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Preprint

This is the submitted version of a paper published in *Composite structures*.

Citation for the original published paper (version of record):

Dionisi, F., Harnden, R., Zenkert, D. (2017)

A model to analyse deformations and stresses in structural batteries due to electrode expansions.

Composite structures, 179: 580-589

<https://doi.org/10.1016/j.compstruct.2017.07.029>

Access to the published version may require subscription.

N.B. When citing this work, cite the original published paper.

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<http://urn.kb.se/resolve?urn=urn:nbn:se:kth:diva-212208>

A model to analyse deformations and stresses in structural batteries due to electrode expansions

Filippo Dionisi^{a,b}, Ross Harnden^{a,*}, Dan Zenkert^a

^a*School of Engineering Sciences, Department of Aeronautical and Vehicle Engineering, Lightweight Structures, KTH Royal Institute of Technology, SE-100 44 Stockholm, Sweden*

^b*Dipartimento di Scienze e Tecnologie Aerospaziali, Politecnico di Milano, Via La Masa 34, 20156 Milano, Italy*

Abstract

In order to aid design of future structural battery components an analytical model is developed for modelling volume expansions in laminated structural batteries. Volume expansions are caused by lithium ion intercalation in carbon fibre electrodes. An extended version of Classical Lamination Plate Theory (CLPT) is used to allow analysis of unbalanced and unsymmetric lay-ups. The fibre intercalation expansions are treated analogously to a thermal problem, based on experimental data, with intercalation coefficients relating the fibre capacity linearly to its expansions. The model is validated using FEM and allows the study of the magnitude of interlaminar stresses and hence the risk of delamination damage due to the electrochemically induced expansions. It also enables global laminate deformations to be studied. This allows information about favourable lay-ups and fibre orientations that minimise deformations and the risk of delamination to be obtained. Favourable configurations for application to a solid state mechanical actuator are also given.

Keywords: structural batteries, electrode expansion, electrode deformation, analytical model, interlaminar stress, solid state mechanical actuator

*Corresponding author

Email address: harnden@kth.se (Ross Harnden)

1. Introduction

Structural batteries is a concept that aims to combine two functions into the same material: electrical energy storage and mechanical load bearing capability. These functionalities are not carried out by two separate components
5 in a system, but rather by the same material. The concept can be realised by using carbon fibres as an active electrode material in a lithium-ion battery (LIB) making it possible to create a synergetic multifunctionality that has the potential to offer large mass and volume savings at a systems level, and hence create more efficient structures [1].

10 Conventional LIB cells are typically made up of three layers: a cathode, an electrically insulating separator, and an anode, which are immersed in an ionically conductive electrolyte. In basic terms LIBs function by transferring lithium ions (Li-ions) between the anode and the cathode through the electrolyte, while corresponding electrons travel through an external circuit as electrical current.
15 Li-ions are most often inserted into the anode and cathode by a process of intercalation, whereby Li-ions correlate in the microstructure of the electrode material. The ions are extracted by the reverse process, known as deintercalation. During the charging process, Li-ions deintercalate out of the cathode, pass through the electrolyte, and intercalate into the anode.

20 PAN-based carbon fibres are used in a variety of high-performance structures thanks to their high specific stiffness and strength. Their use has enabled substantial weight savings in aircraft, cars, ships etc. PAN-based carbon fibres have also proven to intercalate Li-ions in a similar way to today's state-of-the-art LIB graphite-based anodes, and offer similar electrochemical characteristics [2].
25 These characteristics have paved the way for the development of multifunctional structural batteries.

Several architectures have been proposed for this concept, one of which is the laminated structural battery as schematically illustrated in figure 1. It has distinct similarities with a standard composite laminate. In this concept
30 a battery cell is made up of 3 layers: a carbon fibre anode, a load bearing

separator, and a cathode material which is reinforced with carbon fibres. These 3 layers are immersed in a solid battery electrolyte (SBE) material that acts as a matrix for the carbon fibre reinforcement. The cell can be considered as an unbalanced and unsymmetric laminate, depending on the anode and cathode fibre orientations.

Figure 1: Proposed laminated structural battery architecture

Promising work has recently been carried out developing SBE matrix materials that offer good ionic conductivity and mechanical properties [3]. Thus, for the anode side of the battery, PAN-based carbon fibres can be impregnated with a SBE matrix system creating a multifunctional ply. Likewise, a thin electrically insulating glass fibre weave could be used for the separator, impregnated with the same matrix system. The cathode side is a little more challenging and still requires more research. However, one possible path currently being researched is to mix some intercalating material, e.g. $LiFePO_4$, into the SBE used for the cathode, while using the carbon fibres as a reinforcement to carry the majority of the loads.

Structural batteries have not yet reached maturity enough for implementation and more research is needed — a comprehensive review of this subject area is carried out in [1]. One of the areas in need of more research is the modelling

of structural battery laminates.

50 It has been discovered that when carbon fibres are intercalated with Li-ions they expand longitudinally by up to 1%, and radially by as much as 5% [4]. This may pose practical problems for structural battery implementation, as such large strains would result in large deformations and interlaminar stresses at the component level, which could result in delamination. Conversely, it has
55 been proposed that such strains could be exploited in other applications such as solid state mechanical actuators [5]. However, due to the inherent anisotropy of long fibre laminated composite materials, and the ability therefore to tailor a laminate’s properties by varying fibre orientations and lay-up, it is proposed that such deformations and interlaminar stresses could be suppressed or exploited by
60 the way cells are put together. A model in which these effects could be analysed is therefore proposed.

The nature of the carbon fibre expansion due to Li-ion intercalation can be treated as linear with specific capacity following the experimental measurements by Jacques et al. [4] and shown in figure 2. In this work it was shown that
65 the carbon fibre expands almost linearly with the quantity of intercalated Li-ions. The quantity of Li-ions stored in the carbon fibre corresponds to its specific capacity. It should be noted that throughout the rest of this article *capacity* can be assumed to refer to the *specific capacity*, that is the capacity per unit mass of intercalating material. The reversible capacity is considered to
70 be the capacity after the first intercalation/deintercalation cycle, where there is an irreversible capacity loss due to Li-ions becoming trapped in the carbon fibre microstructure, as well as the formation of a thin coating known as the Solid Electrolyte Interphase. These trapped Li-ions are indicated by a small irreversible expansion after deintercalation, shown by the y-offset in figure 2.
75 The reversible capacity depends on the charge current, where a lower current will result in a higher reversible capacity. Following this an analogy to thermal expansion problems can be made, using an intercalation coefficient to linearly relate the fibre expansion to the reversible capacity.

The present study considers the need for an efficient computational method

80 which can give fast and sufficiently reliable results to allow valuable information about the effect of fibre orientation and lay-up on interlaminar stresses and laminate deformations. Given this premise, existing analytical methods have been extended in order to develop a computationally simple model for this specific case, and FEM has been used solely for validation purposes.

Figure 2: The longitudinal expansion strain in an IMS65 PAN-based carbon fibre as a function of capacity and charge current. Linearised and simplified reproduction from [4]

85 Interlaminar stresses exist near the free edges of laminated structures composed of dissimilar laminae. These are caused by the mismatch of material properties between adjacent layers in the case of traction-free edges where no external loads are applied. This problem can be an issue in the design of traditional composite laminates with free edges: the presence of these edge stresses
90 make delamination a common failure mode. Close to free edges in fact the state of stress becomes three dimensional and the predictions given by Classical Lamination Plate Theory (CLPT) become unsuitable. A comprehensive literature review on the research done on the analysis of free-edge effects in multilayered composite plates and shells can be found in [6].

95 Exact solutions for 3D elasticity problems have not yet been fully developed.
 Several authors have however proposed approximate solutions for finding shear
 and transverse interlaminar stresses for a range of load cases. Puppo and Even-
 son [7] and Pipes and Pagano [8] were among the first to study the free-edge
 effect of unidirectional extensional loading in laminated composites. Thereafter
 100 several other authors developed approximate solutions using various assump-
 tions. The study that has been considered as a base for the development of
 the method presented here is the work conducted by Kassapoglou and Lagace
 in [9], [10]. They focused on symmetric laminates under uniaxial loading and
 developed a method based on assumed stress shape functions which could also
 105 be implemented in other cases. In fact Kassapoglou generalised this method
 for various load cases [11] with the exception of the thermal one, although only
 symmetric laminates were considered. Following this work, Lin, Hsu and Ko
 [12] extended the method to unsymmetric cases albeit without consideration
 of thermal loading. Morton and Webber [13] conversely modified the method
 110 to account for thermal loading in the energy function, but concentrated exclu-
 sively on symmetric laminates. Lastly, Stiftinger [14] generalised Kassapoglou's
 method to take unsymmetric laminates into account. This last method has been
 modified in this article for application to structural battery laminates.

As structural battery technology develops, more emphasis will naturally be
 115 placed on the practical design of such structures. In order to begin under-
 standing how structural battery laminates will behave an analytical model is
 developed to obtain interlaminar shear and normal stresses as well as global
 laminate deformations. Input data is taken from Jacques et al. [4] for Toho
 Tenax IMS65 unsized carbon fibres and their intercalation expansions in order
 120 to estimate realistic cell stresses and deformations.

2. Model

2.1. Thermal expansion analogy

The expansion problem due to Li-ion intercalation in carbon fibres is treated analogously to a thermal problem for a laminate. This is justified by the fibres' behaviour during charging as shown in figure 2. The equivalent temperature change ΔT is replaced by the capacity C (quantity of intercalated Li-ions) of the fibres and the 3D thermal coefficients are now the ply intercalation coefficients, relating the capacity to the fibre expansions.

Hence it is possible to write, for both the longitudinal and transverse carbon fibre directions:

$$\varepsilon_{L,T}^f = \alpha_{L,T}^f (C_{rev} + C_{lost_{\varepsilon_{ir}}}) = \alpha_{L,T}^f C \quad (1)$$

where the subscript L,T stands for longitudinal and transverse direction, C_{rev} is the reversible capacity of the fibres and $C_{lost_{\varepsilon_{ir}}}$ is the irreversible capacity loss in the first cycle due to the irreversible expansion of the fibres, and $\alpha_{L,T}^f$ are the fibres' longitudinal and transverse intercalation coefficients. These parameters are functions of the charge current, as it can be interpreted from figure 2. The discrete experimental data from [4] have been interpolated to obtain a set of continuous data to be used in the model.

The smallest laminate unit is that of a three ply full battery cell as schematically illustrated in figure 3. It consists of a carbon fibre anode, a separator (assumed to be quasi-isotropic) and a carbon fibre reinforced cathode, all immersed in a SBE matrix.

The intercalation coefficients and the mechanical properties of the plies have been derived through rule of mixtures. The contraction of the cathode when deintercalating Li-ions has not yet been measured experimentally, however considering a cathode concept in which active material is mixed into the SBE as described above, a 2% isotropic contraction of the matrix is thought to be a reasonable assumption. This results in negative transverse and longitudinal intercalation coefficients — the latter being very small due to the stiffness of the

Figure 3: Schematic of basic structural battery unit consisting of a quasi-isotropic separator ply sandwiched between two UD carbon fibre plies constituting the anode and the cathode

carbon fibres. By treating the expansion as analogous to a thermal problem, it is possible to obtain the far-field stresses (away from the free edges) and the global laminate strains using CLPT. The former are used as inputs for the interlaminar stress calculations, while the latter are used for deriving the global laminate deformations.

2.2. Interlaminar stress model

2.2.1. General assumptions

The plies that constitute the final battery laminate are characterised by different intercalation coefficients, therefore if the plies are bonded together interlaminar stresses will be induced to force all plies to deform equally. Figure 3 shows the laminate global coordinate system (x_1, x_2, x_3) , on which the entire method is based. Considering the case shown in figure 3 stresses at the free edge normal to x_2 must be:

$$\sigma_{22} = 0 \quad \sigma_{12} = 0 \quad \sigma_{23} = 0 \quad (2)$$

A general stacked laminate consisting of N orthotropic plies is considered, as shown in figure 4.

Figure 4: General stacked laminate configuration for analytical model

The coordinate system is located in the middle of the laminate with the x_3 axis pointing upwards. The origin of the local coordinate for each ply is located at the centre point on the bottom surface of the respective ply, such that $0 \leq z \leq t^n$. For convenience a coordinate transformation is introduced:

$$y = b - x_2 \quad (3)$$

so that the origin of y is at the free edge.

The general assumptions made in the model can be summarised as follows [14]:

- Far from the free edge the solution from CLPT is recovered.
- The plies are treated as macroscopically homogeneous.
- The laminate is assumed to be long in the axial x_1 direction which allows the stress distribution to be independent of the x_1 coordinate. This means that the problem of expansion in 1- and 2-directions are separated.

Hence the equilibrium equations for each ply can be written as:

$$\frac{\partial \sigma_{12}^{(k)}}{\partial y} - \frac{\partial \sigma_{13}^{(k)}}{\partial z} = 0 \quad (4)$$

$$\frac{\partial \sigma_{22}^{(k)}}{\partial y} - \frac{\partial \sigma_{23}^{(k)}}{\partial z} = 0 \quad (5)$$

$$\frac{\partial \sigma_{23}^{(k)}}{\partial y} - \frac{\partial \sigma_{33}^{(k)}}{\partial z} = 0 \quad (6)$$

165 Note that no body forces are considered in this case.

2.2.2. Stress shape functions

It is assumed, as suggested in [9], that for each stress the y and z dependence can be functionally separated. Hence the stresses in the k^{th} lamina can be expressed as:

$$\sigma_{ij}^{(k)} = f_{ij}^{(k)}(y)g_{ij}^{(k)}(z) \quad (7)$$

where $f_{ij}^{(k)}(y)$ and $g_{ij}^{(k)}(z)$ are unknown functions to be determined for each ply in the laminate. Substituting equation 7 in equation 4, 5 and 6:

$$\frac{\partial f_{12}^{(k)}(y)}{\partial y} g_{12}^{(k)}(z) - \frac{\partial g_{13}^{(k)}(z)}{\partial z} f_{13}^{(k)}(y) = 0 \quad (8)$$

$$\frac{\partial f_{22}^{(k)}(y)}{\partial y} g_{22}^{(k)}(z) - \frac{\partial g_{23}^{(k)}(z)}{\partial z} f_{23}^{(k)}(y) = 0 \quad (9)$$

$$\frac{\partial f_{23}^{(k)}(y)}{\partial y} g_{23}^{(k)}(z) - \frac{\partial g_{33}^{(k)}(z)}{\partial z} f_{33}^{(k)}(y) = 0 \quad (10)$$

Considering that $f_{ij}^{(k)}(y)$ and $g_{ij}^{(k)}(z)$ have been assumed to be independent the equations above result in six equations for local equilibrium. These are given below:

$$\frac{\partial g_{13}^{(k)}(z)}{\partial z} - g_{12}^{(k)}(z) = 0 \quad (11)$$

$$\frac{\partial g_{23}^{(k)}(z)}{\partial z} - g_{22}^{(k)}(z) = 0 \quad \frac{\partial g_{33}^{(k)}(z)}{\partial z} - g_{23}^{(k)}(z) = 0 \quad (12)$$

$$\frac{\partial f_{12}^{(k)}(y)}{\partial y} - f_{13}^{(k)}(y) = 0 \quad (13)$$

$$\frac{\partial f_{22}^{(k)}(y)}{\partial y} - f_{23}^{(k)}(y) = 0 \quad \frac{\partial f_{23}^{(k)}(y)}{\partial y} - f_{33}^{(k)}(y) = 0 \quad (14)$$

This grouping of equations 11 to 14 shows that the required number of unknown functions to be assumed is four for each ply, since 12 and 14 are coupled. In order to recover the solution from CLPT, so that not only symmetric laminates can be analysed, $g_{12}^{(k)}$ and $g_{22}^{(k)}$ are assumed to vary as linear functions of z . These are given below:

$$g_{12}^{(k)} = B_1^{(k)}z + B_2^{(k)} \quad (15)$$

$$g_{22}^{(k)} = B_3^{(k)}z + B_4^{(k)} \quad (16)$$

Substituting 15 and 16 into 11 and 12 the following are obtained:

$$g_{13}^{(k)} = \frac{B_1^{(k)}}{2}z^2 + B_2^{(k)}z + B_5^{(k)} \quad (17)$$

$$g_{23}^{(k)} = \frac{B_3^{(k)}}{2}z^2 + B_4^{(k)}z + B_6^{(k)} \quad (18)$$

$$g_{33}^{(k)} = \frac{B_3^{(k)}}{6}z^3 + \frac{B_4^{(k)}}{2}z^2 + B_6^{(k)}z + B_7^{(k)} \quad (19)$$

Considering now the $f_{ij}^{(k)}(y)$ functions, as explained in [14], it must be pointed out that to satisfy the equilibrium conditions $\sigma_{33}^{(k)}$ must change sign at least once, so that the total force is zero. Moreover $\sigma_{33}^{(k)}$, $\sigma_{13}^{(k)}$ and $\sigma_{23}^{(k)}$ must decay to zero with increasing distance from the free edge. A linear combination of two exponential functions in y satisfies the requirement, although this does not guarantee additional sign changes that end up being the major restriction in the model. In fact FEM analysis shows that σ_{33} can have more than one sign change for some stacking sequences. Kassapoglou argues in [9] that a mode that traverses the y -axis intuitively represents a higher energy mode (analogous to plate vibration and buckling modes), thus the assumed form represents the lower energy state while still satisfying the requirements of equilibrium. It is then assumed that

$$f_{22}^{(k)}(y) = A_1^{(k)}e^{(-\phi y)} + A_2^{(k)}e^{(-\phi \lambda y)} + A_3^{(k)} \quad (20)$$

$$f_{12}^{(k)}(y) = A_5^{(k)} e^{(-\phi y)} + A_4^{(k)} \quad (21)$$

And hence by substituting 20 and 21 in 13 and 14

$$f_{13}^{(k)}(y) = -A_5^{(k)} \phi e^{(-\phi y)} \quad (22)$$

$$f_{23}^{(k)}(y) = -A_1^{(k)} \phi e^{(-\phi y)} - A_2^{(k)} \phi \lambda e^{(-\phi \lambda y)} \quad (23)$$

$$f_{33}^{(k)}(y) = A_1^{(k)} \phi^2 e^{(-\phi y)} + A_2^{(k)} \phi^2 \lambda^2 e^{(-\phi \lambda y)} \quad (24)$$

The interlaminar stresses in an unsymmetric laminate are thus fully defined by a set of constants, A_1 to A_5 and B_1 to B_7 . These constants can be determined by using boundary conditions and interface traction continuity, as well as two parameters which can be determined through the minimization of the complementary energy - described later.

2.2.3. Boundary conditions and interfacial traction continuity

The boundary and interface traction continuity conditions can be expressed as follows, as clearly exposed in [12]:

- At the free edge $y=0$, for every ply:

$$\sigma_{12}^{(k)} = 0 \quad \sigma_{22}^{(k)} = 0 \quad \sigma_{23}^{(k)} = 0 \quad (25)$$

While away from free edge the CLPT solution has to be recovered, hence:

$$\lim_{y \rightarrow \infty} \{\sigma_{13}^{(k)}, \sigma_{23}^{(k)}, \sigma_{33}^{(k)}\} = 0 \quad \lim_{y \rightarrow \infty} \sigma_{12}^{(k)} = \tilde{\sigma}_{12}^{(k)} \quad \lim_{y \rightarrow \infty} \sigma_{22}^{(k)} = \tilde{\sigma}_{22}^{(k)} \quad (26)$$

where the $\tilde{\sigma}$ refers to the results from CLPT. By using these boundary conditions it is possible to evaluate the $A_i^{(k)}$ coefficients:

$$A_1^{(k)} = -\lambda \quad A_2^{(k)} = -\frac{\lambda}{\lambda - 1} \quad A_3^{(k)} = A_4^{(k)} = -A_5^{(k)} = 1 \quad (27)$$

- On the bottom and top surfaces of the laminate the stresses have to be:

$$\sigma_{13}^{(r)} = 0 \quad \sigma_{23}^{(r)} = 0 \quad \sigma_{33}^{(r)} = 0 \quad r = 1, N \quad (28)$$

while at every interface between the plies they become:

$$\sigma_{13}^{(k)} = \sigma_{13}^{(k+1)} \quad \sigma_{23}^{(k)} = \sigma_{23}^{(k+1)} \quad \sigma_{33}^{(k)} = \sigma_{33}^{(k+1)} \quad k = 1, \dots, N-1 \quad (29)$$

Hence it is possible to evaluate

$$B_1^{(k)} = \frac{1}{t^{(k)}} (\tilde{\sigma}_{12_{top}}^{(k)} - \tilde{\sigma}_{12_{bottom}}^{(k)}) \quad (30)$$

$$B_2^{(k)} = \tilde{\sigma}_{12_{bottom}}^{(k)} \quad (31)$$

$$B_3^{(k)} = \frac{1}{t^{(k)}} (\tilde{\sigma}_{22_{top}}^{(k)} - \tilde{\sigma}_{22_{bottom}}^{(k)}) \quad (32)$$

$$B_4^{(k)} = \tilde{\sigma}_{22_{bottom}}^{(k)} \quad (33)$$

$$B_5^{(k)} = \sum_{j=1}^{k-1} B_1^{(j)} \frac{t^{(j)^2}}{2} + B_2^{(j)} t^{(j)} \quad (34)$$

$$B_6^{(k)} = \sum_{j=1}^{k-1} B_3^{(j)} \frac{t^{(j)^2}}{2} + B_4^{(j)} t^{(j)} \quad (35)$$

$$B_7^{(k)} = \sum_{j=1}^{k-1} B_3^{(j)} \frac{t^{(j)^3}}{6} + B_4^{(j)} \frac{t^{(j)^2}}{2} + t^{(j)} B_6^{(j)} \quad (36)$$

175 where t^j indicates the thickness of the j-ply. Note that for ply 1 the coefficients B_5 , B_6 and B_7 are equal to zero.

Finally different values for λ and ϕ could have been assumed for each ply, but the condition of traction continuity would have led to the result of λ and ϕ being constant throughout the laminate, as already assumed.

The final stress functions for the k^{th} ply can thus be written as follows:

$$\sigma_{12}^{(k)} = (1 - e^{-\phi y})(B_1^{(k)}z + B_2^{(k)}) \quad (37)$$

$$\sigma_{22}^{(k)} = [1 - \frac{\lambda}{\lambda - 1}(e^{-\phi y} - \frac{1}{\lambda}e^{-\lambda\phi y})](B_3^{(k)}z + B_4^{(k)}) \quad (38)$$

$$\sigma_{13}^{(k)} = \phi e^{-\phi y}(B_1^{(k)}\frac{z^2}{2} + B_2^{(k)}z + B_5^{(k)}) \quad (39)$$

$$\sigma_{23}^{(k)} = \phi \frac{\lambda}{\lambda - 1}(e^{-\phi y} - e^{-\lambda\phi y})(B_3^{(k)}\frac{z^2}{2} + B_4^{(k)}z + B_6^{(k)}) \quad (40)$$

$$\sigma_{33}^{(k)} = \phi^2 \frac{\lambda}{\lambda - 1}(\lambda e^{-\lambda\phi y} - e^{-\phi y})(B_3^{(k)}\frac{z^3}{6} + B_4^{(k)}\frac{z^2}{2} + B_6^{(k)}z + B_7^{(k)}) \quad (41)$$

$\sigma_{11}^{(k)}$, which has dropped out of the equilibrium equations, can be found, as in [15], through the 3D strain-stress relationship of an anisotropic laminate:

$$\begin{Bmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \varepsilon_{33} \\ \gamma_{23} \\ \gamma_{13} \\ \gamma_{12} \end{Bmatrix}^{(k)} = \begin{bmatrix} S_{11} & S_{12} & S_{13} & 0 & 0 & S_{16} \\ S_{12} & S_{22} & S_{23} & 0 & 0 & S_{26} \\ S_{13} & S_{23} & S_{33} & 0 & 0 & S_{36} \\ 0 & 0 & 0 & S_{44} & S_{45} & 0 \\ 0 & 0 & 0 & S_{45} & S_{55} & 0 \\ S_{16} & S_{26} & S_{36} & 0 & 0 & S_{66} \end{bmatrix}^{(k)} \begin{Bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33} \\ \sigma_{23} \\ \sigma_{13} \\ \sigma_{12} \end{Bmatrix}^{(k)} + \begin{Bmatrix} \epsilon_1 \\ \epsilon_2 \\ \epsilon_3 \\ 0 \\ 0 \\ \epsilon_{12} \end{Bmatrix}^{(k)} \quad (42)$$

where the ϵ_i terms are the free intercalation expansions due to charging with respect to the global coordinate system. Hence

$$\sigma_{11} = \frac{1}{S_{11}^{(k)}}(\varepsilon_{11} - \alpha_{11}C - S_{12}^{(k)}\sigma_{22}^{(k)} - S_{13}^{(k)}\sigma_{33}^{(k)} - S_{16}^{(k)}\sigma_{12}^{(k)}) \quad (43)$$

Assuming then that the interlaminar stresses do not affect the strain state of the laminate it follows that:

$$\varepsilon_{11}^{(k)} = \tilde{\varepsilon}_{11}^{(k)} \quad (44)$$

where

$$\tilde{\varepsilon}_{11}^{(k)} = S_{11}^{(k)} \tilde{\sigma}_{11}^{(k)} + S_{12}^{(k)} \tilde{\sigma}_{22}^{(k)} + S_{16}^{(k)} \tilde{\sigma}_{12}^{(k)} + \alpha_{11} C \quad (45)$$

Finally it can be stated that:

$$\sigma_{11}^{(k)} = \frac{1}{S_{11}^{(k)}} [S_{11}^{(k)} (B_8^{(k)} z + B_9^{(k)}) + S_{12}^{(k)} (B_3^{(k)} z + B_4^{(k)}) + S_{16}^{(k)} (B_1^{(k)} z + B_2^{(k)}) - (S_{12}^{(k)} \sigma_{22}^{(k)} + S_{13}^{(k)} \sigma_{33}^{(k)} + S_{16}^{(k)} \sigma_{12}^{(k)})] \quad (46)$$

where

$$B_8^{(k)} = \frac{1}{t^{(k)}} (\tilde{\sigma}_{11_{top}}^{(k)} - \tilde{\sigma}_{11_{bottom}}^{(k)}) \quad B_9^{(k)} = \tilde{\sigma}_{11_{bottom}}^{(k)} \quad (47)$$

2.2.5. Complementary energy

As reported in [13] the complementary energy for a thermo-elastic laminate, with null traction and body forces, can be written in general as:

$$\Pi = - \int_V G dV \quad (48)$$

With G representing the thermodynamic potential, or Gibbs function per unit volume, which in this problem can be written as

$$G = -\frac{1}{2} \sigma^t S \sigma - \sigma^t \alpha C \quad (49)$$

Recalling the assumption that the stresses are independent of the x coordinate, the energy can be evaluated per unit length. It is then assumed that the laminate is wide enough that:

$$e^{-\phi b} = 0 \quad e^{-\lambda \phi b} = 0 \quad (50)$$

Hence,

$$\Pi = \sum_{k=1}^N \left[\int_0^b \int_0^{t^{(k)}} \left(\frac{1}{2} \sigma^{(k)t} S^{(k)} \sigma^{(k)} + \sigma^{(k)t} \alpha^{(k)} C \right) dz dy \right] \quad (51)$$

where the energy is evaluated for each ply and then summed, while only half the width is considered due to symmetry. The expanded form of the energy has been obtained using the software Maple however is not reported here for conciseness. The derivatives of the energy are instead reported below.

2.3. Minimization of energy

The final result is the one that minimises the energy in the laminate. Hence by differentiating the energy equation with respect to the two unknown parameters it is possible to obtain two non-linear equations, shown below:

$$\begin{aligned}\frac{\partial \Pi}{\partial \lambda} &= \lambda^4 \phi^4 f_2 + 2\lambda^3 \phi^4 f_2 + \lambda^2 \phi^2 [f_3 - 2(f_6 + f_8 - f_9)] + \\ &\quad 2\lambda^2 (f_1 + f_7 + f_{10}) + 2\lambda [3f_1 + 2(2f_7 + f_{10})] + \\ &\quad 3f_1 + 4f_7 + 2f_{10} = 0\end{aligned}\tag{52}$$

$$\begin{aligned}\frac{\partial \Pi}{\partial \phi} &= 3\lambda^3 \phi^4 f_2 + \lambda^2 \phi^2 [f_3 + f_4 - 2(f_6 + f_8 - f_9)] + \lambda \phi^2 f_4 + \\ &\quad \lambda^2 [3f_1 + f_5 + 2f_{10} + 2f_{11} + 6f_7] + \\ &\quad \lambda [5f_1 + f_5 + 4f_{10} + 2f_{11} + 8f_7] + \\ &\quad 3f_1 + 4f_7 + 2f_{10} = 0\end{aligned}\tag{53}$$

The f -coefficients are expressed in Appendix A. The two equations are solved simultaneously with various initial values to obtain the solutions. The absolute minimum for the energy function is then established. Note that only real positive values of λ and ϕ are possible solutions, otherwise the expression for interlaminar stresses will grow rather than tend to zero away from the free edges.

2.4. Special cases

Special cases exist for laminate lay-ups as reported in [10], and these can lead to closed form solutions for λ and ϕ .

2.5. Angle ply laminates

In the case of symmetric angle ply laminates CLPT shows that

$$\sigma_{22}^{(k)} = 0\tag{54}$$

Consequently the set of stresses is given by:

$$\sigma_{12}^{(k)} = (1 - e^{-\phi y})(B_1^{(k)} z + B_2^{(k)})\tag{55}$$

$$\sigma_{13}^{(k)} = \phi e^{-\phi y} (B_1^{(k)} \frac{z^2}{2} + B_2^{(k)} z + B_5^{(k)}) \quad (56)$$

$$\sigma_{23}^{(k)} = 0 \quad (57)$$

$$\sigma_{33}^{(k)} = 0 \quad (58)$$

$$\sigma_{11}^{(k)} = \frac{1}{S_{11}^{(k)}} [S_{11}^{(k)} (B_8^{(k)} z + B_9^{(k)}) + S_{16} (B_1^{(k)} z + B_2^{(k)}) - S_{16}^{(k)} \sigma_{12}^{(k)}] \quad (59)$$

Hence λ drops away from the formulation of the energy and only the parameter ϕ needs to be taken into account (thus only the derivative $\frac{\partial \Pi}{\partial \phi}$) which leads to the solution of ϕ :

$$\phi = \sqrt{-\frac{f_5 + 2f_{11}}{f_4}} \quad (60)$$

If ϕ is imaginary in equation 60, no solution can be found and the method is not applicable.

2.6. Cross ply laminates

In the case of symmetric cross ply laminates CLPT shows that

$$\sigma_{12}^{(k)} = 0 \quad (61)$$

Thus the stresses become:

$$\sigma_{22}^{(k)} = [1 - \frac{\lambda}{\lambda - 1} (e^{-\phi y} - \frac{1}{\lambda} e^{-\lambda \phi y})] (B_3^{(k)} z + B_4^{(k)}) \quad (62)$$

$$\sigma_{13}^{(k)} = 0 \quad (63)$$

$$\sigma_{23}^{(k)} = \phi \frac{\lambda}{\lambda - 1} (e^{-\phi y} - e^{-\lambda \phi y}) (B_3^{(k)} \frac{z^2}{2} + B_4^{(k)} z + B_6^{(k)}) \quad (64)$$

$$\sigma_{33}^{(k)} = \phi^2 \frac{\lambda}{\lambda - 1} (\lambda e^{-\lambda \phi y} - e^{-\phi y}) (B_3^{(k)} \frac{z^3}{6} + B_4^{(k)} \frac{z^2}{2} + B_6^{(k)} z + B_7^{(k)}) \quad (65)$$

$$\sigma_{11}^{(k)} = \frac{1}{S_{11}^{(k)}} [S_{11}^{(k)}(B_8^{(k)}z + B_9^{(k)}) + S_{12}^{(k)}(B_3^{(k)}z + B_4^{(k)}) - (S_{12}^{(k)}\sigma_{22}^{(k)} + S_{13}^{(k)}\sigma_{33}^{(k)})] \quad (66)$$

Recalculating the expression for the energy and the derivatives 52 and 53:

$$\lambda(\lambda - 1)(\lambda^2\phi^4f_2 - f_1) = 0 \quad (67)$$

The possible solutions are then:

200

- $\lambda = 0$, not of interest since it results in the CLPT solution

- $\phi = \sqrt{\frac{1}{\lambda} \sqrt{\frac{f_1}{f_2}}}$

This could result in a non-real solution, in which case the next possibility should be applied.

- $\lambda = 1$.

In this case, the two basic stress shapes $e^{-\phi y}$ and $e^{-\phi \lambda y}$ coincide, hence as suggested in [10] and [13], a new initial shape for σ_{22} is proposed. In particular if a characteristic equation shows repeated roots, the exponential solutions are $e^{-\phi y}$ and $ye^{-\phi y}$. Thus f_{22} becomes:

$$f_{22} = A_1 ye^{-\phi y} + A_2 e^{-\phi y} + A_3 \quad (68)$$

and again applying the boundary conditions as done above

$$A_1 = -\phi \quad A_2 = -1 \quad A_3 = 1 \quad (69)$$

Hence, the new stress shapes can be written as

$$\sigma_{22}^{(k)} = [1 - (1 + \phi y)e^{-\phi y}](B_3^{(k)}z + B_4^{(k)}) \quad (70)$$

$$\sigma_{23}^{(k)} = \phi^2 ye^{-\phi y} (B_3^{(k)} \frac{z^2}{2} + B_4^{(k)} z + B_6^{(k)}) \quad (71)$$

$$\sigma_{33}^{(k)} = \phi^2 (1 - \phi y) e^{-\phi y} (B_3^{(k)} \frac{z^3}{6} + B_4^{(k)} \frac{z^2}{2} + B_6^{(k)} z + B_7^{(k)}) \quad (72)$$

And ϕ can be found in the closed form:

$$\phi = \sqrt{\frac{-(f_3 - 2f_6) + \sqrt{(f_3 - 2f_6)^2 - 12f_2[11f_1 + 8f_{10}]}}{6f_2}} \quad (73)$$

Delamination of composite laminates results from the out-of-plane stresses σ_{13} , σ_{23} and σ_{33} . Brewer and Lagace proposed a Quadratic Delamination Criterion [16] to predict the risk of delamination based on admissible shear and transverse strength. First an average stress is defined as suggested by Kim and Soni [17] as:

$$\bar{\sigma}_{ij} = \frac{1}{h_0} \int_0^{h_0} \sigma_{ij} dy \quad (74)$$

where σ_{ij} is the interlaminar stress of the analysed interface and h_0 is the so called critical length. The latter is assumed to be the average of the thicknesses of the two adjacent plies considered [14], with the exception of the case of $\bar{\sigma}_{33}$ where it is assumed to be the distance between the free edge and the point where σ_{33} changes sign [16]. The criterion can then be expressed as:

$$f_{risk} = \left(\frac{\bar{\sigma}_{13}}{Z_{13}}\right)^2 + \left(\frac{\bar{\sigma}_{23}}{Z_{23}}\right)^2 + \left(\frac{\bar{\sigma}_{33}^t}{Z_{33}^t}\right)^2 < 1 \quad (75)$$

where Z_{ij} is the interlaminar strength in the corresponding direction and f_{risk} is the delamination risk coefficient. Note that as suggested in [14] only positive σ_{33} is considered (the superscript t designates traction) as the compressive terms do not influence the delamination risk.

The average stresses can be derived directly by integrating equations 39, 40 and 41 or equivalents for the special cases. For instance in the general case:

$$\bar{\sigma}_{13}^{(k)} = \frac{1 - e^{-\phi h_0}}{h_0} (B_1^{(k)} \frac{z^2}{2} + B_2^{(k)} z + B_5^{(k)}) \quad (76)$$

$$\bar{\sigma}_{23}^{(k)} = -\frac{\lambda e^{-\phi h_0} - e^{-\lambda \phi h_0} - \lambda + 1}{h_0(\lambda - 1)} (B_3^{(k)} \frac{z^2}{2} + B_4^{(k)} z + B_6^{(k)}) \quad (77)$$

$$\bar{\sigma}_{33}^{(k)} = \frac{\lambda \phi (e^{-\phi h_0} - e^{-\lambda \phi h_0})}{h_0(\lambda - 1)} (B_3^{(k)} \frac{z^3}{6} + B_4^{(k)} \frac{z^2}{2} + B_6^{(k)} z + B_7^{(k)}) \quad (78)$$

210 This criterion requires the determination of appropriate strength parameters. Some examples of experimental derivations of these parameters can be found in [18] and [19].

2.8. Limitations

Despite the solutions that have been obtained for the laminates studied,
215 there are cases where no real solutions exist for the coefficients λ and ϕ . Certain
combinations of *thermal* and mechanical properties and loads exist where the
method cannot return solutions, for instance when the coefficients are not real.
Webber and Morton gave the algebraic conditions for the existence of solutions
for simple cases such as cross ply laminates, as reported in Appendix B of
220 [13]. The complexity of the equations for the general method however make
it extremely difficult to predict which conditions result in unsolvable problems.
Only a few combinations have been found where the method presented a so
called *blind spot* in the parameter space [13].

Although the coefficients presented here are limited to the intercalation load
225 case, the response to mechanical load could also be implemented by adding the
specific terms to the energy function in equation 51.

2.9. Validation

The method has been validated against existing data from literature and a
FEM analysis using *Abaqus*. In particular the pure thermal cases reported in
230 [13], [14] and [20] with specific ply properties have been simulated using the
presented model and all cases show good agreement. A 3D laminate has been
built in *Abaqus* to simulate the expansion of the various plies of the battery
laminate. The plies that have been considered here are a carbon fibre anode, a
glass fibre separator and a carbon fibre cathode. 3D elements have been used
235 for obtaining the normal stress σ_{33} , in particular 20-node quadratic bricks with
reduced integration (C3D20R), as recommended in [21]. For obtaining shear
stresses, continuum shell elements have been used, as suggested in [22]. Mesh
bias toward the free edge has been implemented and a predefined temperature
field as been input with the value of $\Delta T = C$. Finally the stresses have been
240 plotted following a path in the considered interface. The data used in the FEM
and in the analytical model are reported in table 1. These are specific to a charge
current of $15[\frac{mA}{g}]$ which corresponds to a cell first cycle capacity of $345[\frac{mAh}{g}]$.

Table 1: Ply data

	<i>Anode</i>	<i>Separator</i>	<i>Cathode</i>
t [mm]	0.5	0.3	1.0
V_f	0.6	0.5	0.6
E_L [MPa]	175200	13500	175200
E_T [MPa]	5386	13500	5386
G_{LT} [MPa]	2702	3945	2702
ν_{LT}	0.3	0.25	0.3
ν_{T3}	0.3	0.25	0.3
α_L [$\frac{g}{mAh}$]	2.18e-5	0	-1.48e-7
α_T [$\frac{g}{mAh}$]	6.08e-5	0	-1.12e-5

The comparison is presented in figures 5 and 6 for the [A/S] interface of
245 one single cell with the lay-up ([A/S/C]) [0/0/90], where A stands for anode,
S separator and C cathode, from bottom to top. The angles are expressed in
degrees, with respect to the global coordinate system, as shown in figure 3.
The analytical model appears to be in close agreement with the results from
the FEM analysis, however the latter generally predicts higher magnitudes of
250 interlaminar stresses σ_{33} close to the free edge, while being fundamentally similar
in the region $y/2t > 0.01$.

Figure 5: σ_{23} between [A/S]

Figure 6: σ_{33} between [A/S]

3. Results and discussion

The assumed material properties for the plies are given in table 1 which includes the assumed coefficients of expansion (L and T). Free edge stresses are evaluated for faces I and II as indicated in figure 3. Placing two cells stacked one on top of the other represents the simplest lay-up to produce a symmetric battery laminate (two cells) thus ensuring there is no global bending of the stack. The two lay-ups analysed here are: $[A/S/C/C/S/A]$ and $[C/S/A/A/S/C]$. It is further restricted in this case to cross ply laminates in order to avoid in-plane shear deformations, hence the cathodes and anodes are placed exclusively at 0° or 90° . For the two lay-ups the results will naturally be the same and only two different orientations need to be considered: for instance considering $[A/S/C/C/S/A]$ the cases to consider are $[0/0/0/0/0/0]$ and $[0/0/90/90/0/0]$. In other configurations directions 1 and 2 would simply be exchanged. The results are studied for varying charge currents. The in-plane strains 1 and 2 for these two configurations are shown in figure 7.

Figure 7: In-plane strains in symmetric two-cell battery laminate with lay-up $[A/S/C]_s$

Placing both the anode and cathode in the same orientation appears to reduce the overall expansion of the battery laminate compared to using a $[0/90]$ lay-up. Considering the symmetric cases mentioned above, the risk of delamination is analysed using the quadratic criterion given in equation 75. The values for shear and tensile stresses are taken from [16]: in plane shear strength $Z_{13} = 100MPa$, $Z_{23} = 100MPa$ and $Z_{33} = 50MPa$. These are generic values for unidirectional carbon fibre/epoxy laminates and will not necessarily be representative for a future structural battery but are used herein for the sake of comparison. The cells are charged at a charge current of $15mA/g$ that gives a capacity of $345[\frac{mAh}{g}]$ which is the maximum capacity for which there is experimental data. The value of the delamination criterion f_{risk} from equation 75 is presented in figures 8 and 9 for two configurations: $[A/S/C]_s$ and $[C/S/A]_s$ for varying anode and cathode lay-up angles. The results for face I is basically the same as for face II, but mirrored around an anode angle of 45° (a certain f_{risk} on face II at a given angle θ , the same f_{risk} will be found on face I at the angle $90^\circ - \theta$).

As seen in figure 8, the laminate $[A/S/C]_s$ is at risk of delamination ($f_{risk} > 1$) for orientations $[15/0/15]_s$ (on face II) and $[75/0/75]_s$ (on face I). The case of $[C/S/A]_s$ is different as shown in figure 9. The cathode shrinks in the outer plies of the laminate and the anode expands in the middle hence the risk of delamination in the interface between the two cells (where the two anodes are in contact) is high. This should indeed be considered while designing the architecture of the full battery laminate: having the anode as outer plies appears to be a safer configuration because the normal stresses are mostly compressive and hence reduce the risk of delamination.

Energy storage is not the only possible application for the described battery system. The deformation of the cell could provide another function, for instance in the context of solid state mechanical actuators. By controlling the lay-up and charging parameters it is possible to obtain a desired displacement. Some possible examples could be to maximise the pure twist of the laminate or bending around one axis.

Figure 8: Comparison of the delamination risk on face II for the two-cell battery laminate with lay-ups $[A/S/C]_s$ at a charge current of 15 mA/g

In order for example obtain a pure twist with no other curvatures around the x_1 or x_2 axes, an antisymmetric angle-ply can be designed. The stack of two cells in the forms $[A/S/C/C/S/A]$ and $[C/S/A/A/S/C]$ are again used as “base” cases. The possible lay-ups are shown in table 2. The charge current considered is $15mA/g$, however the current required to obtain a specific curvatures can be easily calculated using the model. The maximum value of the twisting curvature κ_{12} which can be obtained is also shown in table 2. Note that the values corresponding to the two lay-ups are different because the distances of the plies from the laminate mid-plane which influences the twist.

An example of maximum twist is illustrated in figure 10. The figure is plotted to scale of a $[A/S/C/C/S/A]$ in orientation $[10/0/0/0/0/-10]$ at a charge current of $15mA/g$. Approximately the same twisting curvature can be obtained with a $[C/S/A/A/S/C]$ with orientations $[15/0/0/0/0/-15]$, however, that configuration will result in significantly higher free edge delamination stresses.

Creating a bending actuator can be envisaged in a similar manner using a

Figure 9: Comparison of the delamination risk on face II for the two-cell battery laminate with lay-ups $[C/S/A]_s$ at a charge current of 15 mA/g

Table 2: Maximum curvature laminate orientations

<i>Lay – up</i>	$ \kappa_{12} _{max}$	<i>Orientation</i> $_{max}$
$[A/S/C/C/S/A]$	$3.875e - 3$	$[10/0/0/0/0/ - 10]$
$[C/S/A/A/S/C]$	$3.615e - 3$	$[15/0/0/0/0/ - 15]$
<hr/>		
	$ \kappa_{11} _{max}$	
$[A/S/C/C/S/A]$	$4.792e-3$	$[90/0/90/0/0/0]$
$[C/S/A/A/S/C]$	$4.851e-3$	$[90/0/90/0/0/0]$

cross-ply lay-up sequence. For a symmetric two-cell configuration the maximum bending curvatures κ_{11} are included in table 2.

315 4. Conclusions

This study has presented a method to analyse the global deformations and interlaminar stresses in a structural battery laminate that arise from large vol-

Figure 10: Twist of 200 by 40 mm laminate with lay-up [A/S/C/C/S/A] in orientation [10/0/0/0/0/-10]. Plotted to scale. Legend refers to delamination risk.

ume changes in carbon fibres when intercalated with Li-ions. The method is based on an extension of CLPT by Lagace and Kassapoglou [9] that includes
 320 3D stress shape functions near the laminate edges and has been modified here to allow unbalanced and unsymmetric lay-ups to be analysed. Although several orders of magnitude larger, the intercalation expansions were modelled analogously to thermal expansions — with intercalation coefficients relating the electrode capacity linearly to its expansions. The method was verified using
 325 3D FEM simulations which indicate that the method is accurate enough for design purposes. The model allows the study of the magnitude of interlaminar stresses and hence the risk of delamination damage due to the electrochemically induced expansions. This risk was evaluated using a Quadratic Delamination Criterion developed by Brewer and Lagace [16]. It was clearly seen that a symmetric structural battery laminate in sequence [Anode/Separator/Cathode]_s develops significantly lower interlaminar stresses than the opposite configuration [Cathode/Separator/Anode]_s. The model further allows for the design of solid
 330 state actuation mechanisms. This can be achieved by making laminate configu-

335 rations that maximise global curvatures thanks to the carbon fibre expansions.

This work contributes significantly to the understanding of the practical design of structural battery laminates, and provides a useful tool for future development of the technology.

5. Acknowledgements

340 We thank the Swedish Research Council, projects 621-2012-3764 and 621-2014-4577, the Swedish Energy Agency, project 37712-1 and the strategic innovation programme LIGHTer (provided by Vinnova, the Swedish Energy Agency and Formas) for financial support. The Swedish research group Kombatt is acknowledged for its synergism throughout this work.

Appendix A. f -coefficients

345 The following f -coefficients for the expression of the energy derivatives have been obtained:

$$f_1 = \frac{1}{2} \sum_{k=1}^N (B_3^{(k)^2} \frac{t^{(k)^3}}{3} + B_3^{(k)} B_4^{(k)} t^{(k)^2} + B_4^{(k)^2} t^{(k)}) \tilde{S}_{22} \quad (\text{A.1})$$

$$\begin{aligned} f_2 = \frac{1}{2} \sum_{k=1}^N t^{(k)} [& B_3^{(k)^2} \frac{t^{(k)^6}}{252} + B_3^{(k)} B_4^{(k)} \frac{t^{(k)^5}}{36} + (4B_3^{(k)} B_6^{(k)} + 3B_4^{(k)^2}) \frac{t^{(k)^4}}{60} + \\ & (B_3^{(k)} B_7^{(k)} + 3B_4^{(k)} B_6^{(k)}) \frac{t^{(k)^3}}{12} + (B_4^{(k)} B_7^{(k)} + B_6^{(k)^2}) \frac{t^{(k)^2}}{3} + \\ & B_6^{(k)} B_7^{(k)} t^{(k)} + B_7^{(k)^2}] \tilde{S}_{33} \end{aligned} \quad (\text{A.2})$$

$$\begin{aligned} f_3 = \frac{1}{2} \sum_{k=1}^N [& B_3^{(k)^2} \frac{t^{(k)^5}}{20} + B_3^{(k)} B_4^{(k)} \frac{t^{(k)^4}}{4} + (B_4^{(k)^2} + B_3^{(k)} B_6^{(k)}) \frac{t^{(k)^3}}{3} + \\ & B_4^{(k)} B_6^{(k)} t^{(k)^2} + B_6^{(k)^2} t^{(k)}] S_{44} \end{aligned} \quad (\text{A.3})$$

$$f_4 = \frac{1}{2} \sum_{k=1}^N [B_1^{(k)^2} \frac{t^{(k)^5}}{20} + B_1^{(k)} B_2^{(k)} \frac{t^{(k)^4}}{4} + (B_2^{(k)^2} + B_1^{(k)} B_5^{(k)}) \frac{t^{(k)^3}}{3} + B_2^{(k)} B_5^{(k)} t^{(k)^2} + B_5^{(k)^2} t^{(k)}] S_{55} \quad (\text{A.4})$$

$$f_5 = \frac{3}{2} \sum_{k=1}^N (B_1^{(k)^2} \frac{t^{(k)^3}}{3} + B_1^{(k)} B_2^{(k)} t^{(k)^2} + B_2^{(k)^2} t^{(k)}) \tilde{S}_{66} \quad (\text{A.5})$$

$$f_6 = \frac{1}{2} \sum_{k=1}^N [B_3^{(k)^2} \frac{t^{(k)^5}}{30} + B_3^{(k)} B_4^{(k)} \frac{t^{(k)^4}}{6} + (2B_3^{(k)} B_6^{(k)} + B_4^{(k)^2}) \frac{t^{(k)^3}}{6} + (B_4^{(k)} B_6^{(k)} + B_3^{(k)} B_7^{(k)}) \frac{t^{(k)^2}}{2} + B_4^{(k)} B_7^{(k)} t^{(k)}] \tilde{S}_{23} \quad (\text{A.6})$$

$$f_7 = \frac{1}{2} \sum_{k=1}^N [B_1^{(k)} B_3^{(k)} \frac{t^{(k)^3}}{3} + (B_2^{(k)} B_3^{(k)} + B_1^{(k)} B_4^{(k)}) \frac{t^{(k)^2}}{2} + B_2^{(k)} B_4^{(k)} t^{(k)}] \tilde{S}_{26} \quad (\text{A.7})$$

$$f_8 = \frac{1}{2} \sum_{k=1}^N [B_1^{(k)} B_3^{(k)} \frac{t^{(k)^5}}{30} + (3B_1^{(k)} B_4^{(k)} + B_2^{(k)} B_3^{(k)}) \frac{t^{(k)^4}}{24} + (2B_1^{(k)} B_6^{(k)} + B_2^{(k)} B_4^{(k)}) \frac{t^{(k)^3}}{6} + (B_1^{(k)} B_7^{(k)} + B_2^{(k)} B_6^{(k)}) \frac{t^{(k)^2}}{2} + B_2^{(k)} B_7^{(k)} t^{(k)}] \tilde{S}_{36} \quad (\text{A.8})$$

$$f_9 = \frac{1}{2} \sum_{k=1}^N [B_1^{(k)} B_3^{(k)} \frac{t^{(k)^5}}{20} + (B_1^{(k)} B_4^{(k)} + B_2^{(k)} B_3^{(k)}) \frac{t^{(k)^4}}{8} + (B_1^{(k)} B_6^{(k)} + B_3^{(k)} B_5^{(k)} + 2B_2^{(k)} B_4^{(k)}) \frac{t^{(k)^3}}{6} + (B_2^{(k)} B_6^{(k)} + B_4^{(k)} B_5^{(k)}) \frac{t^{(k)^2}}{2} + B_5^{(k)} B_6^{(k)} t^{(k)}] S_{45} \quad (\text{A.9})$$

$$f_{10} = \sum_{k=1}^N (\alpha_2^{(k)} - \frac{S_{12}}{S_{11}} \alpha_1^{(k)}) [B_3^{(k)} C_{rev} \frac{t^{(k)^2}}{2} + B_4^{(k)} C_{rev} t^{(k)}] \quad (\text{A.10})$$

$$f_{11} = \sum_{k=1}^N (\alpha_{12}^{(k)} - \frac{S_{16}}{S_{11}} \alpha_1^{(k)}) [B_1^{(k)} C_{rev} \frac{t^{(k)^2}}{2} + B_2^{(k)} C_{rev} t^{(k)}] \quad (\text{A.11})$$

with

$$\tilde{S}_{22} = (S_{22}^{(k)} - \frac{S_{12}^{(k)^2}}{S_{11}}) \quad (\text{A.12})$$

$$\tilde{S}_{23} = (S_{23}^{(k)} - \frac{S_{12}^{(k)} S_{13}^{(k)}}{S_{11}}) \quad (\text{A.13})$$

$$\tilde{S}_{26} = (S_{26}^{(k)} - \frac{S_{12}^{(k)} S_{16}^{(k)}}{S_{11}}) \quad (\text{A.14})$$

$$\tilde{S}_{33} = (S_{33}^{(k)} - \frac{S_{13}^{(k)^2}}{S_{11}}) \quad (\text{A.15})$$

$$\tilde{S}_{36} = (S_{36}^{(k)} - \frac{S_{13}^{(k)} S_{16}^{(k)}}{S_{11}}) \quad (\text{A.16})$$

$$\tilde{S}_{66} = (S_{66}^{(k)} - \frac{S_{16}^{(k)^2}}{S_{11}}) \quad (\text{A.17})$$

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