

THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

On the multifunctional performance of structural batteries

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Cover:

Microscale mechanical model of a coated fibre using the commercial finite element (FE) software COMSOL.

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ABSTRACT

The structural battery composite is a composite material that can store electrical energy (i.e. work as a battery) while simultaneously provide mechanical integrity in a structural system. Due to its inherent ability to store energy this material offers significant weight savings on a system level. For this reason, this type of material has the potential to revolutionize future design of electric vehicles and devices.

Because of the multifunctional nature of this material novel design frameworks and multiphysics models are needed to predict and evaluate its multifunctional performance. Furthermore, charging and discharging the structural battery will generate heat and alter the volume and elastic properties of the constituents during operation. This will affect the effective properties of the material and generate internal stresses which can cause mechanical and/or electrical failure. For these reasons, it is crucial to be able to predict the multifunctional performance of the material and how it varies during operation.

In this thesis, modelling frameworks to predict and analyse the multifunctional performance of structural batteries is developed. The frameworks are used to estimate the multifunctional performance of different material designs and to study the mechanical consequences from electrochemical cycling. We demonstrate how the material design can be altered to enhance different performances. Furthermore, significant changes in effective elastic properties for the structural battery composite with change in state of charge (SOC) are found. This illustrates the need to consider changes in the elastic properties with SOC when designing structural battery components. Finally, it is shown that the properties of the constituents, charge/discharge current, lamina dimensions and residual stresses have significant effect on the internal stress state and the elastic properties of the composite lamina. The results also show that the heat generated during electrochemical cycling must be accounted for when evaluating the internal stress state in structural batteries.

Keywords: Carbon fibre reinforced plastics, Multifunctional composites, Energy storage, Mechanical properties, Electrical properties, Modelling

PREFACE

The work in this thesis was carried out from April 2017 to April 2019 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 under Grant Agreement Number 738085 and USAF, contract FA9550-17-1-0338.

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Göteborg, April 2019

David Carlstedt

THESIS

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

- Paper A** David Carlstedt, Wilhelm Johannisson, Dan Zenkert, Peter Linde, Leif E. Asp “Conceptual design framework for laminated structural battery composites” *Proceedings of 18th European Conference on Composite Materials (ECCM18)* (2018)
- Paper B** David Carlstedt, Erik Marklund, Leif E. Asp “Effects of state of charge on elastic properties of 3D structural battery composites” *Composite Science and Technology* (2019). DOI: 10.1016/j.compscitech.2018.10.033
- Paper C** David Carlstedt, Leif E. Asp “Thermal and diffusion induced stresses in a structural battery under galvanostatic cycling” *Submitted for publication*

Paper A: The approach originates from discussions among the authors. David Carlstedt and Leif Asp wrote the paper. Wilhelm Johannisson, Dan Zenkert and Peter Linde proof-read the paper and contributed with suggestions and comments.

Paper B: Leif Asp suggested the approach. Erik Marklund developed the original analytical CCA model. David Carlstedt developed the numerical implementation. David Carlstedt and Leif Asp developed the methodology and wrote the paper. Erik Marklund proof-read the paper and contributed with suggestions and comments.

Paper C: David Carlstedt suggested the approach and developed the numerical implementation. David Carlstedt and Leif Asp developed the methodology and wrote the paper.

Paper not included in thesis:

Wilhelm Johannisson, Niklas Ihrner, Dan Zenkert, Mats Johansson, David Carlstedt, Leif E. Asp, Fabian Sieland, Multifunctional performance of a carbon fiber UD lamina electrode for structural batteries *Composite Science and Technology* (2018). DOI: 10.1016/j.compscitech.2018.08.044

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Part I

Extended Summary

1 Introduction

1.1 Background

A route to high-performance lightweight electric vehicles and devices is smart designs, utilizing multifunctional materials such as structural battery composites. The structural battery composite is a composite material with ability to store electrical energy (i.e. work as a battery) while simultaneously provide mechanical integrity in a structural system. This material is made from carbon fibre reinforced polymer where the carbon fibres simultaneously work as mechanical reinforcements and active material in the battery cell. The high specific mechanical and electrical properties of carbon fibres [1, 2] make them ideal for multifunctional application. The structural battery composite is often referred to as “massless” energy storage due to its ability to store energy without adding additional weight solely for the purpose of storing energy. This type of material offers significant weight reduction on a system level [3, 4] and has the potential to revolutionize future design of electric vehicles and devices [5, 6, 7].

Airbus, who is a leading aircraft producer, aspires to make all-electric regional aircraft by 2050. An example of their aspiration can be seen from the announced partnership between Airbus, Rolls-Royce, and Siemens to develop the E-fan X shown in Figure 1.1. The development of this electric-hybrid aircraft demonstrator is a significant step forward in hybrid-electric propulsion for commercial aircraft.



Figure 1.1: The E-fan X hybrid-electric flight demonstrator. From [8].

Due to the low energy to weight ratio of existing monofunctional battery technologies, unrealistically high energy storage per passenger is needed to realise all electric regional aircraft. To enable this type of technology new energy storage solutions are needed. Structural battery composites provide a potential solution to this problem, providing

“massless” energy storage within the structural system. In addition to energy storage, the material provides the possibility to distribute the energy stored, reducing the need for cables, further lowering the weight of the aircraft. Consequently, this material can have significant impact on sustainable transportation meeting future requirements on reduced emissions.

The current research is part of a large research initiative on structural power composites initiated by Airbus GmbH to advance the maturity of this material. This research initiative is a continuation of earlier work done within the STORAGE [9] and KOMBATT [10] projects. The current thesis focus on analysis methods and evaluation of the multifunctional performance of structural battery composites. To evaluate the performance of this composite material, multiple physical phenomena need to be considered in concert. Hence, novel design frameworks and multiphysics models are needed to predict and evaluate the multifunctional performance of structural batteries.

1.2 Aim, scope and limitations

The main focus of this thesis is to develop models to predict and evaluate the multifunctional performance of structural battery composites. This thesis is part of the research project "Structural power for future civil aircraft" (SORCERER) which has the overall objective to advance structural power materials such that they can start to be adopted in Large Passenger Aircraft (LPA). In order to reach the targeted maturity of the technology models for evaluating the performance of the material are needed. Moreover, models are needed to predict the behaviour of the material under different loading conditions and to allow for improved material designs. The aims for this thesis are presented as follows.

Develop a model to predict the multifunctional performance of structural battery composites. The purpose of this tool is to be able to design an A4-sized demonstrator (as part of the SORCERER project) using laminated structural batteries such that requirements on mechanical and electrical performance are fulfilled. This tool is described in **Paper A** along with proposed designs for the demonstrator.

Evaluate mechanical consequences (i.e. changes in elastic properties and internal stress state) due to changes in the electrochemical state. Models for such purposes are developed and described in **Paper B** and **Paper C**.

2 Structural battery architectures

Two types of architectures for the structural battery composite have been proposed. The first one is the laminated battery architecture, which was first proposed by Wetzel et al. [11] and later demonstrated by Ekstedt et al. [12] and Carlson [13]. In this design the individual laminae have separate functions and work as electrode, separator, current collector, etc. The work by Johannisson et al. [14] on laminated structural battery negative half-cells illustrates the maturity of this concept and indicates highly promising performance of this material. The second architecture was first developed by Asp and co-workers [5, 13, 15] and is often referred to as the 3D battery or Micro battery architecture. In this design the individual carbon fibres are coated with a thin polymer electrolyte coating and work as individual battery cells in the composite lamina. State-of-the-art versions of the two battery architectures are illustrated in Figure 2.1.

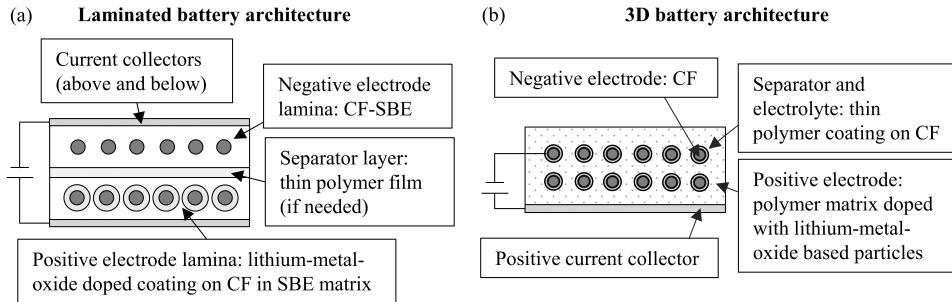


Figure 2.1: Schematic illustrations of the structural battery composite architectures (CF=carbon fibre, SBE=Solid battery electrolyte [16]). (a) The laminated battery. (b) The 3D battery.

Both architectures allow for the individual laminae to be stacked in different configurations, as done in ordinary composite laminates. This offers further tailoring of the design to provide improved multiaxial mechanical performance. As the laminae in Figure 2.1 are electrically conductive they need to be insulated from neighbouring plies. The structural battery composite is also sensitive to moisture (as ordinary lithium ion batteries) and must be protected from exposure to moist. Furthermore, the cells need to be connected to an external circuit via the current collectors and connection points. Some of these practical issues and additional discussions on the application of the structural battery composites in load carrying structures have been presented by Asp and Greenhalgh [17] and will be studied further in the continuation of the project.

In this thesis both architectures are studied. The design framework in **Paper A** is applied to laminated batteries but is also applicable for 3D batteries. Moreover, in **Paper B** and **Paper C** the 3D battery is studied but the developed frameworks are also applicable for laminated batteries.

2.1 Laminated battery

The upper lamina in Figure 2.1a corresponds to the negative electrode in the battery cell. This lamina consists of structural battery electrolyte (SBE) developed at KTH by Ihrner et al. [16] reinforced with carbon fibres (CF). The lower lamina, corresponding to the positive electrode, also consists of SBE but is instead reinforced with coated carbon fibres. This coating contains lithium metal oxide based particles (e.g. LiFePO_4). The carbon fibres in the negative electrode lamina and the particles in the coating in the positive electrode lamina are the active electrode materials in the battery cell. To avoid short-circuit, i.e. to assure that the positive and negative electrode do not come in contact with each other, a separator layer might be needed between the two laminae. The separator layer needs to be electrically insulating (i.e. not conduct electrons) while allowing transportation of ions between the electrodes. The separator layer also needs to be thin to minimize the ohmic loss, while rigid enough to prevent penetration and allow mechanical load transfer. The separator could potentially be removed given that the SBE is rigid enough and properly distributed to assure that the active electrode materials do not come in contact.

2.2 3D battery

In the 3D battery design concept (Figure 2.1b) each fibre works as an individual battery cell. The carbon fibres work as both negative electrodes in each battery cell and current collectors on the negative side. The individual fibres are coated with a thin solid polymer electrolyte which works as a combined electrolyte and separator layer. This polymer electrolyte coating is assumed to be made of the polymer system used by Leijonmarck et al. [18]. The surrounding polymer matrix is assumed to be made of a bi-continuous polymer network developed by Ihrner et al. [16]. The polymer matrix is doped with positive electrode particles (e.g. LiFePO_4) to make it work as positive electrode in the battery cell. The matrix is also doped with an electrically conducting material, e.g. carbon black (cb), to enable electrical conduction within the matrix. The benefit of using this design concept is the significantly reduced distance between electrodes [5].

3 Multifunctional performance

Due to the inherent multifunctionality of the material the mechanical and electrical performance need to be considered simultaneously in the material design. In the work by O'Brien et al. [19] a framework to estimate the multifunctional performance of structural capacitors was developed. This framework has been used by Greenhalgh et al. [20] to evaluate the multifunctional performance of structural supercapacitors. Building on these studies a framework to assess the multifunctional performance of structural batteries has been developed. In the framework in **Paper A** the mechanical performance is assessed by estimating the elastic properties while the electrical performance is assessed estimating the energy density (which refers to the amount of energy the battery can store per weight).

The mechanical performance of the composite lamina is assessed by estimating the in-plane elastic properties using Classical Laminate Theory (CLT). The i^{th} component of the extensional A_{ij} , coupling B_{ij} and bending D_{ij} stiffness matrices, respectively, are defined as

$$A_{ij} = \sum_k^n (\bar{Q}_{ij})_k (h_k - h_{k-1}), \quad (3.1)$$

$$B_{ij} = \frac{1}{2} \sum_k^n (\bar{Q}_{ij})_k (h_k^2 - h_{k-1}^2), \quad (3.2)$$

$$D_{ij} = \frac{1}{3} \sum_k^n (\bar{Q}_{ij})_k (h_k^3 - h_{k-1}^3), \quad (3.3)$$

where $(h_k - h_{k-1})$ represents the thickness of the k^{th} ply and n the number of plies. The i^{th} component of the stiffness matrix \bar{Q}_{ij} is defined with respect to the directional properties of the composite lamina based on the elastic properties of the constituents. These properties can be derived using micromechanical models or be based on experimental data. Hence, the elastic properties of the composite laminate depend on volume fractions, elastic properties of constituents, lamina thicknesses, orientations, etc. The electrical performance is accessed estimating the energy density defined as

$$\Gamma = V_n C_C, \quad (3.4)$$

where V_n is the nominal voltage during discharge and C_C is the specific capacity of the component (Ah/kg). The specific capacity is defined with respect to the amount of electrode materials available in the cell as

$$C_C = \min(C_{\text{neg}}, C_{\text{pos}})/w_C = \min(C_f m_f, C_p m_p)/w_C. \quad (3.5)$$

In Eq. (3.5) C_f and C_p are the specific capacity of the fibres (subscript f) and lithium metal oxide particles (subscript p), respectively. These capacities refers to the abilities of the active electrode materials to store electrical energy. The mass of the electrode materials (fibre and particles) m_f and m_p and the weight of the component (w_C) depend on the volume fractions, lamina thicknesses and material densities. Hence, the mechanical and electrical performance both depend on the laminate configuration and by varying volume fractions, lamina thicknesses, etc. the multifunctional performance of the material can be altered.

3.1 Demonstrator

In this work the developed framework has been used in the conceptual design of the A4-sized demonstrator. The proposed layout of the demonstrator is schematically illustrated in Figure 3.1.

The proposed design of the demonstrator consists of 10 laminated battery cells connected in series between protective reinforcement plies. The cells are connected in

series to provide an output voltage of 28 V, assuming that the nominal cell voltage during discharge for each cell is 2.8 V, to meet the power demand for the application. This assumption is based on the active materials considered in this work (carbon fibres versus LiFePO_4). By altering the laminate configuration different designs are achieved where the different performances are enhanced. The trade-off between electrical and mechanical performance for three different laminate configurations is illustrated in Figure 3.2.

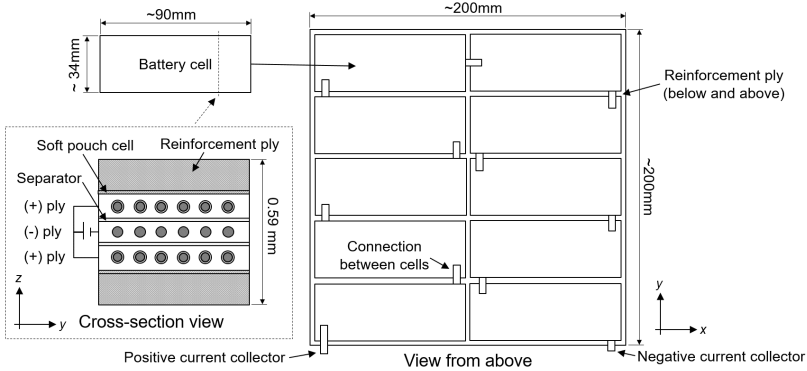


Figure 3.1: Conceptual design of the demonstrator for which the developed design framework (from **Paper A**) has been used to estimate its mechanical and electrical performance.

The three laminate configurations in Figure 3.2 have different volume fractions of constituents, lamina thicknesses, etc. This illustrates the trade-off between the electrical and mechanical performance for different material designs. Hence, for the given design of the demonstrator the energy density can be significantly improved by altering the material design but at the cost of reduced elastic properties.

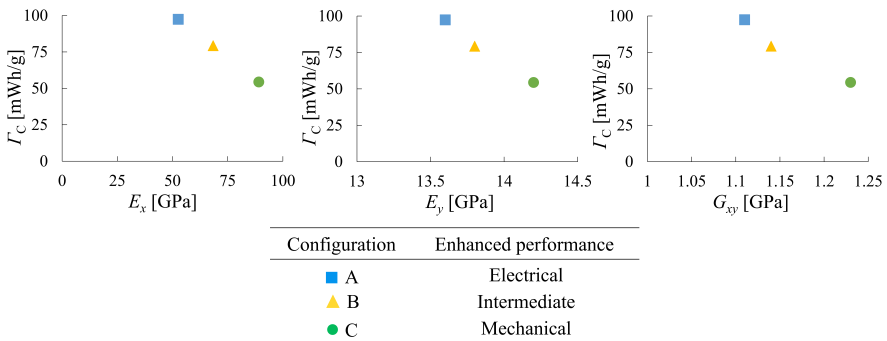


Figure 3.2: The energy density plotted against the elastic moduli for three laminate configurations. This illustrates the trade-off between the electrical and mechanical performance for different material designs.

4 Mechanical consequences from electrochemical cycling

During electrochemical cycling electrical energy is converted to chemical energy (or vice versa) via redox reactions. These redox reactions are reduction and oxidation processes that occur on the surfaces of the electrode materials during charge/discharge and causes change in the lithium concentration in the active electrode materials and generates or absorbs heat. The change in lithium concentration in the active electrode materials causes volume changes and alters the elastic properties of the materials. Earlier studies by Jacques et al. [21] show that intercalation of lithium ions into carbon fibres causes the fibre to expand approximately 1% in longitudinal direction and 8–13% in the transverse plane. It is also known that positive electrode materials particles expand/shrink due to lithiation/delithiation. For example, Mn_2O_4 particles expand/shrink approximately 7% due to lithiation/delithiation [22]. Moreover, it has been shown that the elastic properties of battery electrode materials such as e.g. carbon fibres, graphite and positive electrode particles are affected by lithium-ion concentrations [22, 23, 24, 25]. In addition to these changes, heat is generated inside the battery cell during electrochemical cycling due to resistive heating (related to the flow of charged species in the different phases). For these reasons, electrochemical cycling will generate heat, cause the electrode materials to expand or shrink and alter the elastic properties of the constituents. This will affect the effective properties of the material and induce internal stresses. Hence, to assess the mechanical performance of this material at different electrochemical states these effects need to be accounted for.

For ordinary composite materials the mechanical strains that produces stresses are defined as

$$\epsilon^{\text{M}} = \epsilon - \epsilon^{\text{T}} - \epsilon^{\text{H}}, \quad (4.1)$$

where ϵ are the applied elastic strains and ϵ^{T} and ϵ^{H} are the thermal and hygroscopic strains, respectively. In a structural battery composite no hygroscopic expansion will be present as the battery cell needs to be protected against moisture. For this reason, the mechanical strains defined for ordinary composite materials stated in Eq. (4.1) can be redefined for structural battery composites as

$$\epsilon^{\text{M}} = \epsilon - \epsilon^{\text{T}} - \epsilon^{\text{E}}, \quad (4.2)$$

where the hygroscopic strains ϵ^{H} are replaced with the electrochemical strains ϵ^{E} . These strains are linked to the normalized lithium concentration \bar{C} in the active electrode material defined as

$$\bar{C} = C/C_{\text{sat}}, \quad (4.3)$$

where C is the lithium concentration at a given time t and C_{sat} is the saturated lithium concentration (defined in mol/m^3). In Eq. (4.2) the thermal and electrochemical strains (ϵ^{T} and ϵ^{E}) are defined as

$$\epsilon^{\text{T}} = \alpha \Delta T, \quad (4.4)$$

$$\epsilon^{\text{E}} = \beta \Delta \bar{C}, \quad (4.5)$$

where ΔT and $\Delta \bar{C}$ are the change in temperature and change in normalized lithium concentration, respectively. The first order tensor containing the coefficients of thermal expansion is defined as $\boldsymbol{\alpha}$. The expansion coefficients due to change in lithium concentration ($\boldsymbol{\beta}$) are defined according to standard convention for hygroscopic expansion. This convention is used due to the resemblances in the physical phenomena and as no hygroscopic expansion will be present in this type of material.

The internal stress state can now be expressed as

$$\boldsymbol{\sigma} = \bar{\mathbf{Q}} : (\boldsymbol{\epsilon} - \boldsymbol{\epsilon}^T - \boldsymbol{\epsilon}^E). \quad (4.6)$$

In Eq. (4.6) the stiffness tensor $\bar{\mathbf{Q}}(\bar{C}, T)$ depends on the normalized lithium concentrations in the active electrode materials and the temperature. These dependencies are linked to the variation of the elastic properties and volume of the constituents with lithium concentration and temperature [16, 21, 22, 23, 24, 25, 26, 27].

In this work, only the mechanical consequences from electrochemical cycling under galvanostatic conditions (constant charge/discharge current) are studied and the analysis is assumed to be one-way. Hence, the developed models only studies how changes in volume and elastic properties of constituents and heat generation associated with electrochemical cycling affect the mechanical performance. The mechanical stresses are assumed not to affect the electrochemical capacity and flow of current within the cell. This assumption is motivated by observations made by Jacques et al. [28] who found the electrochemical capacity of T800 carbon fibres to be unaffected by mechanical loading. Moreover, mass transport limitations in the electrolyte are not accounted for (related to electrical performance). Under these conditions the electrochemical state can be approximated based on the diffusion of lithium in the active electrode materials and the flow of charged species within the battery cell.

The change in lithium concentration in the active electrode materials (fibres and particles) are governed by Fick's second law of diffusion which reads

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left(D \frac{\partial C}{\partial x} \right). \quad (4.7)$$

In this work it is assumed that the concentration gradient in the electrode particles is zero. Hence, the lithium concentration within the particles is assumed constant at each time step. Furthermore, it is assumed that the diffusion coefficient (D) is constant during electrochemical cycling. These assumptions are made to simplify the analytical expressions and to allow for superposition. Under the assumption that the diffusion coefficient of the fibre is invariant, Eq. (4.7) can be rewritten in cylindrical coordinates describing the lithium concentration in the fibres as

$$\frac{\partial C_f}{\partial t} = D_f \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_f}{\partial r} \right), \quad (4.8)$$

where D_f is the assumed diffusion coefficient of the fibre. As only galvanostatic cycling is considered the flux of lithium into the active material is assumed constant and determined based on the applied current.

The change in temperature due to heat generation during electrochemical cycling is derived as follows. The thermal energy balance is expressed as

$$mc_p \frac{\partial T}{\partial t} = Q_{\text{gen}} + q_{\text{ext}}, \quad (4.9)$$

where m and c_p are the mass and the specific heat for the system, respectively. Furthermore, Q_{gen} is the generated heat and q_{ext} is the heat exchange with the surroundings. The generated heat during electrochemical cycling is defined as

$$Q_{\text{gen}} = Q_{\text{ohm}} + Q_{\text{rev}} + Q_{\text{rxn}}, \quad (4.10)$$

where Q_{ohm} is the ohmic heat generation rate, Q_{rev} is the reversible heat generation rate and Q_{rxn} is the reaction heat generation rate. Under the assumption that Ohm's law is applicable, the ohmic heat generation rate in Eq. (4.10) can be defined with respect to the current I and internal resistance R as

$$Q_{\text{ohm}} = I^2 R, \quad (4.11)$$

where the internal resistance $R(T)$ depends on the temperature. This dependency is linked to temperature dependency of the conductivity of the different phases [16, 29] in which the current flows. In this work, the heat generation terms Q_{rev} and Q_{rxn} are neglected since they are assumed to be small in comparison with Q_{ohm} due to the relatively high electrical resistance of the constituents compared to counterparts in ordinary lithium ion batteries. Moreover, adiabatic conditions are assumed (i.e. no transfer of heat between the thermodynamic system and its surroundings $q_{\text{ext}} = 0$). This can be motivated by the fact that each battery cell will generate the same heat and given the low thermal conductivity of the surrounding polymer matrix only minor heat dissipation will occur for critical cells. Moreover, the individual laminae will be stacked in a laminate and need to be protected against moisture and oxygen. This means that heat transfer between the battery cells and the surrounding will be further limited in the final application. Given these assumptions Eq. (4.9) can be redefines as

$$\frac{\partial T}{\partial t} = \frac{I^2 R}{mc_p}. \quad (4.12)$$

The change in lithium concentration and temperature can now be derived by integrating Eqs. (4.8) and (4.12) over the time t as

$$\Delta \bar{C} = \frac{1}{C_{\text{sat}}} \int_0^t C(r, t) dt, \quad (4.13)$$

$$\Delta T = \int_0^t T(t) dt, \quad (4.14)$$

where $t \in [0; t_c]$ and t_c is the time for charge/discharge. Accounting for the change in volume and elastic properties of the constituents, associated with change in lithium

concentration and temperature, the effective properties of the composite and the internal stress state can now be estimated for different electrochemical states.

5 Modelling of structural batteries

In this thesis a set of modelling frameworks have been developed to study the mechanical consequences from electrochemical cycling. The modelling work in this thesis is based on earlier work on modelling of structural batteries. For example, Pupurs et al. [30] studied the diffusion-induced stresses in carbon fibres due to lithium concentration gradients associated with electrochemical cycling. Xu et al. [31, 32] have developed a numerical framework using a Multiphysics FE-model to study how the internal stresses in 3D structural batteries are affected by volume change of constituents. Moreover, Dionisi et al. [33] developed an analytical model to predict deformations and stresses in laminated structural batteries due to volume change of the active materials. The current frameworks build upon these studies and adds some of the missing pieces for predicting the mechanical consequences from electrochemical cycling. For example, in previous work on modelling of structural batteries the diffusion-induced stresses (i.e. stresses caused by volume change of electrode materials) have been studied without considering the thermal effects associated with electrochemical cycling. Moreover, how the elastic properties are affected by state of charge (SOC) has not been considered in previous work. The developed models used for these studies are presented in this chapter.

5.1 Semi-analytical models

Two types of semi-analytical modelling frameworks have been developed in this thesis; one to evaluate the elastic properties for different SOC (**Paper B**) and one for evaluating the internal stress state during electrochemical cycling (**Paper C**). These frameworks are illustrated in Figure 5.1 and 5.2, respectively.

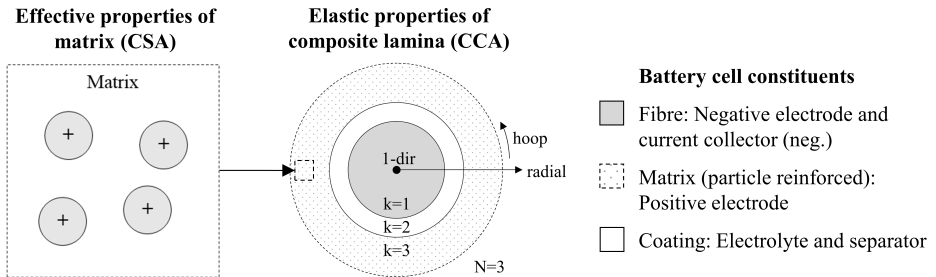


Figure 5.1: Illustration of the semi-analytical framework developed in **Paper B** used to evaluate the elastic properties of the composite lamina for different SOC. The CC model used in this context is referred to as a composite cylinder assemblage (CCA) model.

Both frameworks are based on concentric cylinder (CC) models [34, 35, 36, 37]. In this work the original CC models have been modified and extended to account for changes in volume and elastic properties of constituents and heat generation associated with electrochemical cycling of structural batteries. Moreover, the CC models are linked to a composite spheres assemblage (CSA) model [34] used to determine the effective properties of the particle reinforced matrix.

To account for changes in lithium concentration in the electrode materials and change in temperature associated with electrochemical cycling the CC model in **Paper B** is linked to an axisymmetric diffusion model and a one-dimensional thermal model (Figure 5.2). The diffusion model determines the lithium concentration in the fibres based on an analytical solution of Eq. 4.8 [38]. This model provides the normalized lithium concentration as input to the mechanical model. Moreover, the generated heat is estimated by the thermal model based on a lumped parameter model (often used for ordinary lithium ion batteries [39]). The total generated heat inside the battery is assumed to be a summation of the ohmic heat generated inside the individual material phases. To resolve the internal stress state and the lithium concentration distribution in the fibre the original material phases (fibre, coating and particle reinforced matrix) in the concentric cylinder model are split in additional cylinder sections as illustrated in 5.2.

The developed models are based on repeatable unit cells (RUC), which are often used to evaluate mechanical performance of materials by assessing the effective properties of a simple repeatable volume. The unit cell represents the most basic and smallest volume of a repeatable structure and the design of the unit cell depends on the requirements of the analysis. For mechanical analysis of composite materials RUC representing square and hexagonal packing arrangement or axisymmetric concentric cylinders are often used to evaluate effective elastic properties or stresses. In this work both types of models have been used. The semi-analytical frameworks are based on axisymmetric concentric cylinders while the FE-models used for validation are based on square and hexagonal packing arrangement as well as axisymmetric unit cells.

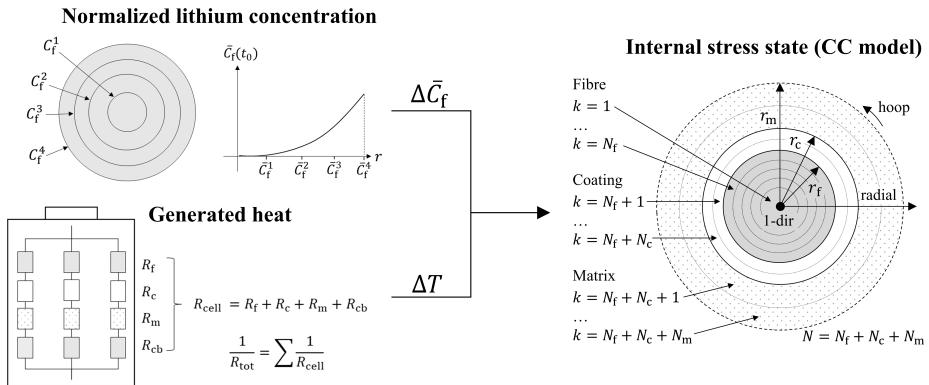


Figure 5.2: Illustration of the semi-analytical framework developed in **Paper C** used to study the internal stress state during electrochemical cycling.

5.2 FE-models

To validate the semi-analytical frameworks additional finite element (FE) models have been developed and set-up in the commercial FE software COMSOL 5.3a. To estimate the effective elastic properties of the composite material numerical homogenisation is performed in accordance to the procedure described by Barbero [40]. For this purpose, parametric FE models for square and hexagonal fibre packing arrangements have been analysed. These models are RUCs and are used to predict the effective elastic properties of the composite lamina for the two arrangements. These values represent upper and lower limits for the effective elastic properties of the composite and were used to validate the predictions from the analytical framework in **Paper B**. The FE models are presented in Figure 5.3a. The effective elastic properties of the composite lamina at different SOC is estimated by varying the volume fractions and elastic properties of the constituents.

The semi-analytical framework developed in **Paper C** to study the internal stress state during electrochemical cycling was validated using an axisymmetric FE model set up in COMSOL. This model is presented in Figure 5.3b. The displacements along the symmetry-axis ($r = 0$) in this model are set to zero and the external surface ($r = r_m$) is free to expand. Generalized plain strain is assumed (as assumed in the semi-analytical framework). The lithium concentration in the fibre is approximated using the axisymmetric diffusion model, which provides input on the resulting expansion and moduli of the fibre. The change in temperature inside the domain is derived using the 1D heat generation model.

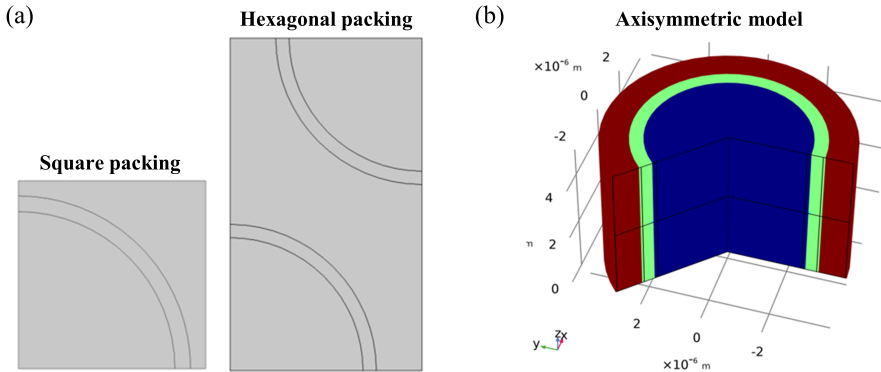


Figure 5.3: Computational models set-up in the commercial FE software COMSOL 5.3a used to validate the developed semi-analytical frameworks. (a) Parametric FE-models for square and hexagonal fibre packing, respectively, used for estimating effective elastic properties. (b) Two-dimensional axisymmetric model used to evaluate internal stresses.

6 Summary of appended papers

Paper A : Conceptual design framework for laminated structural battery composites

In this paper, a framework to estimate the mechanical and electrical performance of laminated structural battery composites is proposed. The proposed framework is used in the conceptual design of an A4-sized demonstrator. The mechanical and electrical performance are assessed estimating the in-plane elastic properties and the energy density of the structural battery composite component, respectively. Based on the trade-off between the mechanical and electrical performance three laminate configurations promoting different performances are proposed. These designs will be used in the build of the physical demonstrator and illustrates the possibility to alter the material design to provide suitable performance based on the purpose of the component.

Paper B : Effects of state of charge on elastic properties of 3D structural battery composites

The paper studies the effects of state of charge (SOC) on the elastic properties of 3D structural battery composites. An analytical model based on available micromechanical models is developed to estimate the effective elastic properties of 3D structural battery composite laminae at different SOC. A parametric study is performed to evaluate 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 composite lamina for extremes in SOC. Furthermore, parametric finite element (FE) models for square and hexagonal fibre packing arrangements are analysed in the commercial FE software COMSOL and used to validate the analytical model. The present study shows that the transverse elastic properties and in-plane shear modulus are strongly affected by the SOC while the longitudinal stiffness is not. For critical configurations the predicted change in transverse stiffness between extremes in SOC is as high as +43%. This shows that it is crucial to take effects of SOC on the elastic properties into account when designing 3D structural battery composite components.

Paper C : Thermal and diffusion induced stresses in a structural battery under galvanostatic cycling

In this paper, a semi-analytical framework to predict the thermal and diffusion induced stresses in a structural battery under galvanostatic cycling is presented. The proposed model is a concentric cylinder (CC) model coupled with an axisymmetric diffusion model and a one-dimensional heat generation model. The present study shows that the heat generated during electrochemical cycling must be accounted for when evaluating the internal stress state in structural battery composites. Furthermore, the results show that the charge/discharge current, lamina dimensions and residual stresses have significant effect on the internal stress state and effective properties of the composite lamina.

7 Concluding remarks and future work

Due to the inherent multifunctionality of structural batteries the mechanical and electrical performance need to be considered simultaneously in the material design. Moreover, electrochemical cycling will generate heat and alter the volume and elastic properties of the constituents during operation. For these reasons novel design frameworks and multiphysics models are needed to predict and evaluate the multifunctional performance of structural battery composites.

In this thesis, modelling frameworks to study the multifunctional performance of structural batteries have been developed. With the developed models it is possible to tailor the material design to meet the requirements of the component and to study the mechanical consequences from electrochemical cycling. In **Paper A** a design framework for estimating the multifunctional performance of structural batteries is proposed. In this work we illustrate how the material design can be altered to enhance different performances. Furthermore, in **Paper B** an analytical framework to study the effects of state of charge on the elastic properties of structural batteries is developed. In this work significant change in effective elastic properties for the structural battery composite with change in SOC is demonstrated. For example, an increase in the transverse stiffness of more than 40% is demonstrated for realistic structural battery composite designs as the battery is fully charged. This illustrates the need to consider change in the elastic properties with SOC when designing structural battery composite components. Finally, in **Paper C** a semi-analytical framework to study thermal and diffusion induced stresses in structural batteries under galvanostatic cycling (constant charge/discharge current) is proposed. This study shows that the heat generated during electrochemical cycling must be accounted for when evaluating the internal stress state in structural batteries. The results also show that the charge/discharge current, lamina dimensions and residual stresses have significant effect on the internal stress state and the elastic properties of the composite lamina.

By combining the developed models it is possible to predict the mechanical performance of structural batteries in the material design as well as during electrochemical cycling. The framework proposed in **Paper A** can be used to tailor the material design of the structural battery composite component. Furthermore, the change in the elastic properties of the composite component for different electrochemical states can be predicted using the framework presented in **Paper B**. Finally, the internal stresses developed during electrochemical cycling can be estimated using the proposed framework in **Paper C**. In addition, by combining the latter two frameworks the change in elastic properties, accounting for the generated heat during electrochemical cycling, can be predicted and evaluated. Hence, the combined set of models provides a toolbox for predicting the mechanical consequences from electrochemical cycling.

Based on the proposed designs of the demonstrator, the physical demonstrator will be built in further work. For the realisation of the physical demonstrator additional requirements on battery cell management, wiring, etc. need to be considered. Moreover, to fully capture the complete multifunctional characteristics of structural battery composites additional performance metrics such as strength (mechanical), power density (electrical), etc. will be vital to include in the design framework.

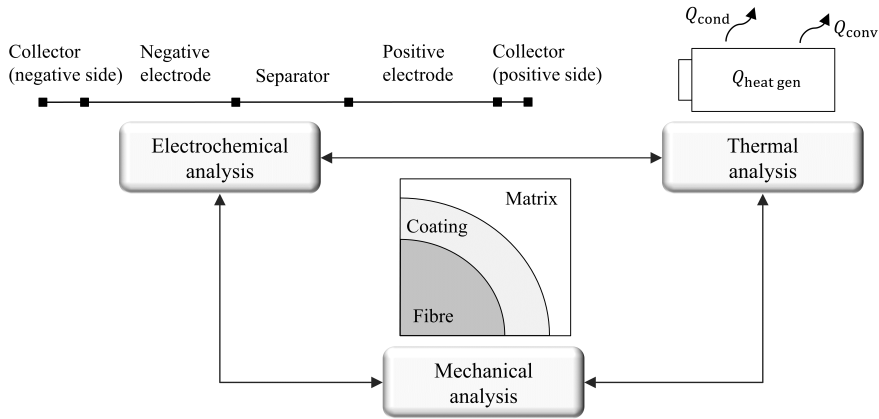


Figure 7.1: Schematic illustration of the coupled analysis in the multiphysics model for structural battery composites.

Moreover, to further advance the understanding of how the performances are affected by the interaction between the physical phenomena, two-way coupled mechanical-thermal-electrical models are needed. A schematic illustration of the coupled analysis in such a model is presented in Figure 7.1. In the current thesis one-way data exchange between the analysis is assumed. This simplifies the analysis significantly and allows for fast assessment of the mechanical consequences from electrochemical cycling. In further studies it would be of interest to allow for two-way data exchanges between the different physical models to be able to estimate the complete performance of the material with respect to the different physical phenomena. Furthermore, this type of model framework can be used to identify critical failure modes [41] to further advance the accuracy and completeness of the design framework for structural battery composites.

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