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A DETAILED SIMULATION MODEL TO EVALUATE THE CRASH SAFETY OF A LI-ION POUCH BATTERY CELL

BENJAMIN SCHAUFELBERGER[1*], ANJA ALTES[1], ANDREAS TRONDL[2], THOMAS KISTERS[3], CLEMENS FEHRENBACH[2], PASCAL MATURA[1] AND MICHAEL MAY[1]

[1] Fraunhofer EMI, Ernst-Zermelo-Straße 4, 79104 Freiburg, Germany
[*]Benjamin.Schaufelberger@emi.fraunhofer.de

[2] Fraunhofer IWM, Wöhlerstraße 11, 79108 Freiburg, Germany

[3] Fraunhofer EMI, Am Klingelberg 1, 79588 Efringen-Kirchen

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Abstract. *In crash situations with an electric vehicle, the integrity of the battery cells is critical for the consequences of the crash. A short circuit triggered by deformation and damage of the internal cell structure can cause overheating of the battery (thermal runaway) and may result in a vehicle fire or even an explosion. Thus, for assessing the crashworthiness of electric vehicles, evaluating the deformation states of potential crash situations with respect to the occurrence of a short circuit is crucial. A particular challenge for building a cell model with acceptable computational time lies in the very different spatial scales regarding the overall cell size and the thickness of individual layers. Cells installed in vehicles have dimensions of several centimeters, whereas the thickness of the individual layers is in the micrometer range. Much research has already been conducted based on homogenized cell models that do not explicitly account for the internal layer structure, and existing material models calibrated to experimental data (e.g. [1]-[3]), while explicitly considering the layered structure is just pursued more recently (e.g. [4]-[7]).*

Within our contribution we introduce a detailed numerical model which, as a part of a multilevel simulation approach, can be used to evaluate the criticality of a deformation state. The model mimics the layered structure of the cell, whereby the constitutive properties were determined by in-house experiments on the respective materials. For validation, bending tests and indentation tests with different punch geometries along with CT-scans at selected indentation depths are available. Comparing the simulation results with the failure sequence and the force-displacement curve from the experiment, a closer view on critical deformations and on their respective stress states is obtained. The results indicate that in-depth understanding and modelling of the failure behavior is crucial for correctly modeling battery cells under crash loading scenarios.

1 INTRODUCTION

Current homogenized cell models are generally able to model a wide variety of cell deformation states sufficiently well. However, up to now a virtual evaluation of the criticality of these deformation states does not have a satisfying validity. Main reason for this is that the occurrence of short circuits, in crash situations expected to be caused by failure of the separator, is primarily a local phenomenon, strongly dependent on the microstructure. Hence, a model appropriate for evaluation must be able to deduce the separator deformation from the macroscopic deformation pattern. More concrete, for such models the ambitious requirements are as follows: capturing macroscopic deformation modes correctly, modeling local deformation behavior and stress states, predicting the failure of the single layers, especially the separator. A model fulfilling all these requirements is, to the best knowledge of the authors, not yet available. Though, Zhu et al. [5] demonstrate a 2D model for punch indentation, which is also able to reproduce the experimentally measured force-displacement curves, due to missing validation data for intermediate penetration depths, the validity of the assumed failure development is unclear.

In the paper at hand, we outline a detailed modeling approach applied for bending and indentation scenarios with 3D and 2D models respectively. Due to the importance of failure modeling the focus is put on this aspect. The presented model benefits from the fact that within BATTmobil, a research project where Fraunhofer Ernst-Mach-Institut (EMI) and Fraunhofer Institute for Mechanics of Materials (IWM) participated, a comprehensive experimental and numerical analysis of a selected Li-ion pouch cell was realized.

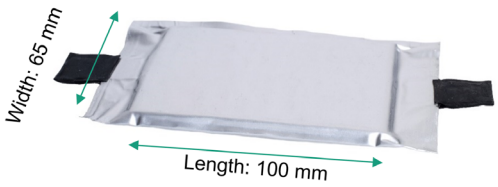
2 OVERVIEW OF EXPERIMENTAL RESULTS

Experimental results presented in the following provide the basis for further modeling. The results from the material characterization are necessary for the calibration of the parameters of the material models. Tests with different intrusion depths, which allow the analysis of the failure sequence, are, besides force-displacement curves, one key aspect for the validation of the deformation and failure behavior.

2.1 Description of the Cell Geometry

For all conducted experiments, a non-commercial Li-ion pouch cell is used. The cell, having a size of ca. 100 mm x 65 mm x 7 mm, consists of 12 coated cathodes (aluminum foils with a lithium-based metal oxide (NMC, lithium nickel manganese cobalt oxide)), 13 coated anodes (copper foils with graphite) and 24 separator foils. An aluminum pouch surrounds the stack of the 99 internal layers. In Figure 1 the cell and the thicknesses of the single components are depicted.

Advantage of the used cell, which was manufactured in a small series at the Fraunhofer Institute for Silicon Technology (ISIT), are: the knowledge of the manufacturing process, the similarity to commercial cells, the availability of the single components (anode, cathode, separator and pouch, being experimentally analyzed in chapter 2.2) and the comparatively small size, which makes it suitable for CT-scanning. The same cell type has already been used for an experimental analysis of rate-dependent failure behavior [8].



	Number of layers	Thickness per layer
Pouch	2	120 µm
Copper	13	18 µm
Graphite	26	83.5 µm
Separator	24	25 µm
Aluminum	12	20 µm
NMC	24	125 µm

Figure 1: Non-commercial Li-ion pouch cell manufactured at Fraunhofer ISIT (left) and a table to outline the thickness of a single layer of the respective material (right). Source [6].

2.2 Material Characterization

Since it is expected that the tensile properties are determined by the pouch, the separator and the metal parts of the electrodes (copper and aluminum for the anode and cathode, respectively), the characteristic of these components is tested in tensile tests. Due to its expected anisotropy, the separator was tested in two directions, for all other materials only unidirectional tests were performed. The coatings (NMC and graphite) have negligible tensile strength, however due to their percent by weight, and the comparatively low stiffness they dominate the compressive behavior of the cell.

In Figure 2 on the left, the stress-strain curves measured in tensile tests are shown. Specimens, cut out of the foils, were tested with a velocity of 1 mm/min. Reported true stress is obtained by relating the measured forces to the current cross section of the specimens, whereas local true strains, in axial and transverse direction, are obtained by digital image correlation (DIC). Compared to all other materials, the metal foils show clearly the highest stiffness and stress levels but also the lowest failure strains. Noticeable is the difference between anode and cathode. Compared to the anode, the cathode has both, lower failure strain and a lower yield stress.

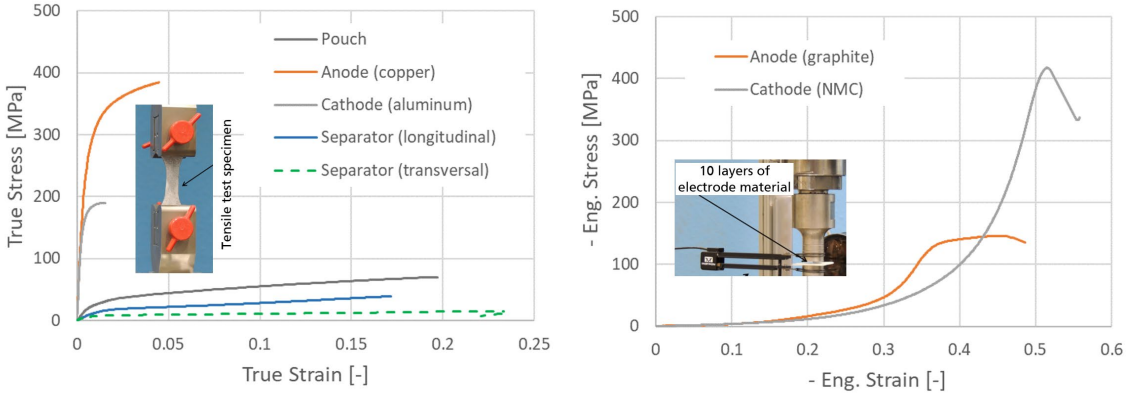


Figure 2: Tensile (left) and compression tests (right) for characterizing the material behavior of cell components. Experiments performed at Fraunhofer IWM.

Compared to the tensile properties of the individual components (depicted in Figure 2 left) the compressive properties, especially of the active materials, are not well understood. Compression tests on specifically casted specimens of graphite anode material are documented in [9]. However, due to the different microstructure (solid material with diameter 16 mm and

height 5 mm) and manufacturing process the applicability of the results for detailed cell models is questionable. Therefore, within the BATTmobil project a different approach was pursued. Stacks of 10 layers of active materials (size 25 mm x 25 mm, height ca. 1.75 mm and ca. 2.75 mm for anode and cathode, respectively) were tested under compression with a rate of 1 mm/min. Before testing, the samples had been soaked in a solvent for 5 h. Comparing the compressive stress-strain curves for anode and cathode in Figure 2 right demonstrates that the stiffness is very similar in the low strain regime (up to roughly 0.2), whereas for larger strains the anode is slightly stiffer but has a lower maximum stress (150 MPa compared to 420 MPa). Engineering strains are calculated by assuming the metal foils being rigid.

2.3 Tests on Battery Cells

In battery tests, cells are typically loaded until a pronounced force drop occurs. Besides these tests, which were performed to identify force-displacement curves over a wide loading range (and used for validation in chapter 4 and 5), also tests with lower indentation depths (interrupted tests) were conducted. Subsequent CT-scanning allows to analyze the development of the failure patterns. By examining the CT-scans, illustrated in Figure 3, it has to be considered that a deformation state after unloading is shown. To avoid relaxation the bended cell was fixed after unloading. (see hollow cylinder in Figure 3 A).

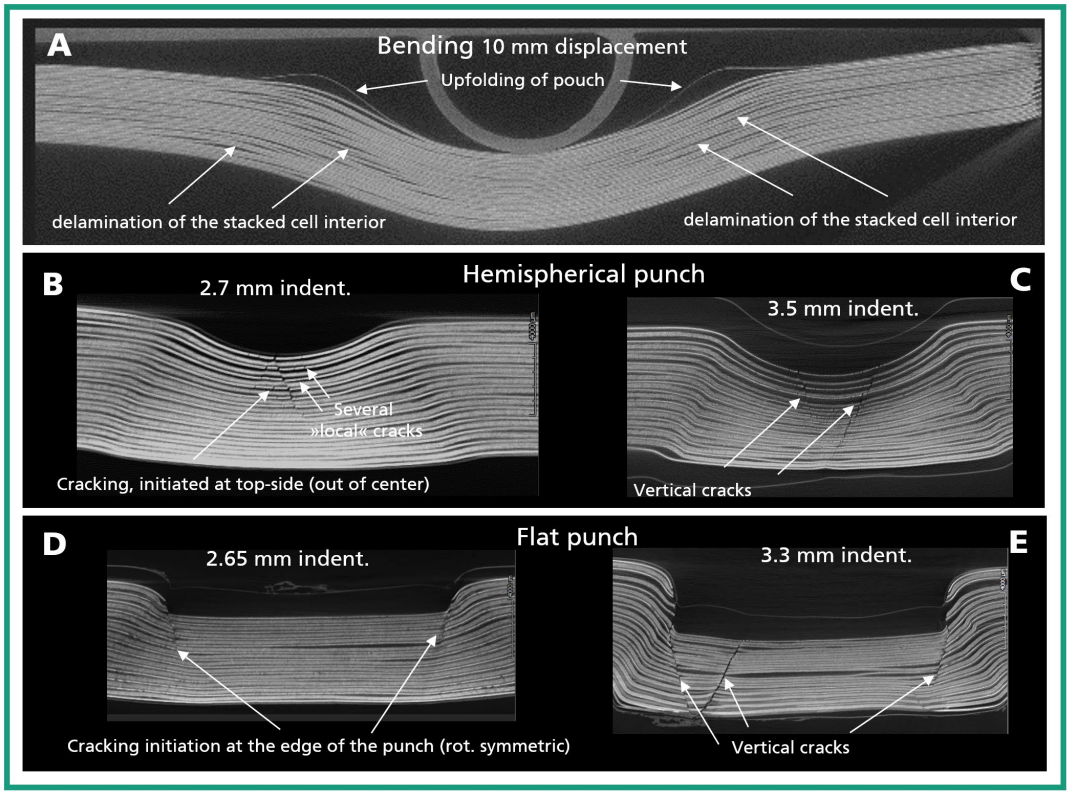


Figure 3: Ex-situ CT-scans of interrupted bending (A), hemispherical (B, C) and flat (D, E) punch indentation tests. Depicted figures are from the length-thickness plane of the cell (compare Figure 1). Anode (due to the high absorptivity of copper the brightest layers) and cathode layers as well as the pouch can be identified separately. Interrupted tests conducted at Fraunhofer IWM, CT-scanning at Fraunhofer EMI.

In bending tests (velocity 1 mm/s) the pouch cell is horizontally placed on two supports (radius 1 cm, distance 6 cm) and loaded with a rod-shaped punch (radius 1 cm). The measured force-displacement curve (see [6] and also Figure 5) has an almost bi-linear shape. Tests with subsequent CT-scanning were performed until 30 mm displacement. Figure 3 A indicates that even for a relatively low displacement of 10 mm an upfolding of the pouch and a delamination of layers occurs. For bending, in none of the analyzed cases a layer-failure could be observed.

Indentation tests were performed with a hemispherical and a flat punch, both having a radius of 1 cm. The load was applied with a velocity of 0.01 mm/s. Besides measuring the force-displacement curve a qualitative analysis of the failure behavior was the major task for the experimental test series. The CT-scans show that before reaching the maximum force (at 3.3 mm for hemispherical and 3.0 mm for flat punch) failure of anode and cathode foils occurs. For hemispherical punch indentation the cracking is initiated (< 2.7 mm) at the top side (Figure 3 B), the crack then propagates further in thickness and width direction. At a displacement of 3.5 mm (Figure 3 C) a distinct second crack is observed. Also, for flat punch indentation cracking is initiated at the top-side, however not at the center line but at the edge of the punch (Figure 3 D). Vertical cracks in width direction develop for larger indentations (> 2.65 mm but < 3.3 mm, see Figure 3 E). It shall also be noticed that in both settings single layers can fail at multiple positions, whereas only a few of these local cracks extend for more than one layer then resulting in pronounced vertical cracks.

3 DESCRIPTION OF THE SELECTED MODELING APPROACH

For simulating the mechanical behavior of pouch cells different, on finite elements (FE) based modeling approaches are possible. Though, for an improved understanding of the identified failure behavior not all approaches are applicable. Frequently used homogenized models for example are unable to model distinct layer-failure. For typically applied boundary condition a variation of stress and strain fields over the thickness is observed. Though, due to inhomogeneity of both of them, using RVEs (representative volume elements) with periodic boundary conditions is not adequate. Therefore, in this work a detailed, representative layer model approach, is used. It is based on modeling the stacked layered structure with representative layers for each component (pouch, copper, aluminum, graphite, NMC and separator). The thicknesses of one layer in the model is thereby calculated by summing up the very thin foils of the experimentally investigated cell, and then subdividing this quantity into the number of representative layers in the model. This allows to reproduce the correct outer dimensions, by considering the local material behavior and the characteristic periodic layering. Moreover, it keeps the computational cost acceptable.

The simplest possible resolution in the model, the so called “period 1”, consists of 13 layers only (see Figure 4): anode (consisting of graphite, copper and graphite), separator, cathode (NMC, aluminium, NMC), separator and anode (graphite, copper and graphite); these layers, representing the cell’s interior, are enclosed by the pouch. In higher “periods” in each iteration this base structure is extended by two separators, one anode and one cathode. In this way a “period 12” model would correlate with the original layering (99 layers). The outlined geometric setting is transferred into a FE model, solved with the commercial software LS-DYNA using explicit time integration. The low thickness of pouch, separator, copper and aluminum foils allows the assumption of membrane behavior, which permits the use of only

one element in thickness direction for each of these layers. In the model, neighbouring layers in the cell-interior are connected by merged nodes, a contact-based interaction is applied between the cell-interior and the pouch.

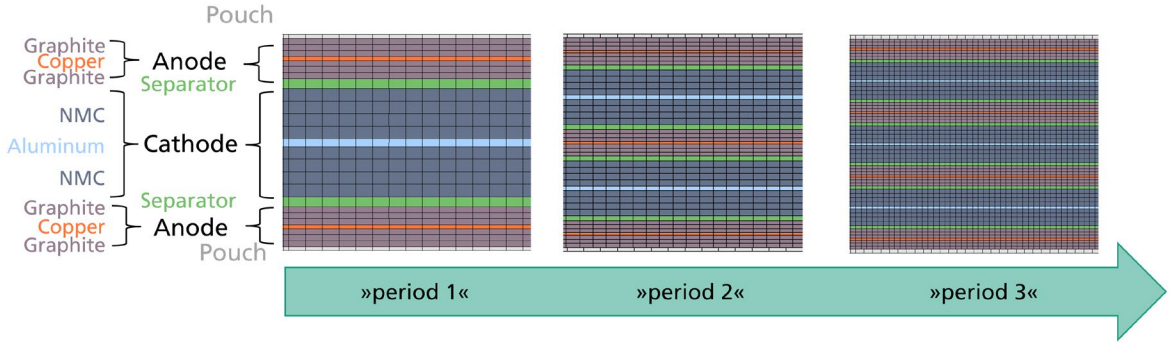


Figure 4: Illustration of the detailed, representative layer model approach which bases on modeling the cell interior with stacked representative layers for each component (material). Based on [6].

To model the material behavior of copper, aluminum and the pouch a linear elastic, piecewise linear plastic material model is used (LS-DYNA material ID 24). Parameters are calibrated to the tensile tests illustrated in Figure 2 left. An anisotropic model (ID 103) is applied for the separator, and a model originally developed for crushable foams (ID 63) for the active materials. Advantage of the foam model, which is also commonly used for homogenized battery cell models (e.g. [1]), is that the compressive behavior is specified by a stress-strain curve. Under tension the model assumes linear behavior until a constant yield stress (named tensile stress cutoff, TSC) is reached.

In the following the outlined modeling approach is applied to simulate the bending and indentation behavior of Li-ion pouch cells. For the indentation simulations, a 2D axisymmetric model is used. Because of this symmetry used, the anisotropy of the separator cannot be modelled, instead the linear elastic, piecewise linear plastic material model (ID 24) is also used here. In bending scenarios, the influence of the surrounding pouch, which cannot be modelled in 2D, is crucial [6], so the use of a 3D model is inevitable. In both configurations, elements with full integration are used.

4 SIMULATION OF DEFORMATION BEHAVIOR UNDER BENDING

The application of the model approach to a bending scenario and subsequent analysis of the basic model parameters developed into a comprehensive study. The procedure for the model development of the bending test has already been published at the 13th European LS-DYNA Conference [6]. In the following, for the sake of completeness, the main conclusions are outlined.

Comparison of 2D and 3D models shows significant differences in the force-displacement curves, whereas only in 3D simulations, the characteristic bi-linear shape (see Figure 5) could be reproduced. Modifications of the pouch in 3D simulations underlined the importance of the closed pouch for the deformation behavior of the cell. Besides the pouch also the layer resolution (“period”) and the tensile strength of the active material have a major impact on the force level, however the initial stiffness remains more or less constant. As illustrated in Figure 5 the force level decreases with increased layer resolution. However, a too low force level can

be compensated by an increase of the tensile strength cutoff (TSC) of the active materials – a parameter which cannot be determined experimentally up to now and which is therefore used for calibrating the force curves in the bending scenario. A further aspect in this context is the more or less unknown shear behavior on the interface between the different layers.

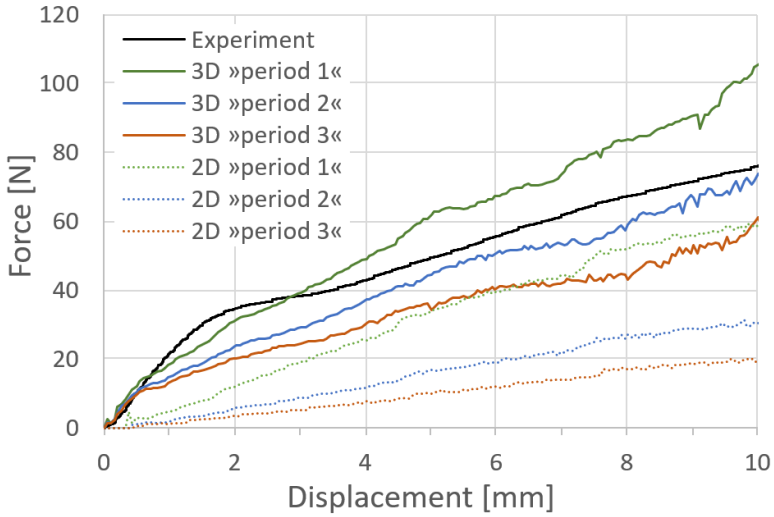


Figure 5: Force-displacement curves for three-point bending: experiment (performed at Fraunhofer EMI) compared to 2D and 3D detailed model simulations. Tensile stress cutoff (TSC) for active material is set to zero. More details are given in [6].

In addition, Figure 6 demonstrates the occurring stresses during bending. In contrast to classical beam bending theory, which for a homogeneous beam implies a stress distribution from compression stress on the top to tensile stresses on the bottom, the stress state in the cell is far more complex, due to the layered structure with distinctly different material properties. Tensile stresses (positive sign, i.e. red in Figure 6) are dominant in copper, aluminum and separator foils whereas locally compressive stresses (negative sign, i.e. blue in Figure 6) occur in NMC and graphite.

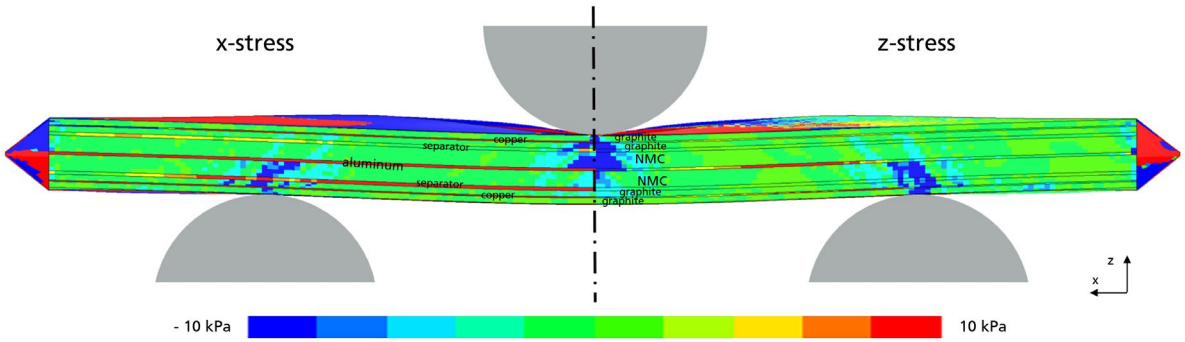


Figure 6: Stresses in x- and z-direction at a displacement of 1 mm simulated with a 3D “period 1” model of the three-point bending test.

5 MODELING THE FAILURE BEHAVIOR IN INDENTATION TESTS

For the time being, the focus is on the principal application of the presented concept and on modeling the experimentally observed failure behavior depicted in Figure 3. In order to meet these objectives, two 2D models were developed. The first model uses only element erosion to model failure, whereas the second one uses element erosion and for selected layers node splitting. Advantage of the chosen 2D approach is that contact modeling, especially after erosion of internal layers, is less complicated than in 3D and also the application of node splitting algorithms is pretty straightforward.

5.1 Hemispherical Punch Indentation using Element Erosion

First, element erosion which is probably the simplest failure modeling technique is used. Using this approach, an element is deleted when a certain parameter, typically a strain, reaches a critical value. Element erosion can be applied to the model without changing the discretization. However, its application results in a non-consistent simulation with regard to mass, volume and energy conservation. The failure parameters are defined as follows: pouch, effective plastic strain 0.18; copper, effective plastic strain 0.04; aluminum, effective plastic strain 0.02 and separator, maximum principal strain 0.22.

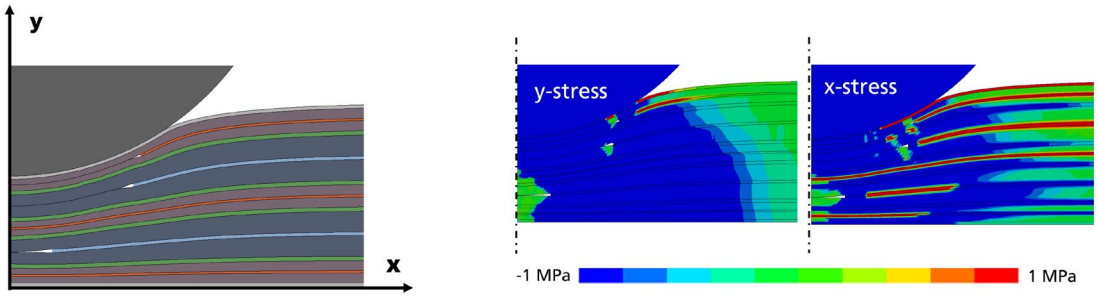


Figure 7: Failure behavior of Li-ion pouch cell indented with a hemispherical punch (penetration depth 2.4 mm), simulated with a 2D axisymmetric “period 2” model using element erosion technique.

Figure 7 demonstrates the failure behavior of the investigated Li-ion pouch cell under hemispherical punch indentation loading. Due to comparatively low failure strains, aluminum and copper layers fail first, aluminum at an indentation of 1.3 mm and copper at 1.6 mm. Failure at the pouch and separator layers starts at 3.1 mm and 4.1 mm respectively. As indicated in Figure 7 these layers fail under an in-plane tensile stress state. Failure of one element results in cascading failure of the neighboring elements within the respective layer. It shall be highlighted that, as the CT-scans of interrupted indentation tests have already shown, the failure of metal foils is also in the simulation initiated considerably before the experimental force-peak (at 3.3 mm).

Comparing the force-displacement curves from experiment and simulation (with erosion) in Figure 8 points out the good accordance of the two curves until first elements (at 1.3 mm indentation) are deleted. For indentations > 1.3 mm element deletion, with a loss of volume and as a consequence also a local loss of contact, results in an underestimation of the force. By neglecting any failure in the model, experimental and simulated force-displacement curves (green curve in Figure 8) agree well until 2.1 mm penetration depth; beyond that, the simulation

behaves slightly too stiff. For the sake of completeness, it shall also be noted that for the loading case under consideration both, the tensile stress cutoff of the active materials (TSC) and the layer resolution ("period") has minor influence on the force-displacement curve.

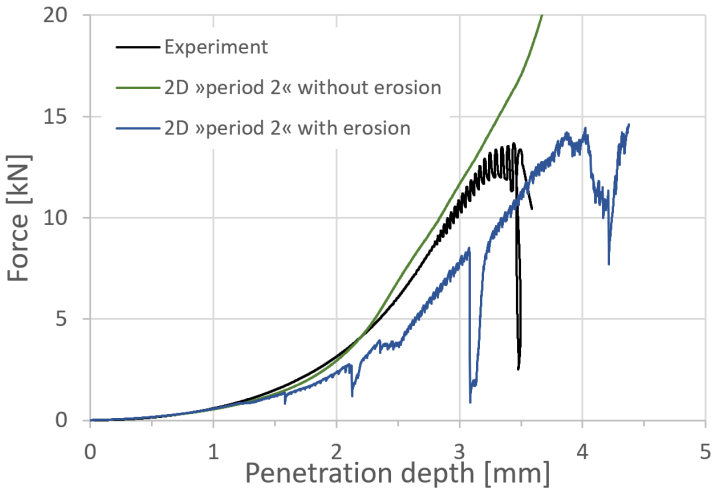


Figure 8: Force-displacement curves for hemispherical punch indentation. 2D axisymmetric simulation results with "period 2" resolution compared to the experiment. Experiment performed at Fraunhofer EMI.

5.2 Modeling Hemispherical Punch Indentation with Node Splitting

As outlined above the early deletion of copper and aluminum elements results in too low stiffness during indentation, so in a next step, failure of these two layers is modeled with node splitting. Applying node splitting requires an adaption of the mesh. Coincident nodes (instead of merged nodes) along with individual tie-break constraints between neighboring elements (within one layer) are needed. For more details on the node splitting algorithm in LS-DYNA see for example [10]. In the model at hand, the critical parameters for disconnection, which in LS-DYNA can only be an effective plastic strain, are 0.02 for aluminum and 0.04 for copper. Due to the fact that copper and aluminum are no longer modeled with merged nodes, the merged node connection to the active materials must be adapted. Hence, it was decided to use a contact-based interaction between all layers.

The force-displacement curves depicted in Figure 9 left shows a good agreement between experiment and simulation until the experimental force peak is reached at 3.3 mm. So, the main objective for applying node-separation – the avoiding of too low stiffness in the range between 1.3 mm and 3.3 mm – is achieved. In the simulation, distinct force drops occur at 2.8 mm and 4.3 mm. These are caused by failure and element deletion of pouch and separator respectively (see also Figure 9 right). Comparing the qualitative failure behavior of copper and aluminum in Figure 9 right with the CT-scans illustrated in Figure 3 underlines the plausibility of the numerically predicted several local cracks in the metal and aluminum foils over the whole deformation zone. Whereas for the separator, which cannot be identified in the CT-scans, the measured voltage-drop indicates that in the experiment the separator fails earlier than in the simulation (3.3 mm instead of 4.3 mm).

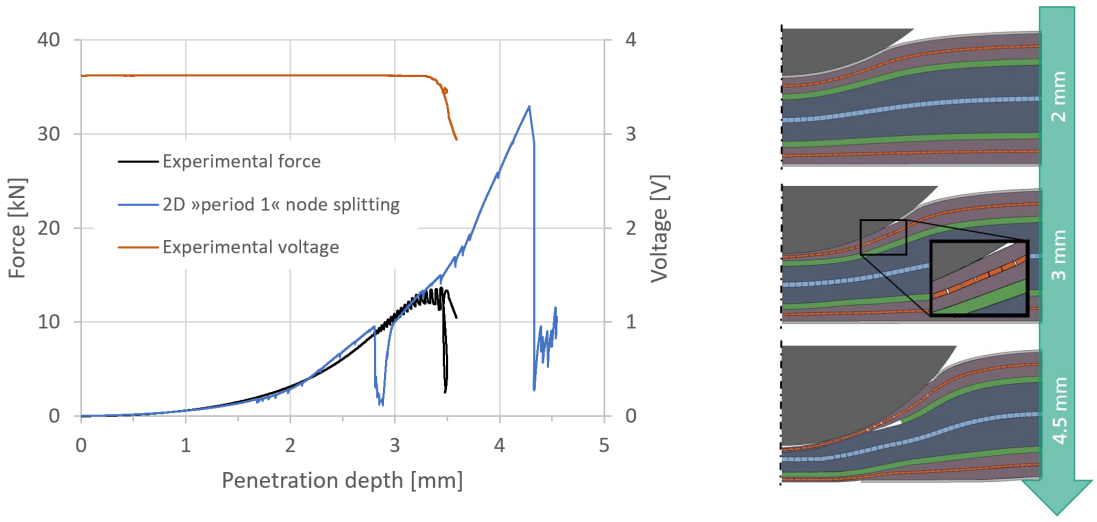


Figure 9: Hemispherical punch indentation modeled with node splitting. Left: comparison of force-displacement curve in experiment (performed at Fraunhofer EMI) and simulation. Right: simulated failure sequence.

5.3 Flat Punch Indentation with Node Splitting

The cell model with node splitting for copper and aluminum is also applied for the flat punch indentation test. Figure 10 left shows that the model predicts the experimental force-displacement curve well until a displacement of ca. 2 mm. For larger displacements the simulation behaves too stiff. The force plateau in the simulation at roughly 40 kN, which is in good agreement with the experimental force peak, is caused by subsequent element erosion of the pouch. Failure of the separator (initiated at 2.6 mm and in good correlation with a second voltage drop in the experiment at 2.4 mm) and ongoing element deletion results in a decline of the force in the simulation. From a qualitative perspective (Figure 10 right), especially the cutting at the edge of the flat punch is well simulated by the model.

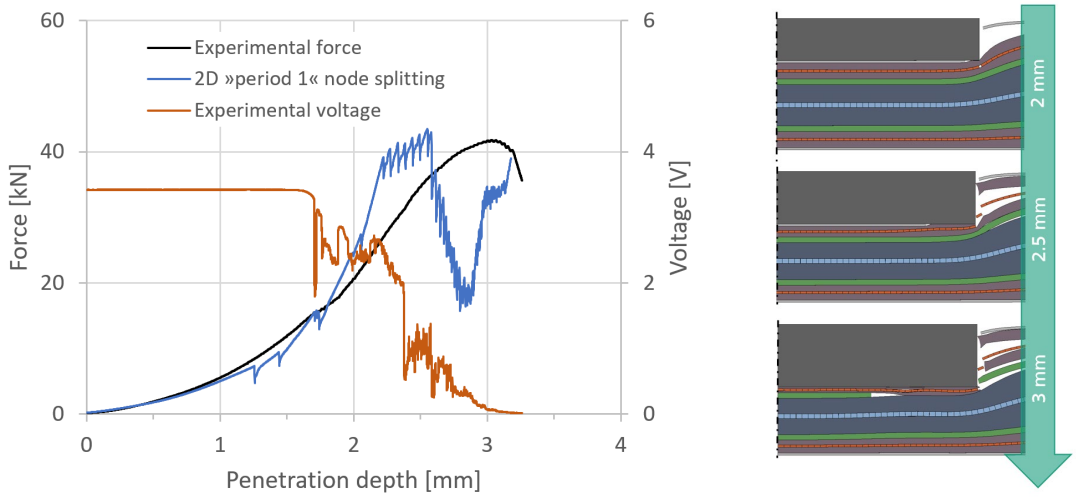


Figure 10: Flat punch indentation modeled with node splitting. Left: comparison of force-displacement curve in experiment (performed at Fraunhofer EMI) and simulation. Right: simulated failure sequence.

5.4 Summary and Discussion of the Failure Modeling

Taking the results from the simulation of hemispherical and flat punch indentation tests, it can be summarized that the model predicts the slope of the force-displacement curve for smaller penetration depths. By applying node splitting the failure of copper and aluminum is simulated well. However, the simulation of the separator failure is not yet satisfying. Whereas the failure initiation in the flat punch simulations correlates with a voltage drop in the experiment, in the simulation of the hemispherical punch indentation the separator fails too late. Simulations (not demonstrated herein) with different layer resolutions (hemispherical punch indentation, using element erosion as failure model) did not show a strong dependence of the separator failure on the layer resolution ("period"), thus the layer resolution can be excluded as one possible reason for this ambiguous result. So, it is expected that the anisotropic behavior of the separator, which is not describable in the axisymmetric model, is the major reason for the observed discrepancy between experiment and simulation. The models also fall short in simulating the experimentally observed force-drop. Assuming crack opening of vertical cracks being the main reason for the force-drop, its correct simulation would require a 3D model as well. Furthermore, in future models also restrictions of the node splitting algorithms have to be considered. Especially modeling pronounced cracks over different layers with strongly different properties (e.g. copper and graphite or aluminum and NMC) is seen as challenging.

6 CONCLUSIONS

The presented simulation models for a Li-ion pouch cell base on explicitly considering the stacked layered structure and the material behavior of the single components in the model. The modeling approach is applied to different types of loading: Three-point bending, where the total cell deformation, layer interaction and the pouch are crucial as well as punch indentation, with local deformations and the material and failure behavior of the cell components being dominant. The main conclusions can be summarized as follows:

- In bending, low tensile and shear strength practically lead to a decoupling of the stress states within the different layers in the cell interior. Besides the pouch, the load is basically carried by the copper and aluminum foils under tension. Although strong cell-deformation occurs, in the case at hand, no layer-failure could be observed.
- The slope of the force-displacement curves in punch indentation tests is determined by the stiffness of the active materials under compression. Characterization tests for stacks of coated electrodes can determine necessary compressive stress-strain curves for the material modeling of the active materials.
- Experiments and simulations of punch tests with different penetration depths demonstrate a progressive failure of the internal layers.
- Due to the loss of volume and the resulting decrease of stiffness, modeling the failure of single layers with an erosion criterion leads to the simulation of too low forces after failure initiation. Advanced failure techniques as node splitting can help to mitigate this issue and therefore result in better simulation results.

In view of the presented outcome, it is expected that in the future detailed simulation models can play an important role in the virtual evaluation of the crash safety of Li-ion cells. Though, for an improved simulation of the experimental failure behavior, further work is needed on the prediction of separator failure and force-peak.

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