Simplified modeling of pouch cells under different loadings

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1 Introduction

Due to increasing requirement on the reduction of CO2-Emissions, the meaning of E-Mobility becomes more and more important. The related development of efficient Li-ions with high charge densities has also a direct impact on the automotive industry. This applies in particular to the crash safety of Li-ion-battery-powered vehicles. The structure of Li-ion batteries is in principle a repetitive layered system. One cell unit consists of two very thin metal foils (typically Al and Cu), which coated with active materials (lithium-metal-oxide/LiMOx and graphite in Fig.1:).

Multilayer setup:
- Thickness of one individual cell-unit ~365 µm
- Pouch-cell consists of ~25-30 individual cell units
- Metallic electrodes (Cu ~10µm / ~3% , Al~15µm / ~4%)
- Separator (Polymer ~25µm / ~14%)
- Active-material with liquid electrolyte and binder (Graphite ~60µm / ~33% ; LiMOx ~85µm / ~46%)
- Active-material represents the main component with a portion of 80%

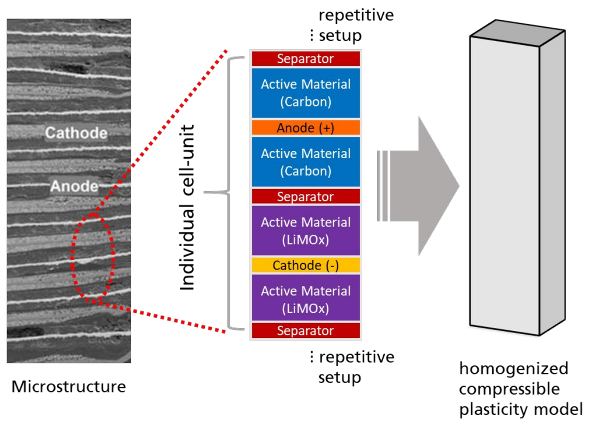

Fig.1: Typical layered setup of a cell-unit within Li-ion battery.

These coated layers represent the anode and the cathode of one unit-cell which are separated by thin polymeric films (separator-foils in Fig.1:). A typical pouch cell consists of hundred and more layers which are embedded in an electrolyte within a bag (pouch, coffee bag). Local deformations resulting from uncontrolled crash loads can lead to critical damage in the separator, which causes an intrinsic short circuit and subsequently an unstable state (thermal runaway) that can result in battery explosions. Therefore, the simulations of intrinsic mechanical deformation states are crucial to predict electrical short circuits.
However, in a full crash simulation of an electrical vehicle it is impossible to model the intrinsic layered system of each pouch cell because there are hundreds of cells installed and each cell consists of more than hundred layers. Due to these facts, it is necessary to develop and apply simplified models, which use a homogenization approach based on the strong repetitive intrinsic layered cell structure [1]. For this modelling approach, anisotropic compressible plasticity models should be well suited, because it is to be expected that the deformation behavior of the cells is different in the layered thickness direction in relation to the in-plane loadings of the parallel layers. Due to the fact of the highly inner repetitive structure of the cell which is encased with a bag-foil, it could be sufficient to use an isotropic compressible plasticity model for the homogenized inner layup combined with surrounding shells to consider the outer bag.
In the following sections numerical investigations are presented to describe the mechanical deformation behavior by using solid elements with different (isotropic and anisotropic) compressible plasticity models for the inner structure, and shell elements with incompressible metal-plasticity for the outer bag. The calculated results are compared to experimental investigations of representative crash loading scenarios, e.g. bending, indentation [2], intrusion and compression in different cell directions. Furthermore, evaluated intrinsic mechanical quantities (volumetric strain) are presented to investigate

the possibility of formulating a mechanical based electrical short circuit criterion for all experimental tested loading cases.

2 Simulation approach for the simplified model of pouch cells

To fit the parameters of the homogenized simplified model different experiments under representative loading scenarios are caried out. For the whole testing regime pouch cells are used with a thickness of around 7mm. The width and length of the cells are 65mm and 110mm respectively (Fig.2:).

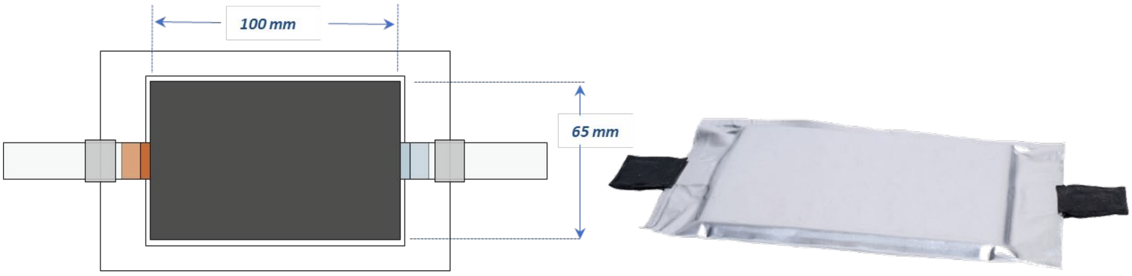

Fig.2: Pouch cell and the simplified analogous model

As mentioned previously, the simplified model consists of the homogenized cell core and the surrounding bag. For the homogenized cell core 8-node solid elements with a compressive plasticity model are used. The bag is modeled with 4-node shell elements by using the same nodes on the outer faces of the cell core. Therefore, no contact must be modeled between the bag and the homogenized cell core. This simplified model approach is shown in Fig.3:. Another important goal in the simplified model development was the high numerical efficiency. Therefore, the model should consist of a number of elements as small as possible, by simultaneous sufficient accuracy. The size of the solid elements is around ~5mm x ~5mm in plain face and 1.75mm in thickness direction. It is important to consider that the element discretization depends on the geometrical size of the expected external penetration objects.

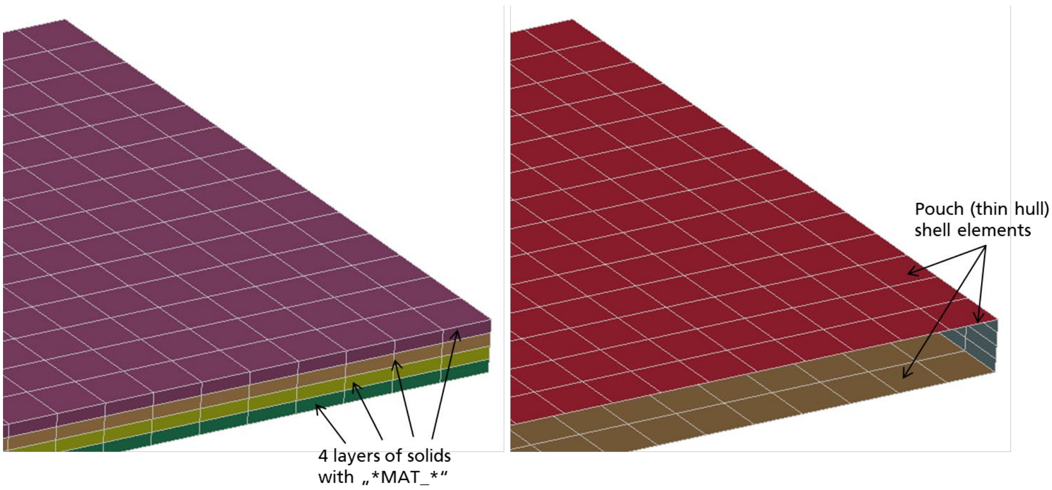

Fig.3: Principle setup and element discretization for the simplified model of a pouch cell

The whole model of the pouch cell consists of 1120 under-integrated 8-node solid elements and 832 under-integrated shell elements with one integration point in thickness direction (membrane behavior). This setup results in a simplified model of less than 2000 elements, which should be also tradable in large crash simulations of complex automotive structures.
Due to the fact that the active material represents the main proportion of the cell material (see Fig.1:), it is assumed that the homogenized material model has a compressible behavior similar to the active material. Therefore, different isotropic and anisotropic material models were used to fit and to simulate the experimental results. Basically, the used compressible plasticity models should be as simple as

possible in its parameterization to limit the effort required for fitting the simulations to the experimental results.

- The isotropic material model ***MAT_CRUSHABLE_FOAM** was applied in its simplified approach. This model was also used in some publications which simulates one or two selected deformation behavior of a pouch cell [3, 4]. However, this model in its simplest form has the disadvantage that the shape of the yield surface is independent of the present hydrostatic pressure (first stress invariant). Therefore, it is sufficient to specify only the hardening (second stress invariant) as a function of the volumetric strain.
- To compare the simple crushable foam model with a more complex isotropic compressible approach, the ***MAT_CSCM** material model was applied. The material model ***MAT_CSCM** [5] is related to the family of Drucker-Prager-Cap models, which have in general a pressure-dependent yield surface. In addition, this model has the advantage of a continuous transition between the linear failure line (for small pressures) and elliptical cap area (for higher pressures), which is based on a multiplicative combination of these two approaches used for the different sections (linear and elliptical) and thus tends to reduce numerical problems in the transition region. The isotropic hardening is implemented with an exponential approach in analytical form, which also has a positive effect on numerical stability.
- Due to characters of the intrinsic layered setup which are explained in Section 1 (Introduction) the anisotropic material model ***MAT_MODIFIED_HONEYCOMB** is also applied. Three (up to six) hardening behaviors regarding to the normal (and/or shear) stress components are available to describe the anisotropy in the model ***MAT_MODIFIED_HONEYCOMB**. The freely definable hardening determines the nonlinear elastoplastic material behavior for all normal and shear stresses separately, whereby the development for all components is completely decoupled and independent from each other.

These three material (***MAT_CRUSHABLE_FOAM**, ***MAT_CSCM**, ***MAT_MODIFIED_HONEYCOMB**) approaches are applied to the simplified model of the pouch cell core and fitted as best as possible to the different experimental tests.

3 Load cases and simulation-setups of different experiments with the simplified model approach of pouch cells

In many publications only one or two load case scenarios were covered in the presented simulations and compared with experimental results [2, 3, 4]. Due to this situation it is mostly possible to fit the model quite well in comparison to the one or two experimental measurements. In practice there are many load cases which are relevant to cover a majority of probable crash scenarios. One interesting question in that context is how accurate the simplified model can describe all experiments. Especially indentation, compression normal and parallel to the intrinsic layered cell plane structure but also bending are expected loadings. Due to this fact one challenge is to cover all relevant loadings as good as possible with the presented simplified model approach. The experimental investigated load cases are shown in Fig.4:

- Normal flat compression test (see upper left in Fig.4:):
  As part of this experiment, a compacting behavior averaged over both active materials (C/Graphite and LiMOx/Lithium-Metal-Oxide) is determined, because it can be assumed that the metallic electrode foils and the separator films are nearly incompressible. Therefore, this testing regime essentially determines the deformation behavior of the simplified model approach because the active material represents the main component in the intrinsic cell core.
- Indentation tests with flat cylindrical (see upper right in Fig.4:) and hemispherical punch geometry:
  In practice, indentations represent relatively likely crash load scenarios. Due to this view there are two different punch geometries (flat and hemispherical head of cylinder) investigated.
- Lateral directed flat compression test (see lower left in Fig.4:):
  Due to the intrinsic structure of Li-ion cells it can be assumed that different deformation patterns for loadings normal and parallel to the layer plane occur.
- Three-point bending test (see lower right in Fig.4:):
  Bending-dominated loadings are also a probable and therefore a relevant load case. In addition, it can be assumed that the thin cell envelope (pouch or bag) has a significant effect on the

deformation behavior, which is the reason for choosing this experiment for estimating the hardening behavior of the bag material.

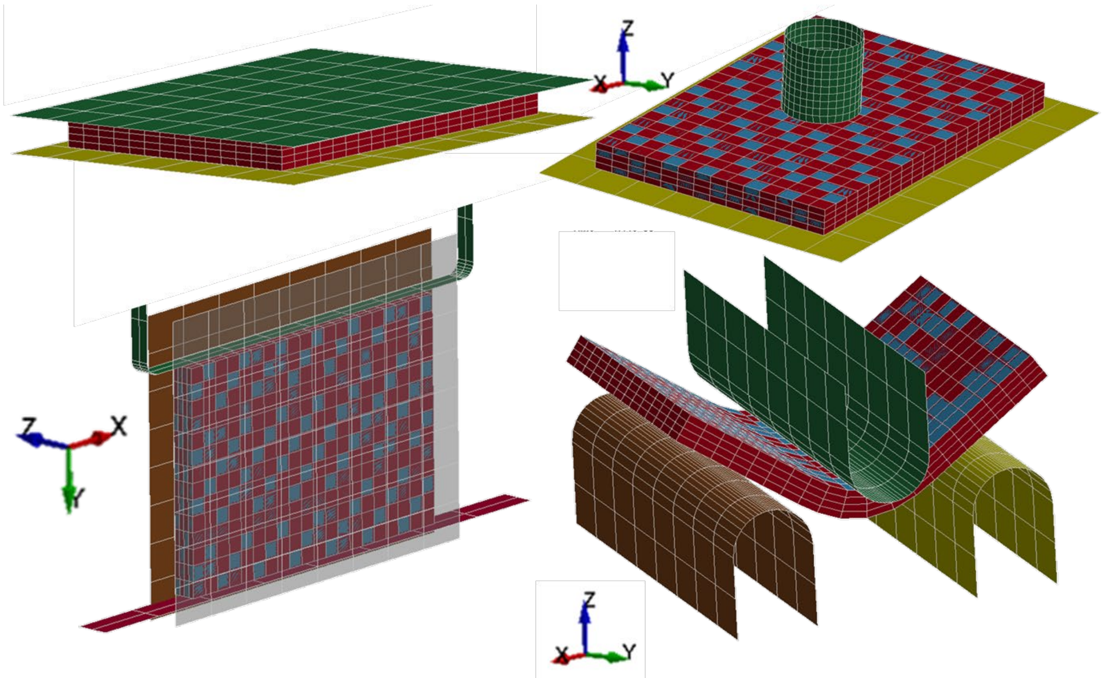

Fig.4: Experimentally investigated and simulated load case scenarios for the simplified model approach of pouch cells

All setups for the simulations of the different experiments are using the same simplified model of the pouch cell (Fig.3:). For the flexible bag (pouch) under-integrated 4-node shell elements with one integration point over the thickness are applied by using the hourglass control type 2 (Flanagan-Belytschko viscous form). The inner homogenized cell-core core domain consists of under-integrated 8-node solid elements and the hourglass control type 6 (Belytschko-Bindeman assumed strain corotational stiffness form) is used. The outer shell elements share the same nodes as the surrounding solid elements of the homogenized cell-core domain (Fig.3:). The material of the outer bag (pouch) is modeled as elastic-plastic by using the **`*MAT_PIECEWISE_LINEAR_PLASTICITY`**. The material parameters for the pouch are experimentally determined with isolated tensile tests of the individual bag [6, 8]. For the cell-core domain the material models which explained in section 2 are used. For each material model the whole series of experiments are simulated and compared to the measurements, which results are explained in the following section 4.
The tools of all experiments are modeled as ideal elastic by using typical parameters for steel (Youngs-modulus of 210000MPa and Poisson-ratio of 0.3). For the frictional contact between the pouch cell and the tools a Coulomb-coefficient of 0.08 is assumed. The whole simulations are calculated under displacement-controlled boundary condition by a predefined velocity of 0.1mm/s, which ensures quasistatic conditions and matches to the experimental setup.
The geometrical discretization of the simplified model of the pouch cell is described in section 2. The element length of the rectangular flat tools is about 10mm and for the pouch cell about 6mm. The element length of the spherical and cylindrical punch as well as the tools for the bending test are between 1mm and 2mm and therefore finer in comparison to the pouch cell.

4 Simulation results and comparison with experimental data

In the following subsections the main results of the three material models for the cell core are presented. To adapt the material parameters the global force-displacement curves of the moved punch are used. The objective target of the fit was to minimize the deviation for all force-displacement curves simultaneously in an averaged manner in comparison to the experimental observations.

4.1 Results for *MAT_CRUSHABLE_FOAM

One restriction of the **`*MAT_CRUSHABLE_FOAM`** (in the basic version) is the shape-independence of the yield surface on the existing hydrostatic pressure (first stress invariant). For this reason, it is sufficient for this (simple) model to specify the hardening (in form of the second stress invariant) only as a function of the volumetric strain. With this approach it is already possible, by using a suitable hardening behavior for load scenarios normal to the intrinsic cell layers, to achieve good agreements between simulations and experiments.

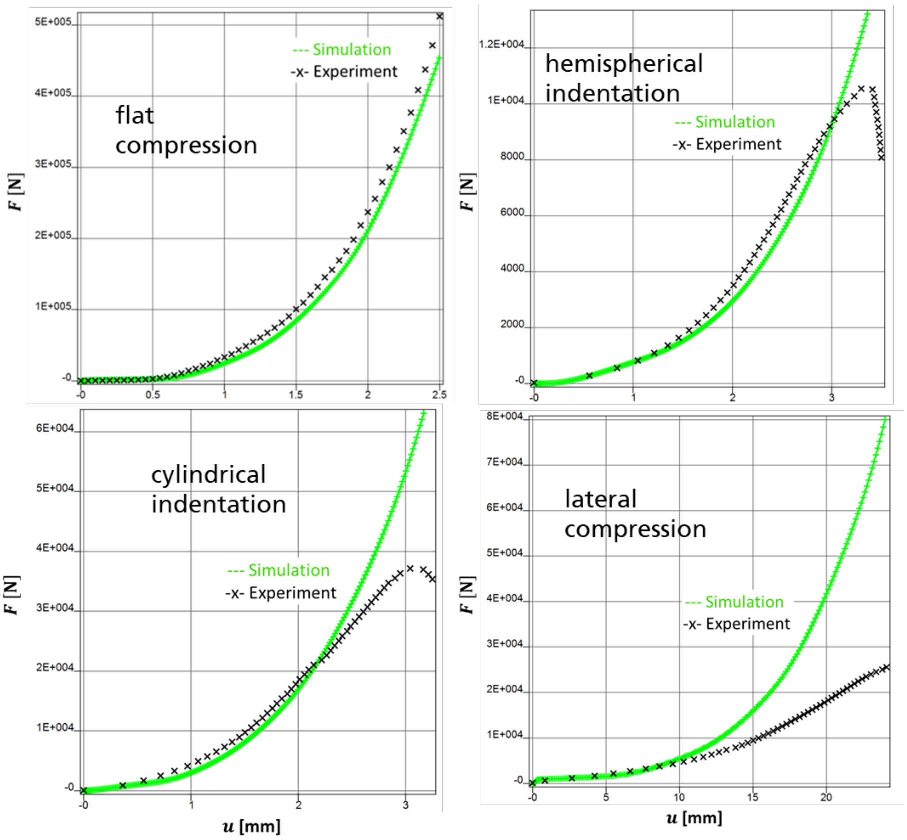

*Fig.5: Comparison between simulation and experiments of force-displacement curves for different load cases by using **`*MAT_CRUSHABLE_FOAM`***

Especially for the flat compression test a good match between simulation and experiment can be achieved, which is shown in upper left diagram of Fig.5:. Similar useful agreement between simulation and experiment can be observed for small and medium punch displacements in the indentation tests (see upper right and lower left in Fig.5:). However, for higher punch displacements, there are larger deviations in the indentation tests compared to the flat compression test, which is related to the absence of a suitable mechanical damage/failure model.
A problem with the use of **`*MAT_CRUSHABLE_FOAM`** occurs for the lateral compression test (lower right diagram in Fig.5:), in which the loading direction is orthogonal to the indentation or flat compression tests and thus parallel to the intrinsic layered setup of the cell. The different behavior for the directed lateral compression test results on the one hand from the mentioned pressure independence of the yield surface and on the other hand from the probable anisotropic behavior parallel and normal to the intrinsic cell layered structure. It can be assumed that with pressure-dominated loads normal to the intrinsic cell layered structure, primarily only the active materials are compacted, whereas with loading parallel to the cell layer structure, complex deformations of the thin electrodes and separator foils (buckling, bending, folding ...) also occur, which have appropriated effects caused by the probable transversal isotropic behavior.

4.2 Results for *MAT_CSCM

Due to the deviations in the global force-displacement curves in case of larger punch displacements for the indentation tests (see upper right and lower left in Fig.5:), as well as the problematic behavior in the lateral compression test (lower right diagram in Fig.5:), the alternative material model **`*MAT_CSCM`** [5] was used. For this modeling approach, a readjustment of the material parameters of the simplified cell core was necessary.

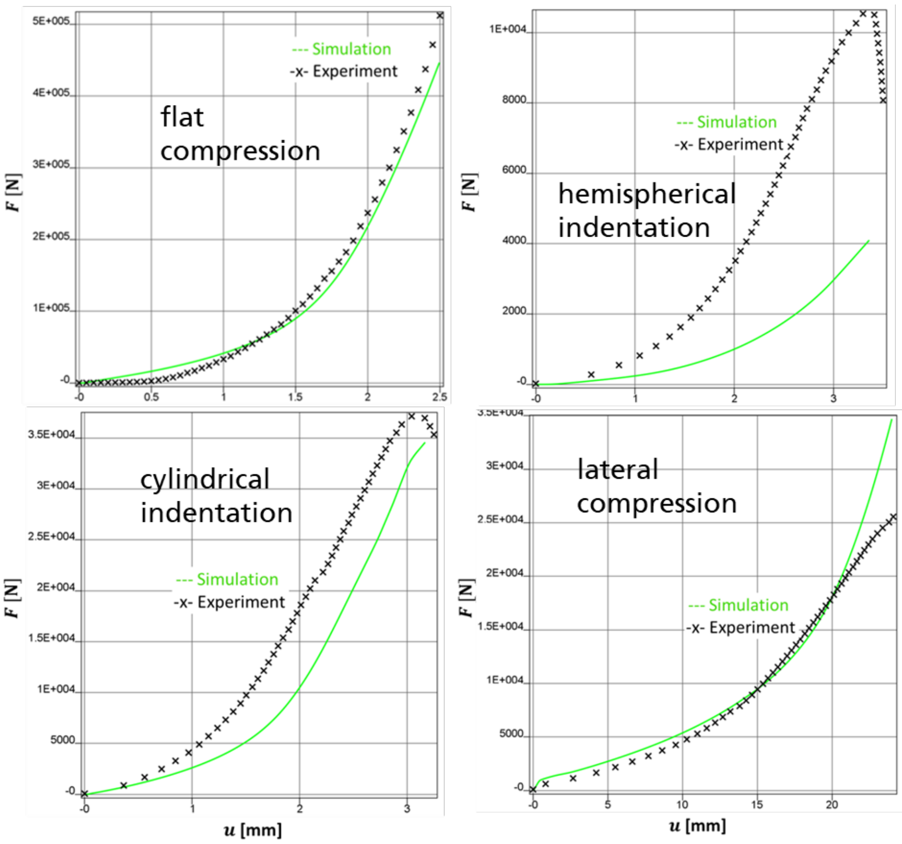

*Fig.6: Comparison between simulation and experiments of force-displacement curves for different load cases by using **`*MAT_CSCM`***

In Fig.6: it can be seen that a suitable adaptation of the **`*MAT_CSCM`** model can achieve a better prediction of the lateral compression test in comparison to the experiment (Fig.6: lower right). The improved adaptation is possible through the already mentioned pressure dependency of Drucker-Prager cap material models, which can be controlled by appropriate parameterization of the flow surface shape (cap eccentricity, slope of the shear failure line, ...). Because in the simple foam model **`*MAT_CRUSHABLE_FOAM`** only the hardening behavior can be parameterized and the shape of the yield surface is independent of the hydrostatic pressure, no simultaneous agreement between simulations and experiments can be achieved.

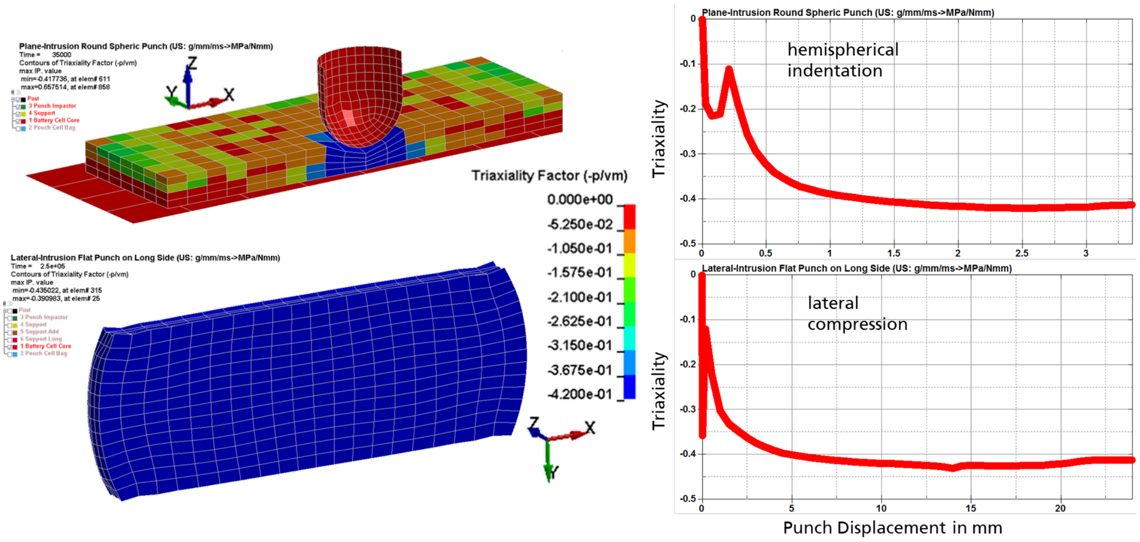

Fig.7: Triaxialities for hemispherical indentation and lateral directed compression (left) and the corresponding evolution of Triaxialities (right)

However, it is also remarkable that the indentation experiment for the hemispherical punch shows a behavior which is too soft compared to the experiment (Fig.6: upper right). This behavior results from an almost identical evolution of the stress history in the dominant deformation zones of both test types (lateral compression test and hemispherical indentation), whereby the final state of triaxiality is shown on the left side of Fig.7:.
The evolutions of the triaxiality in these two tests are very similar (see right side of Fig.7:) but the hydrostatic pressure of the lateral directed compression test rising significant faster. Due to these two circumstances it is not possible to fit both loading regimes simultaneous to the observed test data by using the isotropic model **`*MAT_CSCM`**. Therefore, the hemispherical deformation behavior is too soft, because the model parameters of **`*MAT_CSCM`** are already adapted to the lateral directed compression test.

4.3 Results for *MAT_MODIFIED_HONEYCOMB

To describe the anisotropy in the **`*MAT_MODIFIED_HONEYCOMB`** model, three hardening behaviors regarding to the normal stress components and three hardening behaviors regarding to the shear stress components are available. The freely definable hardening curves determine the non-linear elastoplastic yielding for all normal and shear stresses separately, whereby the evolutions of these quantities are completely decoupled.
For the adjustments in Fig.8: with **`*MAT_MODIFIED_HONEYCOMB`**, it can be noted that the spherical indentation shows a similar problem as already described for the **`*MAT_CSCM`** model. But the deviations of the **`*MAT_MODIFIED_HONEYCOMB`** to the experimental curve are significantly reduced in comparison to **`*MAT_CSCM`**.
Basically, with the anisotropic material model **`*MAT_MODIFIED_HONEYCOMB`**, a good adjustment for all experimental observations can be achieved, which can be clearly seen in Fig.8: compared to Fig.7:.

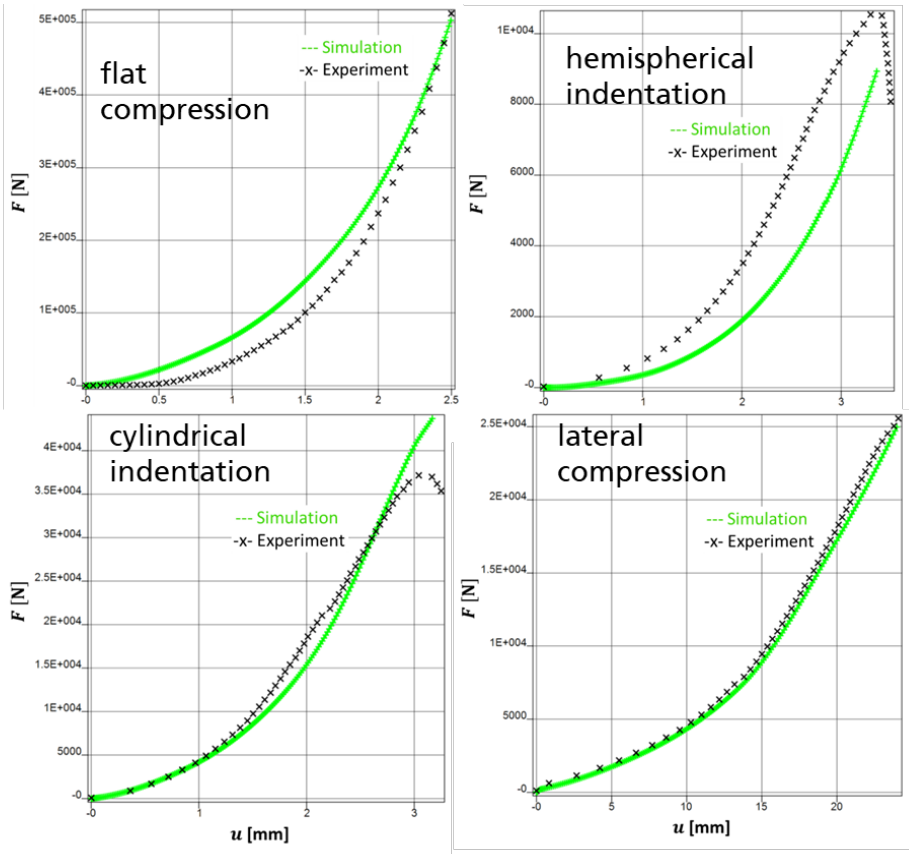

Fig.8: Comparison between simulation and experiments of force-displacement curves for different load cases by using ****MAT_MODIFIED_HONEYCOMB***

According to the results from the numerical simulations, the three-point bending test [6] is significantly dependent on the material properties and the film thickness of the bag (or pouch). A conventional isotropic, incompressible plasticity model (***MAT_PIECEWISE_LINEAR_PLASTICITY**) for metals was used to describe the pouch (bag), which has a film thickness of approx. 0.1 mm.

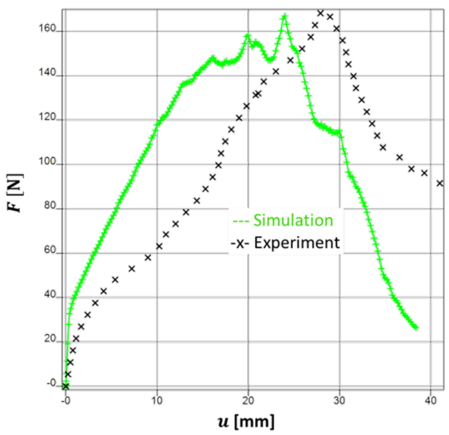

Fig.9: Comparison between simulation and experiments of force-displacement curves for three-point bending test by using ****MAT_MODIFIED_HONEYCOMB***

Fig.9: shows a good agreement between experiment and simulation, although the initial stiffness is a little too high. It can be also remarked that the drop in force is caused by a structural instability in the system, because no failure could be observed in the tested cell or computer tomography CT [7]. Since the simulation without damage or failure approach also shows the drop in force, it can be assumed that the model describes the real deformations for the bending test sufficiently well.

5 Electrical short cut

In most cases, electrical failure is caused by the loss of the insulating capacity of the separator foils, which is in crash scenarios directly related to the mechanical damage and failure behavior of the separator foils. Since the separator foils are not directly described in the homogenized model approach due to reasons of numerical simulation efficiency, the primary task in the development of "electrical failure modeling" is to identify corresponding mechanical quantities in the deformation model to derive an appropriate electrical short circuit criterion. To create or enlarge such defects in the separator foils, corresponded higher mechanical stress fields are necessary, which are in many cases associated with local compression-states of the active materials (C, LiMOx) and are generated by external loads. It can be assumed that these local (mass-)densification are also describable in the homogenized constitutive law of the simplified model, since the predominant proportion of pouch cells consists of the active materials and thus significantly influences the properties of the homogenized material model.

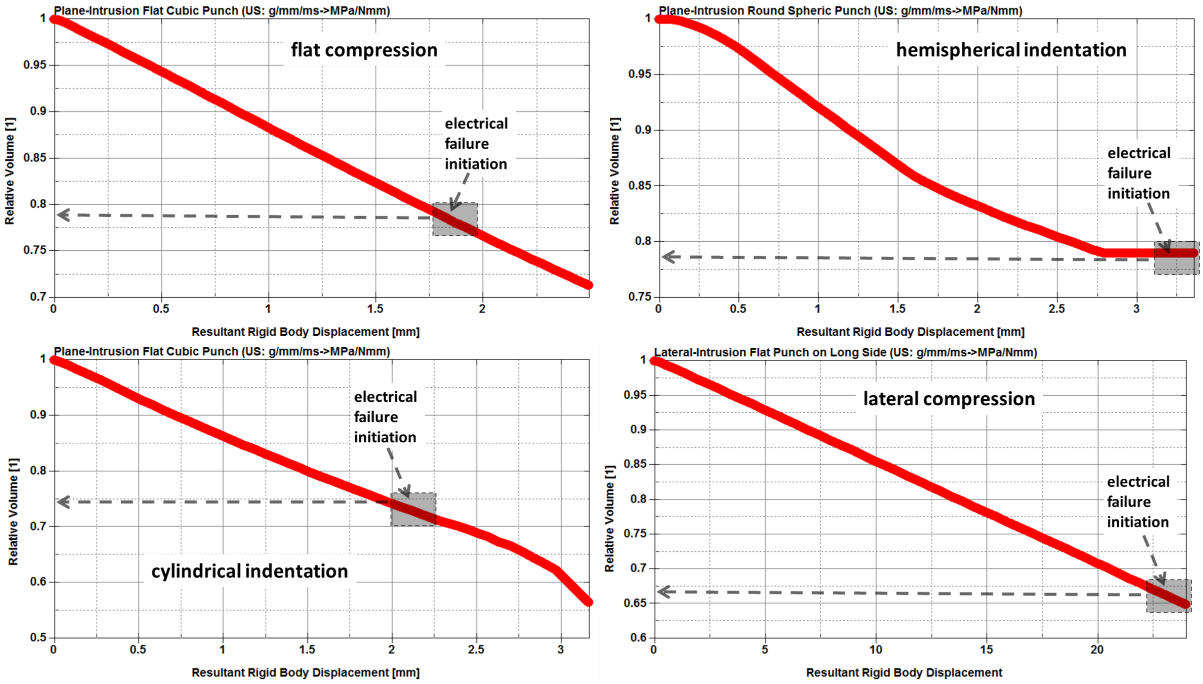

Fig.10: Comparison of (initial) electrical short circuits (grey rectangular) in relation to the evolution of relative volume (directly related to volumetric stain) for different load case scenarios

The rectangular regions with a gray background in Fig.10: indicate the electrical failure initiations which result from the experimental observations by evaluating the electrical potential in relation to the punch displacement [6, 8]. Local evaluations of the volumetric strains (or the directly related relative volumes) as a function of the punch displacements show a direction-dependent growth of this quantity, which probably leads to an anisotropic failure criterion for such an approach, with a significant difference between normal and lateral direction (see Fig.10:). In addition, Fig.10: shows that electrical failure occurs for all tests normal to the inner cell layered structure in a range between 0.75 and 0.78 of the relative volume, which indicates the validity of the use of volumetric strains (or relative volume) as a basis for an electrical short-circuit criterion. The electrical failure initiation in the lateral direction is related to the relative volume 0.66 which is significantly below the value in the normal direction (~0.77) and this result confirms the suggested use of an anisotropic failure model. The three-point bending test shows maximum relative volumes of 0.94 in the simulation and is therefore completely uncritical for electrical failure, which is also consistent with the experimental observations.
All evaluations in this section were done by using the `*MAT_MODIFIED_HONEYCOMB` model.

6 Summary

Due to the high application relevance of pouch cell modelling in automotive crash simulations, different analyzes for available simulation options were carried out to develop a simplified model. The focus was primarily on keeping the model as simple as possible to increase the numerical efficiency and the associated attractiveness in industrial applications. The focus was on the suitable choice of a homogenized material model that best approximates the highly repetitive intrinsic structure of Li-ion cells under the restricted simplified model requirements. Because the development for the modeling of electrical and mechanical failure also based significantly on the mechanical quantities of the deformation model, the choice of a suitable homogenized constitutive law is crucial. The results showed that an anisotropic compressible material model (**`*MAT_MODIFIED_HONEYCOMB`**), which describes the hardening parallel and normal to the intrinsic cell layer structure independently, can reproduce all different experimental results with sufficient accuracy.

Additional local comparative investigations were carried out to develop an electrical failure (short circuit) criterion with the aim to identify corresponding variables from the deformation model. For this approach, it was shown that the volumetric strains (or the directly correlated relative volume) probably represent a suitable variable for the implementation of a criterion for electrical failure. Complementary numerical and experimental investigations with a jellyroll cell type [9] have shown partial alternative results. The discrepancies are probably explainable in the different intrinsic cell structure of a jelly-rolled [9] and a pure stacked cell type which is used in this work.

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