element (FE) software COMSOL Multiphysics[®].

4.2.1 Modelling assumptions

In terms of modelling assumptions, the model description is as earlier mentioned limited to idealized two-dimensional material representations. Further, we do not resolve the electric current within the fibre domains (Ω_f). The fibres (and the positive connector Γ_+ in **Paper B-E**) are instead assumed directly connected via an external circuit and the electric potential is assumed uniform (and equal) in all fibres, $\varphi(\mathbf{x}, t) = \Phi^{+/-}(t)$, for $\mathbf{x} \in \Omega_f = \cup_i^{N_{\text{fibres}}} \Omega_{f,i}$; consequently, the electric field is zero within the fibre domain.

The material properties are characterized as transversely isotropic (isotropy pertains to the cross-section, defined by Cartesian coordinates x_1, x_2 in Figure 4.1) in the fibres, and effectively isotropic at the studied micro-scale in the SBE (effective properties based on the two phases liquid/solid are used). In the fibres we assume that the single active species is Li, which moves into (and out from) the fibre. In the SBE on the other hand we assume that the ion current is carried both by Li^+ and the companion anion X^- , where Li-ions are positively charged (cation) and the companion X-ions (anion) are negatively charged.

4.2.2 Idealized material representations

In the papers attached to this thesis, a number of model geometries have been utilized (schematically illustrated in Figure 4.1). In Figure 4.1a, the definitions of the considered

¹The explicit expressions of the various contributions to the free energy are elaborated in Appendix A in **Paper E**.

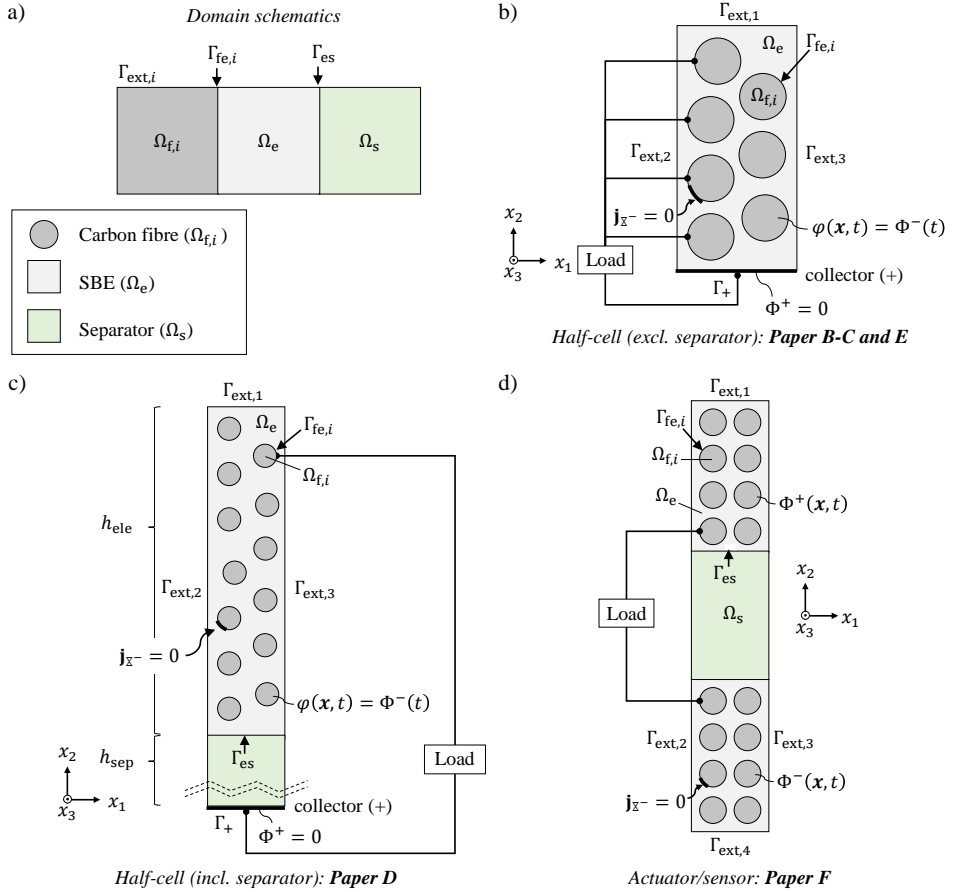


Figure 4.1: (a) Schematic illustration of domains. Idealized material representation of: (b) negative half-cell (studied in **Paper B-C** and **Paper E**), (c) negative half-cell including separator (**Paper D**), and (d) actuator/sensor (**Paper F**), respectively.

domains are presented. Further, in Figure 4.1b the idealized material representation utilized in **Paper B-C** and **Paper E** is shown.² As discussed earlier, we simplify the theoretical analysis by using the similarity of the two electrodes in the laminated architecture (cf. Figure 1.2b). Hence, this geometry is utilized to study the negative half-cell where the positive electrode in the full-cell is replaced by a collector of solid Li-metal (Figure 2.3b). Further, the separator is excluded (for simplicity). The generic/idealized model representation of this negative half-cell corresponds to a repeatable unit in the horizontal (x_1) direction of the negative electrode lamina (where the height of the unit corresponds to the height of the negative electrode). In Figure 4.1c, the model geometry

²It should be noted that there is a slight difference in appearance in the model geometry utilized in **Paper B**. However, the general concept is the same.

from **Paper D** is presented. In this paper the separator phase is added as an additional electrolyte phase. Finally, the model geometry used in **Paper F** is shown in Figure 4.1d. In this model, two carbon fibre-SBE electrodes, separated by a separator phase, are described. Hence, given the varying degree of lithiation in the two electrodes, the final model corresponds to a two (non-reference) electrode cell.

4.2.3 Time-continuous strong format

Governing equations

As starting point, the governing balance equations of momentum, charge (Gauss' law), mass and thermal energy may be expressed in the time-continuous strong format as follows:

$$-\boldsymbol{\sigma} \cdot \boldsymbol{\nabla} = \mathbf{0} \text{ in } \Omega \times \mathbb{R}^+ \quad (4.1a)$$

$$-SF[c_{\text{Li}} - c_{\text{X}}] + \mathbf{d} \cdot \boldsymbol{\nabla} = 0 \text{ in } \Omega \times \mathbb{R}^+ \quad (4.1b)$$

$$\partial_t(Sc_\alpha) + \mathbf{j}_\alpha \cdot \boldsymbol{\nabla} = 0 \text{ in } \Omega \times \mathbb{R}^+ \quad (4.1c)$$

$$\partial_t S + \hat{\mathbf{w}} \cdot \boldsymbol{\nabla} = 0 \text{ in } \Omega \times \mathbb{R}^+ \quad (4.1d)$$

$$C_v \partial_t \theta + \mathbf{q} \cdot \boldsymbol{\nabla} + \mathbf{j}_\alpha \cdot \boldsymbol{\nabla} \mu_\alpha + F[\mathbf{j}_{\text{Li}} - \mathbf{j}_{\text{X}}] \cdot \boldsymbol{\nabla} \varphi = Q_{\text{elec}} \text{ in } \Omega \times \mathbb{R}^+ \quad (4.1e)$$

where \mathbf{d} is the electric flux density vector (dielectric displacement) and \mathbf{q} is the heat flux vector. Further, S is the "fluid storage function" of liquid electrolyte salt, Q_{elec} is the generated heat due to the electric current in the fibres, and C_v is the volume-specific heat capacity. The independent variables for these equations are; \mathbf{u} (displacement field), φ (electric potential), c_α (concentrations), μ_α (chemical potentials), p (pore pressure) and θ (temperature).

Depending on the studied domain (e.g. fibre Ω_f and electrolyte domain Ω_e), different equations are relevant. For example, within the fibre domains we do not resolve the electron current, and assume that there is no seepage of liquid electrolyte or flow of X-ions. Hence, Eqs. (4.1b) and (4.1d) may be disregarded, Eqs. (4.1c) only contains the contribution for the Li-ions and S is replaced with the intrinsic density of the fibre ρ . Moreover, some essential couplings between the physical domains are visible from the governing equations. For example, terms associated with heat generated (or absorbed) due to electro-chemical processes are visible in the thermal energy balance (cf. Eq. (4.1e)).

Constitutive relations

The explicit expressions of the purely energetic quantities, e.g. $\boldsymbol{\sigma}$, \mathbf{d} , and μ_α , may be derived from the introduced partial free energy densities, see e.g. Appendix A in **Paper E** for more information. In addition to the energetic quantities, we utilize a series of (simple) constitutive relations for the dissipative variables (e.g. \mathbf{j}_α , \mathbf{q} , $\hat{\mathbf{w}}$). It should be noted that due to the scope of the thesis being "proof-of-concept", some of the modelling assumptions, e.g. linked to the electro-chemical processes, have been kept simple. However, the framework allows for more complex modelling assumptions in order to achieve better agreement between numerical predictions and experimental data (in future work).

From the constitutive relations, additional couplings between the physical processes become visible, as also discussed in Section 3. For example, the chemical potential of Li in the fibre (μ_{Li}) is affected by the mechanical stress state ($\boldsymbol{\sigma}$), Li-concentration (c_{Li}) and temperature (θ), cf. Eq. (3.4b). Moreover, the stress field is affected by the insertion and thermal induced expansions, cf. Eqs. (3.1a)-(3.1b). Hence, several essential couplings (cf. Figure 1.3) are described in terms of the assumed constitutive relations.

Finally, it should be noted that in **Paper C** ([117]) interpolation expressions are formulated to estimate the effective (microscopic) properties of the SBE as function of its porosity based on computational homogenization utilizing artificially generated Representative Volume Elements (RVEs). Similar interpolation expressions are utilized to estimate the effective properties of the separator phase, which is treated as an additional electrolyte domain, in **Paper D** and **Paper F**. For example, it is interesting to notice that the utilized interpolation rule for the ionic mobilities corresponds to the Bruggeman relation [84, 118] which is commonly used in battery modelling literature.

Interface and boundary conditions

Next, we define the constitutive relations for the interior boundaries, i.e. fibre-electrolyte interfaces Γ_{fe} and the electrode-separator interfaces Γ_{es} , see Figure 4.1. Moreover, relevant boundary conditions are defined in relation to the external boundaries, which comprises $\Gamma_{\text{ext}} = \sum_{i=1}^4 \cup \Gamma_{\text{ext},i}$. It should be noted that $\Gamma_{\text{ext},4}$ in **Paper B-E** is denoted Γ_+ (i.e. the boundary facing the "positive" Li-metal collector). With respect to the external boundary conditions, we apply conditions based on battery designs used in previous and concurrent experimental studies [12, 16, 46, 56, 71].

It should also be noted that although the FE-analysis is two-dimensional, the stress state is three-dimensional due to the assumed stress/strain condition in the x_3 -direction (along the fibres). In the attached papers we opt for conditions that simulate those which are typical for beam (and plate) kinematics, whereby the x_3 -direction is the beam axis. Hence, different mechanical conditions in the x_3 -direction are studied. For example, in the case of "generalized plane stress", we define the additional variable $\epsilon_{33}(x_1, x_2, t) = \bar{\epsilon}_{33}(t)$, and the condition

$$N_{33}(t) := \int_{\Omega} \sigma_{33}(\bullet, t) \, dS = 0 \quad (4.2)$$

where we note that Ω defines a surface in 2D. Here, Eq. (4.2) is the extra condition that is needed to compute $\bar{\epsilon}_{33}(t)$ as part of the FE-problem.

4.2.4 Time-incremental weak format

Based on the strong format of the governing equations, constitutive relations and boundary conditions, we derived the time-incremental weak format for the potentiostatic/galvanostatic problem. It should be noted that we employ a mixed method, where μ_α are treated as independent fields in addition to c_α . This choice requires additional constraint conditions (for more information see e.g. **Paper B**, [119]).

4.2.5 Numerical implementation

The numerical implementation is done in the commercial FE software COMSOL Multiphysics version 5.4. The time-incremental weak format of the governing equations are set up and solved in a monolithic fashion, i.e. without any staggering between the different physical mechanisms. The implementation procedure is schematically illustrated in Figure 4.2.

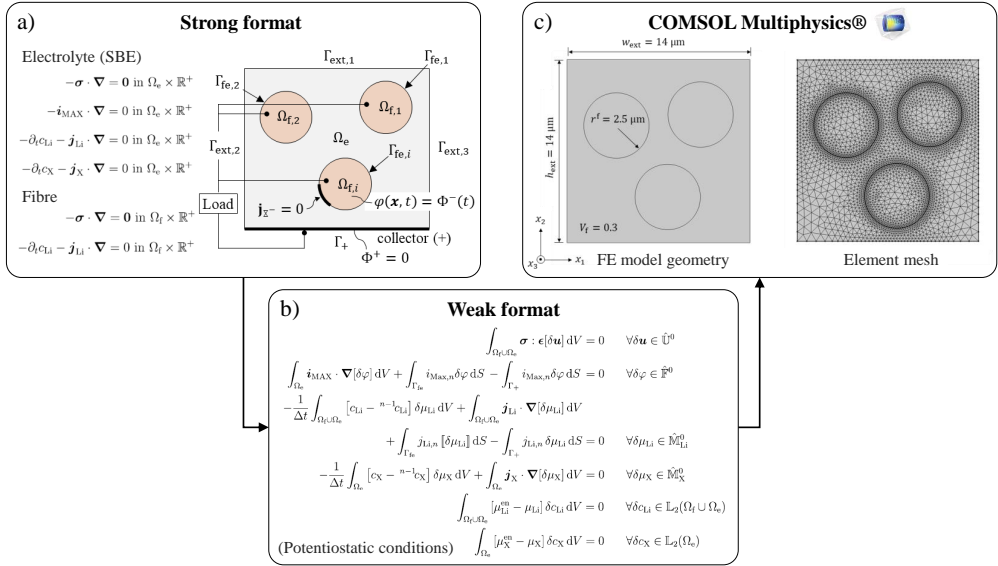


Figure 4.2: Schematic illustration of the implementation procedure. (a) Based on the governing equations, constitutive relations and boundary conditions (in strong format) we derive the (b) time-incremental weak format for the potentiostatic/galvanostatic problem, (c) which is set-up and solved in the commercial FE software COMSOL Multiphysics.

5 Summary of appended papers

Paper A : A structural battery and its multifunctional performance

In this paper, a structural battery composite with unprecedented multifunctional performance is demonstrated, featuring an energy density of 24 Wh kg^{-1} and an elastic modulus of 25 GPa and tensile strength exceeding 300 MPa. The structural battery is made from multifunctional constituents, where reinforcing carbon fibres act as electrode and current collector. A structural battery electrolyte (SBE) is used for load transfer and ion transport and a glass fibre fabric separates the carbon fibre electrode from an aluminum foil-supported lithium-iron-phosphate positive electrode.

Paper B : Electro-chemo-mechanically coupled computational modelling of structural batteries

In this paper, a (thermodynamically consistent) computational modelling framework is developed and exploited for the relevant electro-chemo-mechanical system. Effects of lithium insertion in the carbon fibres, leading to insertion strains, are considered while assuming transverse isotropy. Further, stress-assisted ionic transport is accounted for in addition to standard diffusion and migration. The relevant space-variational problems that result from time discretisation are established and evaluated in some detail. The proposed model framework is applied to a generic/idealized material representation to demonstrate its functionality and the importance of accounting for the electro-chemo-mechanical coupling effects. As a proof of concept, the numerical studies reveal that it is vital to account for two-way coupling in order to predict the multifunctional (i.e. combined electro-chemo-mechanical) performance of structural batteries.

Paper C : Computational modelling of structural batteries accounting for stress-assisted convection in the electrolyte

In this paper, the computational modelling framework developed in **Paper B** is extended to account for stress-assisted convection in the electrolyte. Focus is placed on how the convective contribution to the mass transport within the SBE affects the general electro-chemo-mechanical properties of structural batteries. The numerical results indicate that the convective contribution has only minor influence on the multifunctional performance when the mechanical loading is caused by constrained deformation of constituents during electro-chemical cycling. However, in the case of externally applied mechanical loading that causes severe deformation of the SBE, or when large current pulses are applied, the convective contribution has noticeable influence on the electro-chemical performance. In addition, it is shown that the porosity of the SBE, which affects the effective stiffness as well as the mobility and permeability, has significant influence on the combined mechanical and electro-chemical performance.

Paper D : Experimental and computational characterization of carbon fibre based structural battery electrodes

In this paper, electrodes consisting of carbon fibres embedded in structural battery electrolyte (CF-SBE electrodes) are characterized with respect to their multifunctional (i.e. combined electro-chemical and mechanical) performance. Experimental and numerical

studies are performed on CF-SBE electrode half-cells, whereby the computational modelling framework developed in **Paper B** is exploited. The results clearly show that the electrode thickness, transport properties of the SBE and applied current significantly affect the electro-chemical performance. For example, increasing the electrode thickness of the studied CF-SBE electrode from 50 μm to 200 μm results in a reduction in specific capacity of approximately 70/95% for an applied current of 30/120 A kg^{-1} of fibres, respectively. Moreover, we demonstrate that the developed computational framework is able to capture the overall behaviour of the battery cell (with respect to the electro-chemical analysis), and can predict mechanical strain and stress levels within the regime of small deformations and linear elastic material response. Further, lithium insertion induced longitudinal expansion of carbon fibre electrodes are measured during charge/discharge conditions. In liquid electrolyte the total/reversible longitudinal expansion are found to be 0.85/0.8% while for the CF-SBE electrode the reversible expansion is found to be 0.6%. The fibre expansion in the CF-SBE electrode give rise to residual strains, which is demonstrated numerically.

Paper E : On the coupled thermo-electro-chemo-mechanical performance of structural batteries with emphasis on the thermal effects

In this paper, we extend the computational modelling framework developed in **Paper B** to account for thermal interaction effects. The energy balance then becomes an additional governing equation, and the fully coupled thermo-electro-chemo-mechanical problem is solved for a simplified geometry with the appropriate constitutive relations and interface/boundary conditions. The numerical studies show that the developed framework is able to capture the coupled thermo-electro-chemo-mechanical behaviour. Moreover, it is found that the dominating source for heat generation during galvanostatic cycling is associated with discontinuities in the electrical and chemical potentials at the fibre/electrolyte interface. Further, a limited parameter study shows that the temperature change during electro-chemical cycling is significantly influenced by the applied current, thermal properties of the constituents and heat exchange with the surroundings. Finally, for large temperature variations, e.g. as identified during relevant (dis)charge conditions, the magnitude of the thermal strains in the structural battery electrolyte (SBE) are found to be similar to the insertion induced strains.

Paper F : Hierarchical modelling of a structural sensor-actuator comprising beam action and electro-chemical-mechanical interactions

In this paper, we extend and utilize the developed computational modelling framework (from **Paper B**) to simulate electro-chemically driven actuators and sensors utilizing carbon fibre-SBE electrodes. We focus on the actuation and sensing properties of a beam in conjunction with the appropriate "through-the-thickness" properties. The cross-sectional design is composed of two (partly) lithiated carbon fibre-SBE electrodes on either side of a separator. As a result, the modelling is hierarchical in the sense that (macroscale) beam action is combined with electro-chemo-mechanical interaction along the beam. The predicted deformation in case of the actuator-mode is found to be in close agreement with experimental data available in the literature in the case of small deflections. However for large deflections, geometric nonlinearities need be accounted for. Further, the sensor-mode is found to be well captured by the framework and the

voltage-strain coupling effect is shown to depend strongly on the assumed value of the lithium insertion induced expansion of the fibres (often referred to in terms of the partial molar volume).

6 Concluding remarks

Due to the inherent multifunctionality of structural batteries, the mechanical, electro-chemical and thermal performance need to be considered simultaneously in the material design. Hence, novel design frameworks and multiphysics models are needed to predict and evaluate its combined/coupled multifunctional performance.

In this thesis, a computational modelling framework to predict the coupled thermo-electro-chemo-mechanical performance of structural batteries is developed. In **Paper B** the general/original framework for the electro-chemo-mechanical problem (i.e. assuming isothermal conditions) is developed and described. In this paper, the importance of accounting for two-way coupling between the electro-chemical and mechanical processes is demonstrated. In **Paper C** the contribution to the ion transport from convection, i.e. from seepage of the liquid phase of the SBE, is accounted for. In this paper, we also show that in the case of externally applied mechanical loading that causes severe deformation of the SBE, or when large current pulses are applied, the convective contribution has noticeable influence on the electro-chemical performance. Further, in **Paper D** numerical predictions are compared favourably with experimental data and in **Paper E** the framework is extended to account for thermal interaction effects. In the latter, we show that for large temperature variations, e.g. as identified during relevant (dis)charge conditions, the magnitude of the thermal strains in the structural battery electrolyte (SBE) are similar to the insertion induced strains. Finally, in **Paper F**, the framework is extended to study electro-chemically driven actuators and sensors utilizing carbon fibre-SBE electrodes. In summary, the developed framework provides a foundation for computational modelling of structural battery composites. Due to the generality of the framework it allows for extensions and modifications. Moreover, the modelling framework allows for predicting the combined performance of structural batteries and can therefore be used for, e.g. tailoring the material design to achieve improved performance or to meet the requirements of the component. It also allows for studying essential couplings and the coupled processes during operation/service.

Moreover, in **Paper A** a laminated structural battery with unprecedented multifunctional (i.e. combined mechanical and electro-chemical) performance is demonstrated. The material design (schematically illustrated in Figure 2.3a) utilizes carbon fibres as combined electrode material, current collector and structural reinforcement. Further, a structural battery electrolyte (SBE) is used for load transfer between constituents and ion transport. The positive electrode is made from aluminum foil supported LFP while two types of glass fibre fabric, embedded in SBE, are used as separator. In this paper, we demonstrate a multifunctional composite material featuring an energy density of 24 Wh kg^{-1} and an elastic modulus of 25 GPa and tensile strength exceeding 300 MPa. This combined performance is far beyond any previous attempts (to the authors' knowledge) utilizing similar material architecture. Hence, in addition to provide an efficient material solution for future electric vehicle and devices, **Paper A** and **Paper D** provide inspiration and guidance for the development of future (further improved) laminated structural battery designs.

7 Future work

Four areas related to basic model extensions, multi-scale modelling, durability and material design options for improved multifunctional performance have been identified.

7.1 Basic model extensions

7.1.1 2D to 3D

To start with, the model descriptions in this thesis are limited to two dimensional material representations. By extending the theory and model description to three dimensions (3D), it is possible to e.g. resolve the distribution of electron current within and along the fibres. This would be particularly important when refining the model assumption related to the heat generation due to electron transport in the fibres (discussed in **Paper E**). Moreover, it would allow for estimating the variation of the utilization of the electrode materials, in particular associated with uneven ageing, which is known to be critical for the long term electro-chemical performance of e.g. graphite electrodes [120]. However, it should be noted that due to the involved length scales, strategies for dealing with the multiple scales (in an efficient manner) are needed.

7.1.2 Nonlinear and rate dependent (viscous) material properties

The theory in this thesis is limited to small strain kinematics. One can imagine that there will be cases when this assumption is violated. For example, at low current the cross-section area expansion of the fibres due to lithium insertion has been measured to be almost 14% [70]. Hence, in combination with external mechanical loading, the local strains are expected to be large as compared with the assumption of "small strains". Extending the theory to account for large deformation kinematics [93, 121] (also considering the porous structure of the SBE) is considered an important extension for future work.

In terms of material models, simplified constitutive relations have been utilized in the attached papers. This is linked to the scope of the thesis (proof-of-concept). To improve the agreement between numerical predictions and experimental data refined constitutive relations should be used. For example, with respect to mechanical properties, the material response of the constituents are assumed to be linear elastic. Polymers are known to exhibit a strongly pronounced rate dependent behaviour and significant inelastic deformation are often observed even at small deformations (visco-plastic behaviour). Moreover, in terms of the electro-chemical response refined expressions can be incorporated. For example, extended Butler-Volmer relations can be utilized to describe the current exchange at the electrode-electrolyte interfaces, and concentration dependent material properties can be assumed.

7.2 Multi-scale modelling

Multi-scale models can be envisioned to link the different length scales, e.g. following strategies available in the literature. In this context, Gupta and Gudmundson [103] recently presented a multi-scale model for simulation of electro-chemically induced stresses on scales of active particles, electrode layers, and battery level in conventional Li-ion batteries. Salvadori et al. [97–100] have also done extensive work on multi-scale and computational homogenization approaches for conventional Li-ion batteries. Further, a nested finite element (FE2) multiscale formulation, as proposed by Zhuo et al. [104], can also be envisioned/utilized to bridge the scales in future work.

7.2.1 Positive electrode

As discussed earlier, the coated carbon fibre positive electrode design (cf. Figure 1.2b) was not studied in this thesis due to its low maturity, uncertainty in design, lack of material data and its spatial scales (cf. [48, 49]). However, the developed framework can be used for modelling this type of electrodes in future studies. For this to be possible, the properties of the coating (e.g. particle size, distribution, etc.) need to be incorporated in the model. Further, depending on the particle size in comparison with dimensions of the fibres, the particles must be resolved or homogenized properties of the coating domain may be utilized.

7.3 Durability

Another import area for future research is the durability of the material. Due to its multifunctionality, multiple functions must operate safely during service. Hence, the durability must be assessed with respect to the combined functionalities. To date, some initial studies have been performed in this area. For example, Verners and Simone [122, 123] have performed reactive molecular dynamics simulations to study the mechanical response of material interfaces in fibre based structural batteries. Further, Xu and Varna [124] have studied the risk for propagation of radial matrix cracks and debond cracks at the coating/matrix interface in the 3D battery design, and Hu et al. [125] have studied the interfacial behaviour of modified carbon fibres in structural batteries. However, computational studies, which account for the coupled nature of the damage initiation and evolution process with respect to the multiple physical phenomena involved, are largely lacking. For example, electro-chemical cycling in combination with applied mechanical load will induce stresses and strains and may cause cracks and/or delamination between layers/laminae (Figure 7.1a), or along the fibre-SBE interface, in the fibre or along voids in the SBE (Figure 7.1b). Such failure phenomena are well known to have a pronounced effect on the electro-chemical performance of conventional batteries (see e.g. [126, 127]), and to the mechanical properties of fibre composites. Hence, to accurately model damage between the material interfaces (possibly undergoing large deformations), suitable constitutive relations accounting for the coupled processes need to be defined while assuring that the fundamental balance laws are fulfilled (cf. [128, 129]).

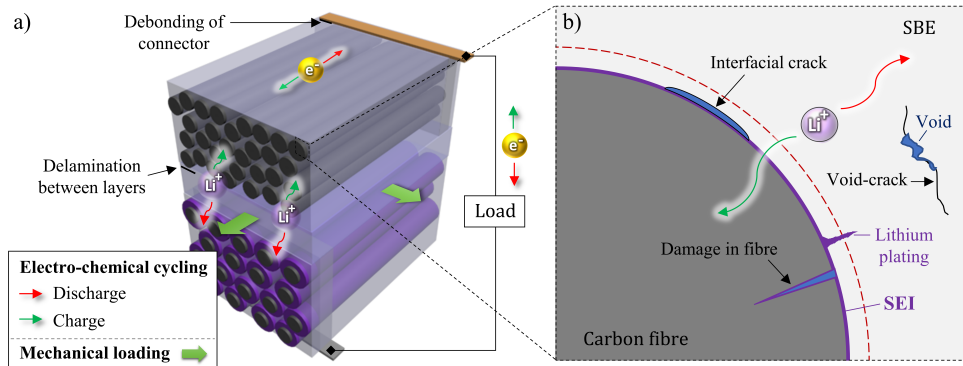


Figure 7.1: (a) Examples of possible damage mechanisms during operation. (b) Damage mechanisms at micro-scale. For example, during electrochemical cycling the fibres will expand and cracks may develop and propagate along the fibre-SBE interface, in the fibre or along voids in the SBE.

7.4 Material design for optimized electro-chemo-mechanical performance

Based on the results from the experimental studies in **Paper A** and **Paper D**, a number of areas have been identified in order to improve the multifunctional performance of future structural battery composites. To start with, the importance of having thin, porous, and stiff separators was confirmed in **Paper A** and **Paper D**. Moreover, the thickness and fibre volume fraction of the carbon fibre-SBE electrode were identified as critical design parameters for the performance. For example, the energy density of the structural battery in **Paper A** was highly affected by the added mass of the separator phase (glass fibre fabric embedded in SBE). This was due to the relatively thick separator solutions utilized, whereby the separator thickness was 125 and 185 μm . This can be compared with a conventional Li-ion battery separator, whose thickness is typically around 25 μm . Hence, a viable alternative is to use thinner (and highly porous) separators. Further, in **Paper D** we demonstrated that the electro-chemical performance of carbon fibre-SBE electrodes is highly influenced by the electrode thickness. In particular, for high currents the energy density of the material was significantly reduced as compared with a reference sample utilizing conventional liquid electrolyte. This reduction is assumed to be linked to the fact that the effective transport properties of the SBE are orders of magnitude lower as compared with conventional liquid electrolytes. Hence, the electro-chemical performance is highly affected by the properties of the SBE and significantly improved performance is expected when utilizing thin electrodes or improving the transport properties of the SBE.

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