AN ANISOTROPIC CONSTITUTIVE APPROACH FOR ANALOGOUS MODELING OF LI-ION CELLS

Modelling, Simulations and Experimental Investigations

Fraunhofer IWM

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AGENDA

- Motivation and introduction
- Continuum model for Li-ion-cell
- Setup of analogous model for Li-ion-cell
- Investigated representative crash loading scenarios
- Parameter identification
- Deformation behavior for different loadings
  (compression normal and lateral, indentation, bending)
- Summary

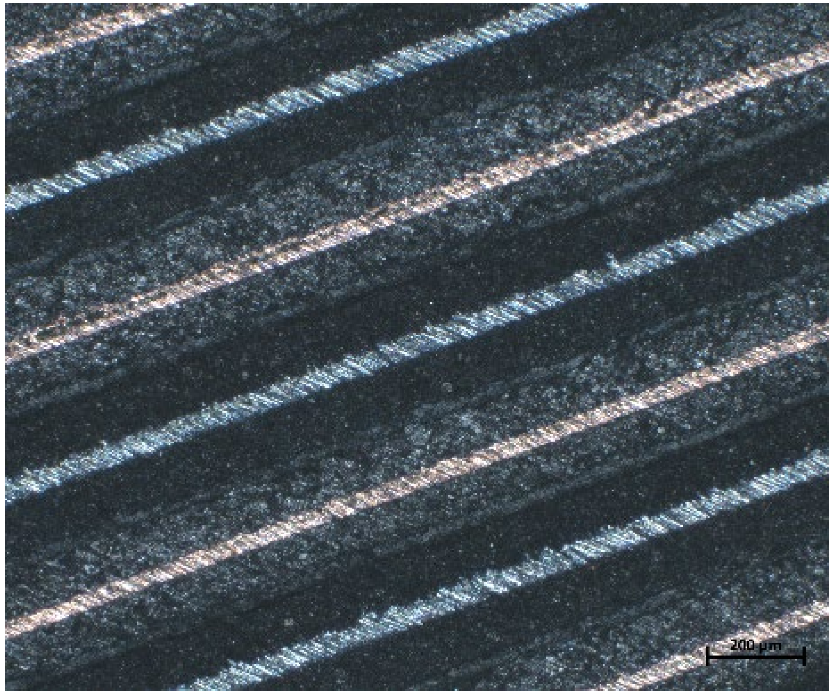
Motivation: pouch cell, principal structural setup, tasks and goals

Principal structural setup:

- With active material (graphite C and lithium metal oxide / LiMOx) coated thin metallic electrode foils (copper / Cu and aluminum / Al)
- Polymeric separator for anodic-cathodic separation
- Outer flexible foil (coffee bag) for enclosing inner layered structure

Typical pouch cell with flexible enclosing outer foil (coffee bag)

Cut and specimen preparation

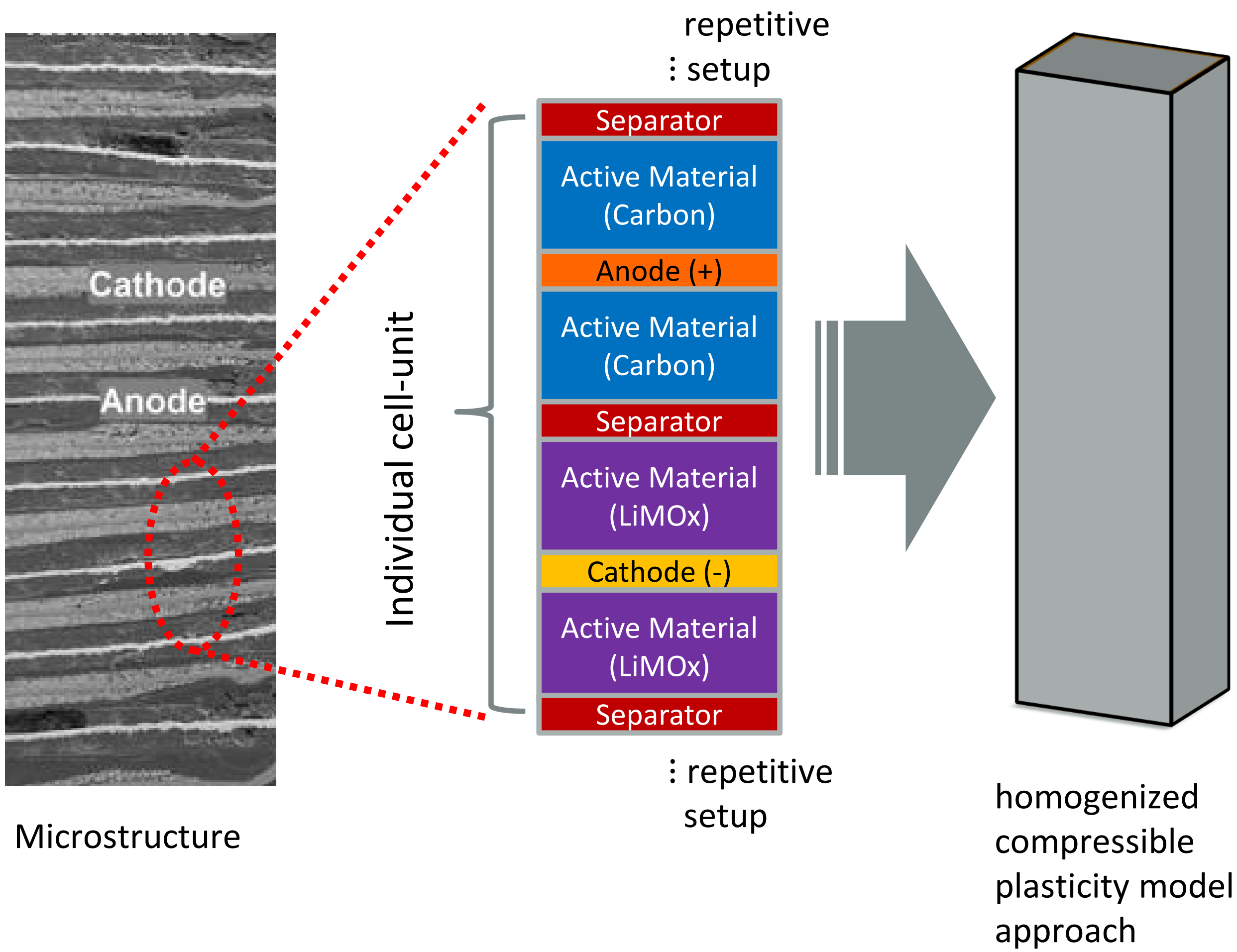


Microstructure (light microscopy, Fraunhofer EMI)

Main tasks:

- Repetitive complex inner structure (few hundred layers) are not in detail representable for efficient models
- Development of efficient simplified analogous models to simulate deformation (short-circuit behavior) for crash

Introduction: Real intrinsic layered setup and model approach



Repetitive 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%
- Active materials are compressible granular materials which suspended in electrolyte

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Introduction: continuum model approach

- Main topics for alternative model development
  - less complex anisotropic model approach (from experimental investigations)
  - physical motivated from inner layered cell-structure:
    transversal isotropic behavior, compressibility, ...
  - using proven conventional plasticity model framework/approach
    (no completely uncoupled directional behavior which could lead to artificial effects, …)
  - model approach should be simple and easy to calibrate

- Main topics of new developed homogenized model approach
  - modified Deshpande Fleck model [1] with definition from a weighted hydrostatic stress [2]
  - associated flow rule
  - strain hardening depending on deviatoric and hydrostatic strain

[1] : Deshpande V.S., Fleck N.A., (2000) Isotropic constitutive models for metallic foams. J Mech and Physics of Solids

[2] : Benzerga A.A, Besson J., (2001) Plastic potentials for anisotropic porous solids. Euro. J Mech

Continuum model for Li-ion-cell

- weighted hydrostatic stress σ_h

$$\sigma_h = h\sigma_{11} + h\sigma_{22} + (1-2h)h\sigma_{33} = \sigma : \mathbf{J}$$

$$\mathbf{J} = \begin{bmatrix} h & & \\ & h & \\ & & 1-2h \end{bmatrix} \qquad h \in [0, 1]$$

h : material parameter for anisotropy (h =1/3: isotropic)

3: thickness direction
- $h \in [0, 1/3]$ direction 3 more compressible (relevant for battery)
- $h \in [1/3, 1]$ directions 1 and 2 more compressible

[2] : Benzerga A.A, Besson J., (2001) Plastic potentials for anisotropic porous solids. Euro. J Mech

- Asymmetrical yield function in (σ_e, σ_h) space. Elliptical form in compression, von Mises line in tension

$$\varphi = \tilde{\sigma} - B \leq 0$$

$$\tilde{\sigma} = \sqrt{\sigma_e^2 + \alpha^2 \sigma_h^2} \qquad \alpha = 0 \text{ for } \sigma_h > 0$$

$\sigma_e = \sqrt{\frac{3}{2} S_{ij} S_{ij}}$: von Mises effective stress

S_{ij} : deviatoric stress

B : yield stress

α : parameter for the shape of the yield surface

[1] : Deshpande V.S., Fleck N.A., (2000) Isotropic constitutive models for metallic foams. J Mech and Physics of Solids

Continuum model for Li-ion-cell

- Normality rule (associated flow)

$$\dot{\boldsymbol{\epsilon}}^{\mathrm{p}} = \dot{\lambda}\,\frac{\partial\,\varphi}{\partial\,\boldsymbol{\sigma}}$$

$\dot{\boldsymbol{\epsilon}}^{\mathrm{p}}$ plastic strain rate

$\dot{\lambda}$ plastic multiplier determined from the consistency relation $\dot{\varphi} = 0$

$$\dot{\boldsymbol{\epsilon}}^{\mathrm{p}} = \dot{\boldsymbol{\epsilon}}^{\mathrm{D}} + \dot{\boldsymbol{\epsilon}}^{\mathrm{V}}$$

plastic strain rate can de splitted into a deviatoric and a volumetric portion

$$\dot{\varepsilon}_{\mathrm{D}} = \sqrt{\frac{3}{2}\dot{\boldsymbol{\epsilon}}^{\mathrm{D}} : \dot{\boldsymbol{\epsilon}}^{\mathrm{D}}}$$

deviatoric von Mises effective plastic strain rate

$$\dot{\varepsilon}_{\mathrm{V}} = \dot{\boldsymbol{\epsilon}}^{\mathrm{V}} : \mathbf{1}$$

volumetric plastic strain rate

- Hardening rule depending on deviatoric and volumetric plastic strain

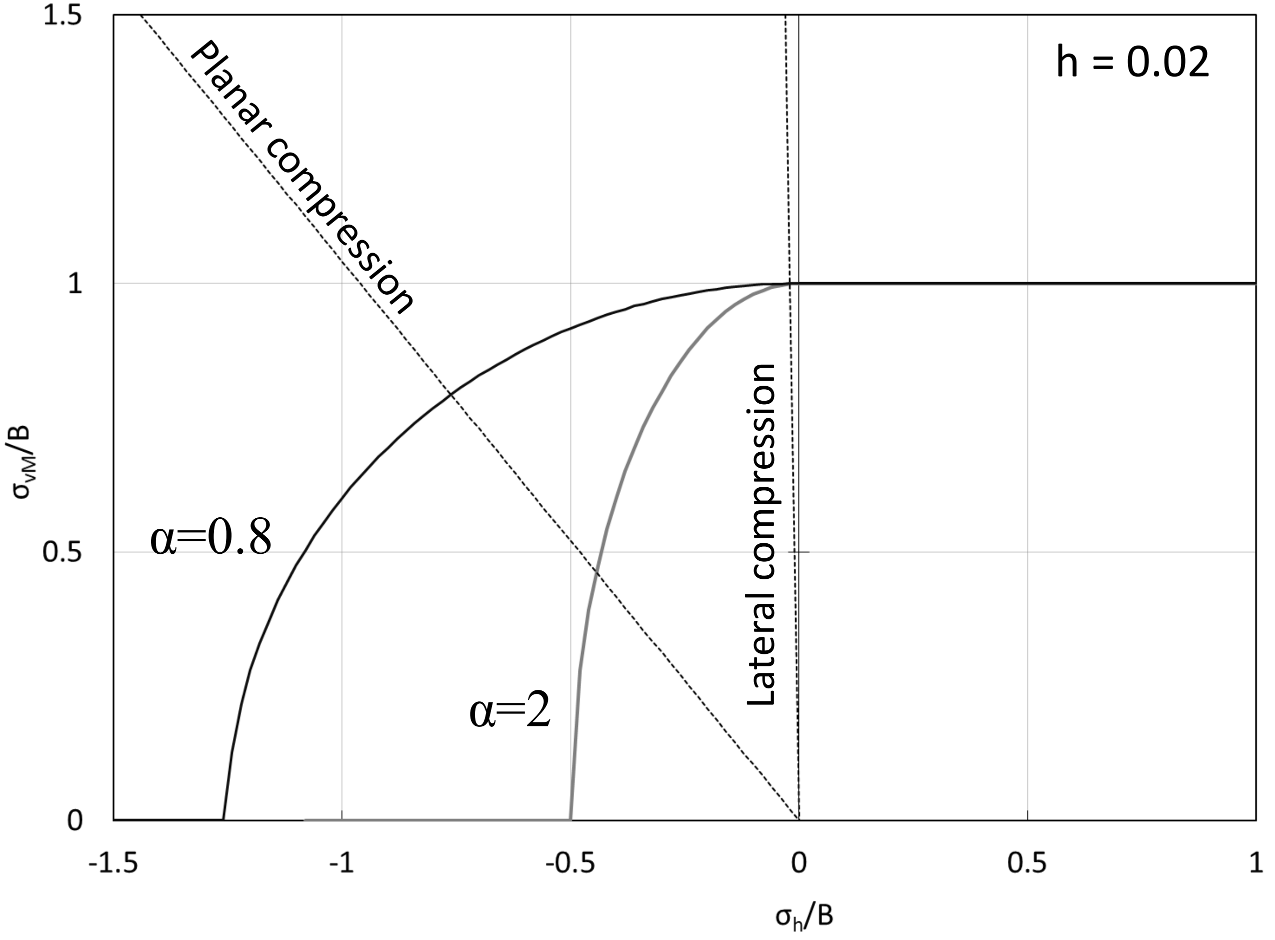
$B = B_D(\varepsilon_D) + B_V(\varepsilon_V)$

exponential deviatoric hardening: $B_D = Y_{0D} + A_D\,(1\text{-}\exp(\text{-}B_D\varepsilon_D))$

polynomial volumetric hardening: $B_V = Y_{0V} + A_V\varepsilon_v^{n_V}$

Y_{0D}, A_D, B_D, Y_{0V}, A_V, n_V: material parameters for hardening

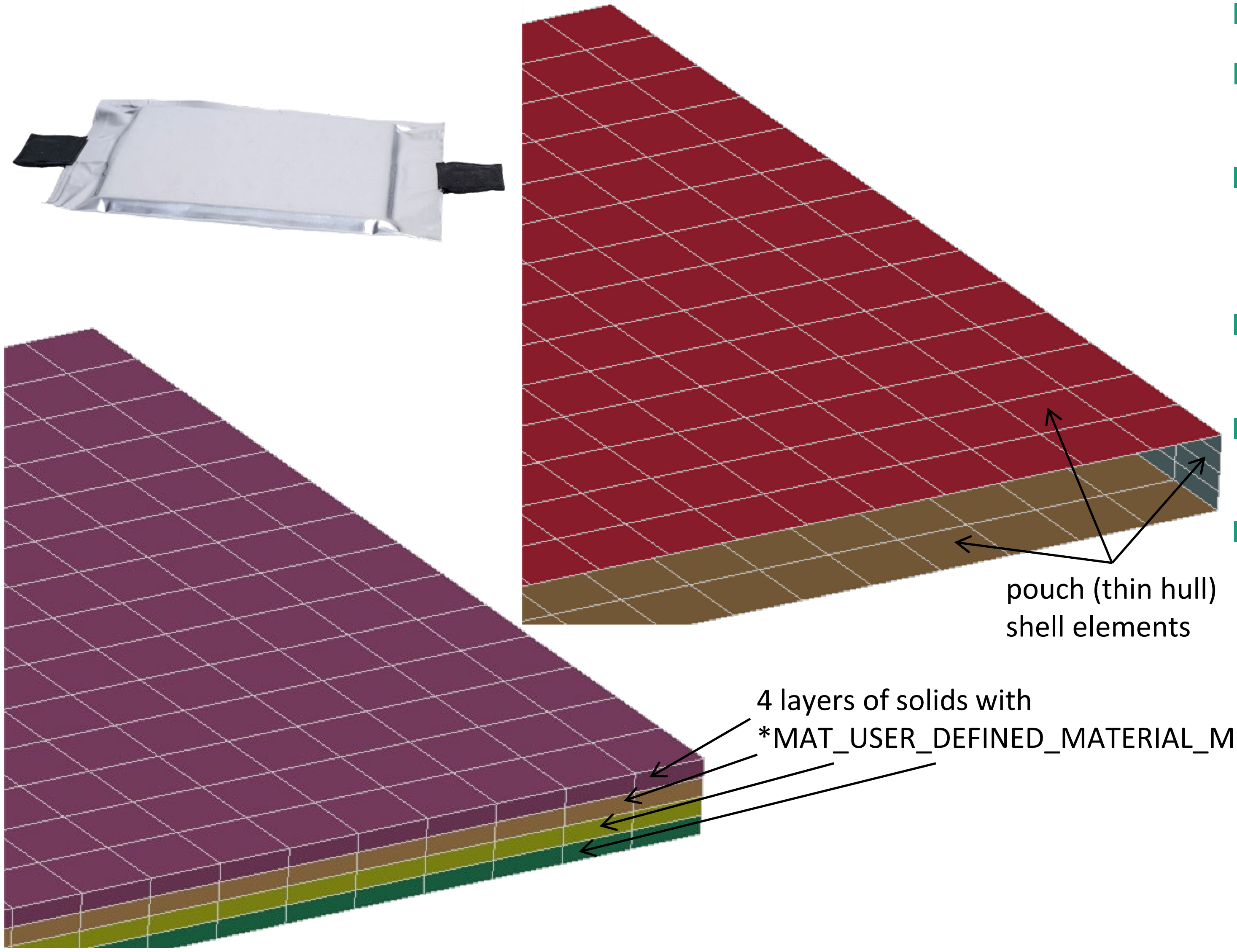
Continuum model for Li-ion-cell



Remarks:

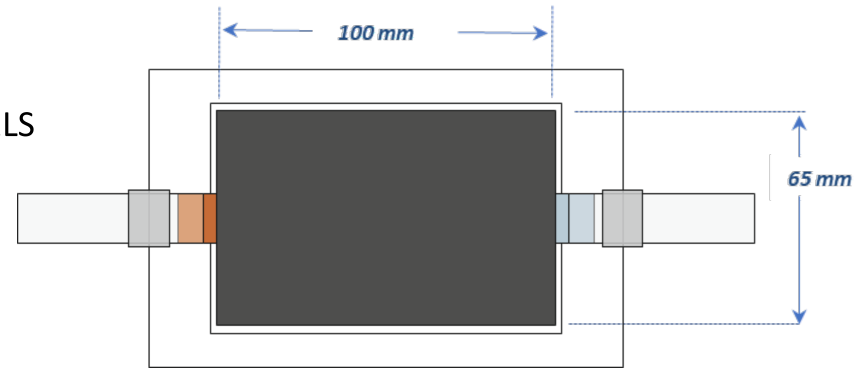
- Under compression the model shows stress state dependent yielding (like model of Fleck-type). The compressibility is due to the compressible active materials, which are the major component in a Li-ion-cell
- Under tension loadings the model behaves like a classical von Mises model (no volume variation)

Setup of the simplified analogous model



Characteristics of the model setup:

- Thickness of pouch cell ~7mm
- Under-integrated 8-node solid elements (HG 6) for inner layered structure
- Under integrated 4-node shell elements (HG 2) for pouch (bag), which uses same nodes as outer solids (no contact)
- Discretization ~5mm x ~5mm x ~1.75mm (overall: 1120 solid & 832 shell elements)
- *MAT_USER_DEFINED_MATERIAL_MODELS for homogenized inner materials
- *MAT_PIECEWISE_LINEAR_PLASTICITY for outer pouch (bag)



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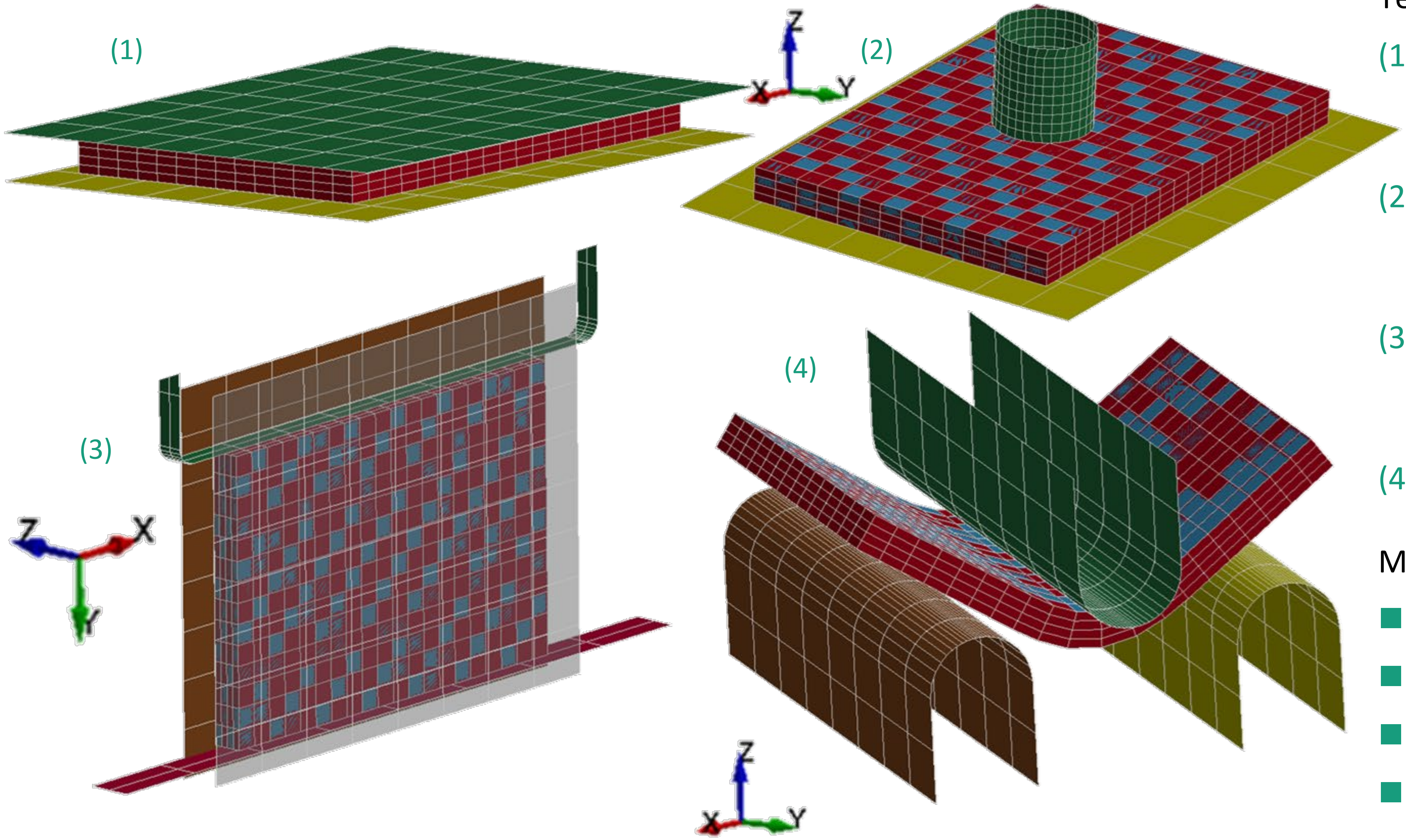
Investigatedrepresentative crash loading scenarios



Test setups:

(1) Compression test in thickness direction (loading normal to inner layered structure)

(2) Indentation test in thickness with flat and hemispherical heads of cylindrical indenters

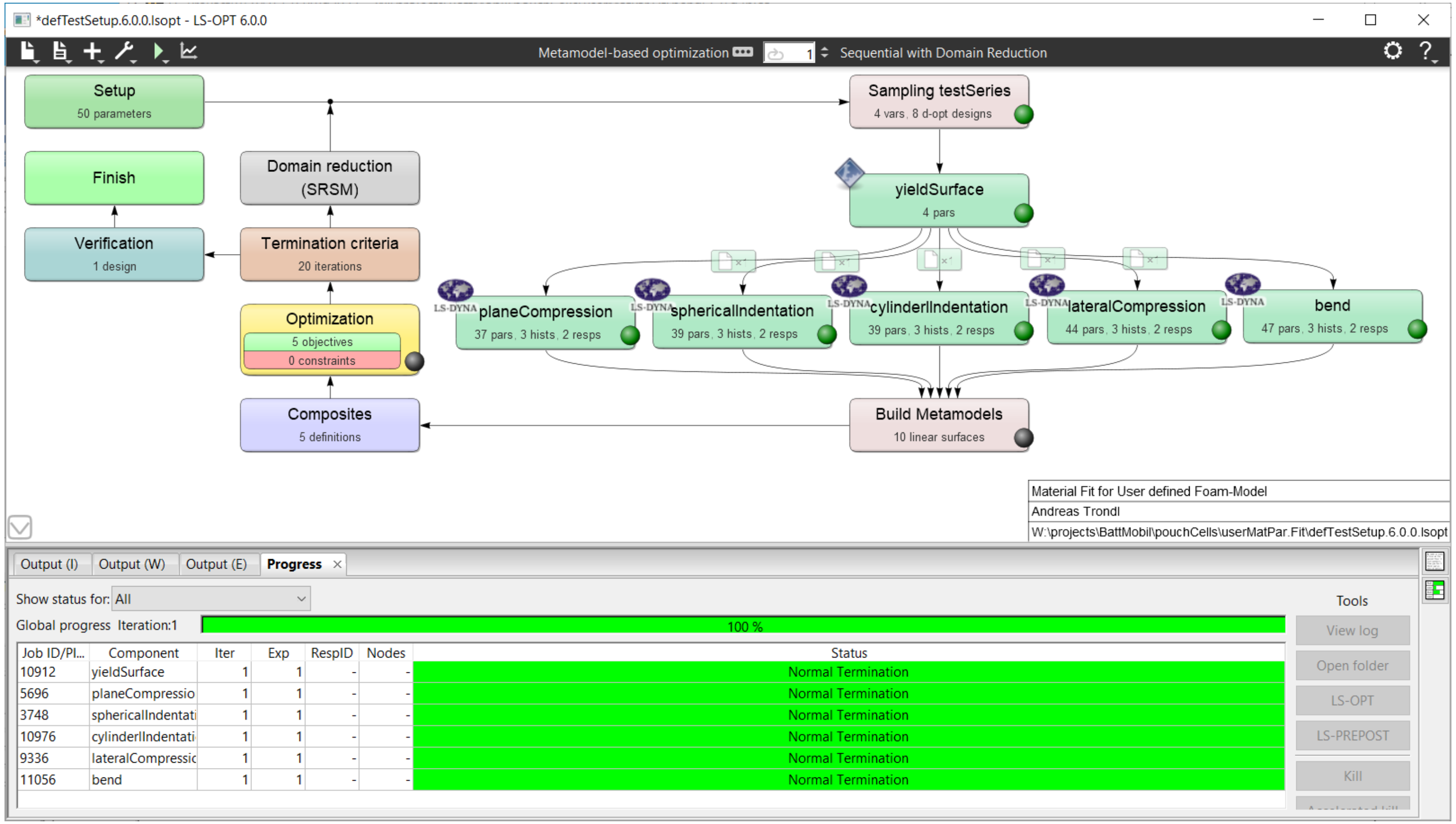
(3) Lateral directed compression test (loading parallel to inner layer structure)

(4) 3-point bending test

Model setup:

- Punch speed 0.1mm/s
- Tool-pouch friction 0.08
- Rigid body tools
- Tool discretization 1-2mm up to 6mm

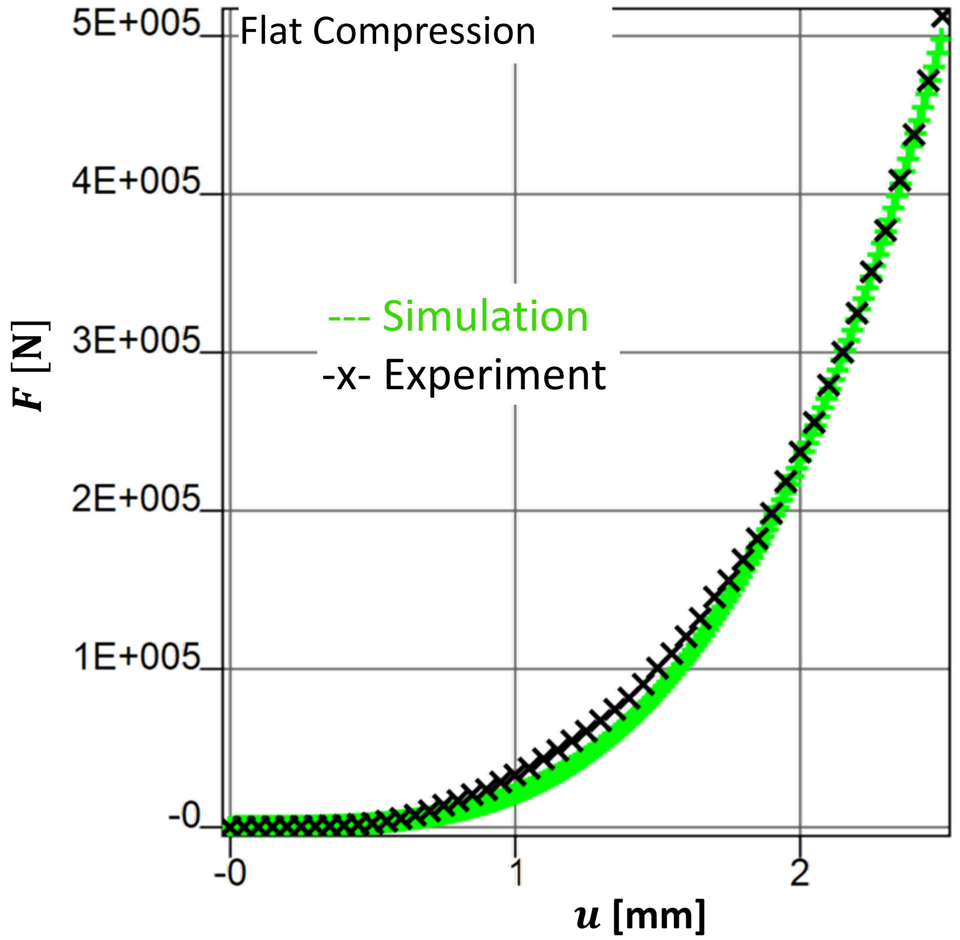
Deformation behavior of the material model, parameter identification

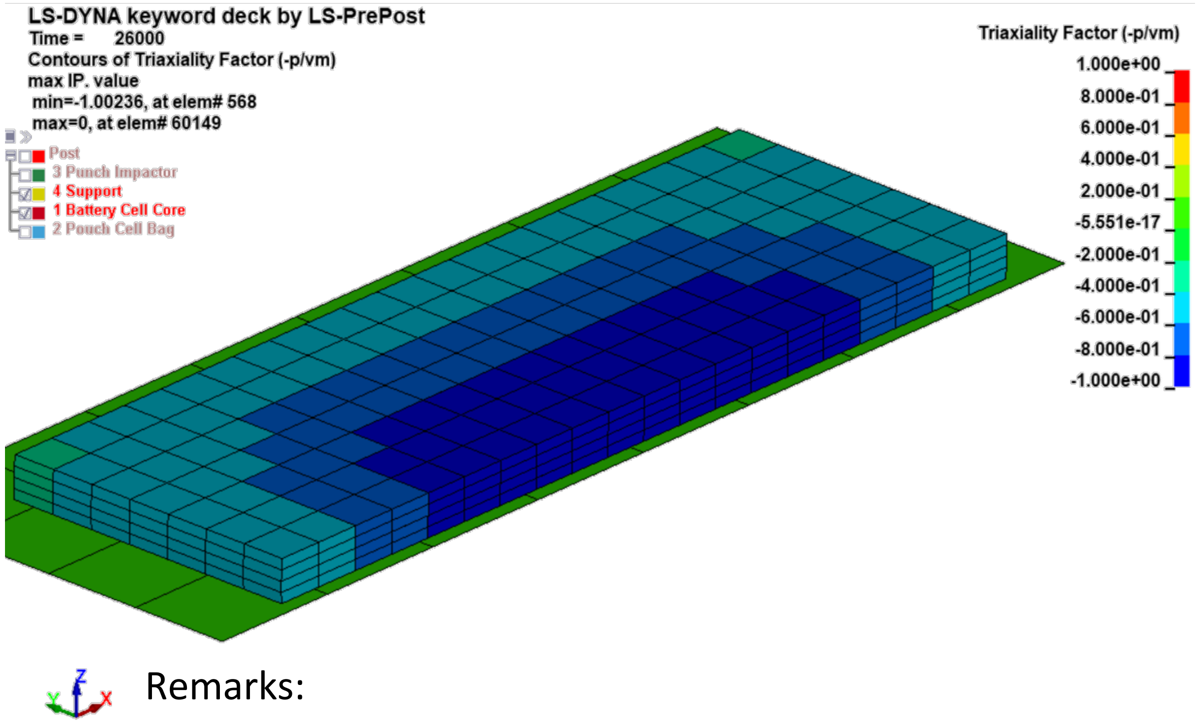


Setup for parameter-fit:

- Using of LS-Opt
- Objective optimization goal are the global characteristic of force-displacement curve(s)
- Simultaneous parameter-fit in an average sense for all tests (weight factors: flat compression 2, other tests 1)
- Only volumetric part for hardening is used
- Free parameters: h, α, A_V, n_V; (Y_{0V} small constant value)

Compression test in thickness direction

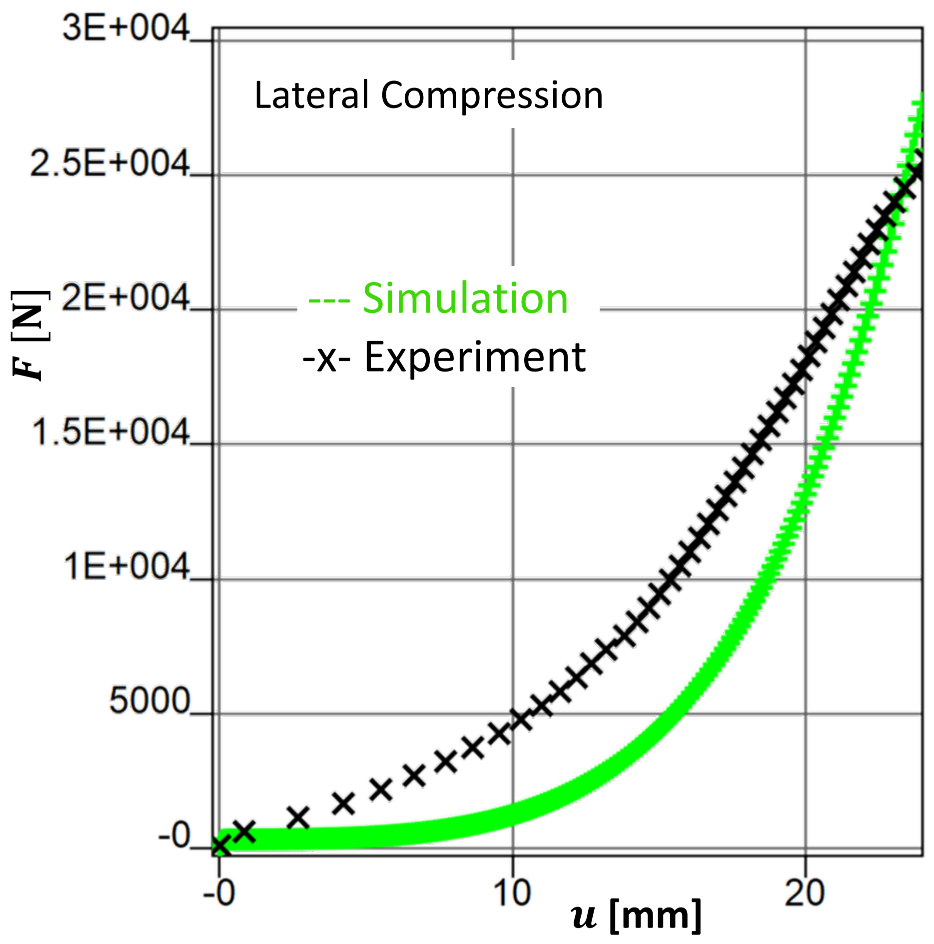


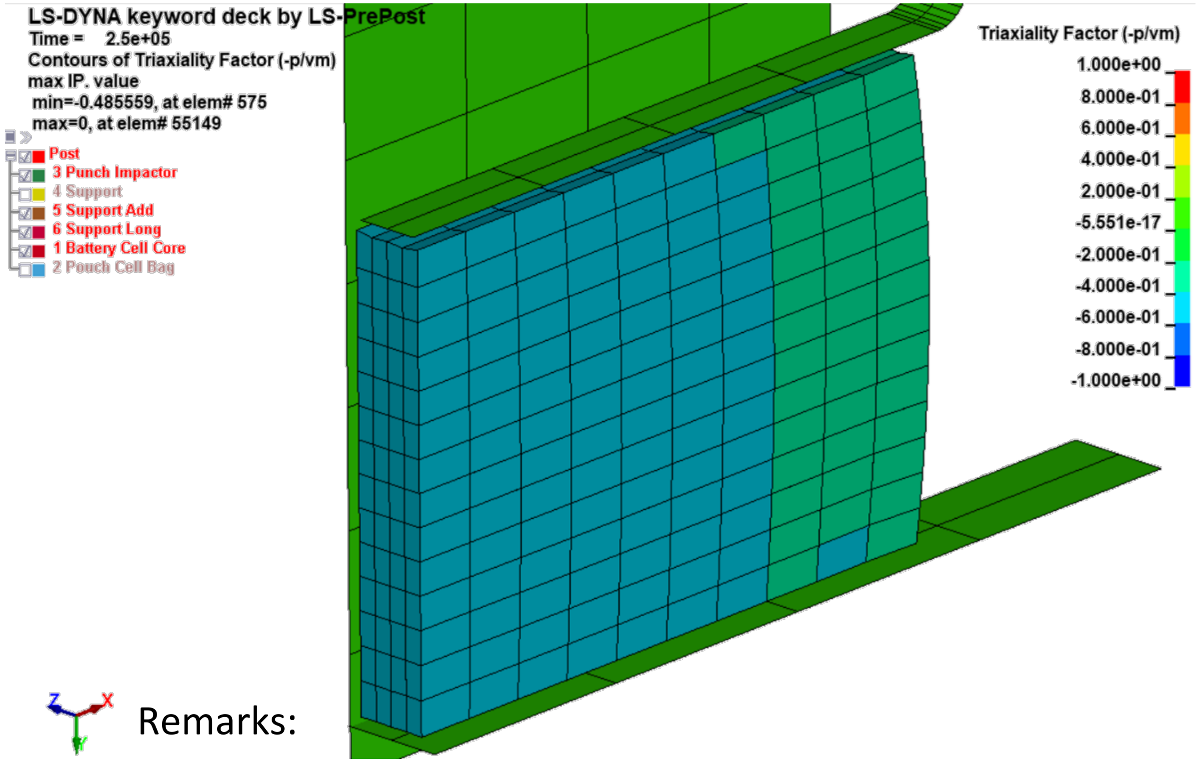


Remarks:

- Value of stress-triaxiality nearly -1 in center
- Stress-triaxiality in center region and outer region nearly constant
- No variation of stress-triaxiality in thickness
- Global F-u characteristic fits well to experiments

Lateral compression test





Remarks:

- Constant stress-triaxiality of -0.5 in cell-center
- Smaller stress-triaxiality (~-0.3) in outer regions of cell
- No variation of stress-triaxiality in thickness
- Moderate deviations in global F-u characteristic to experiments (softer at beginning, stiffer at ending)

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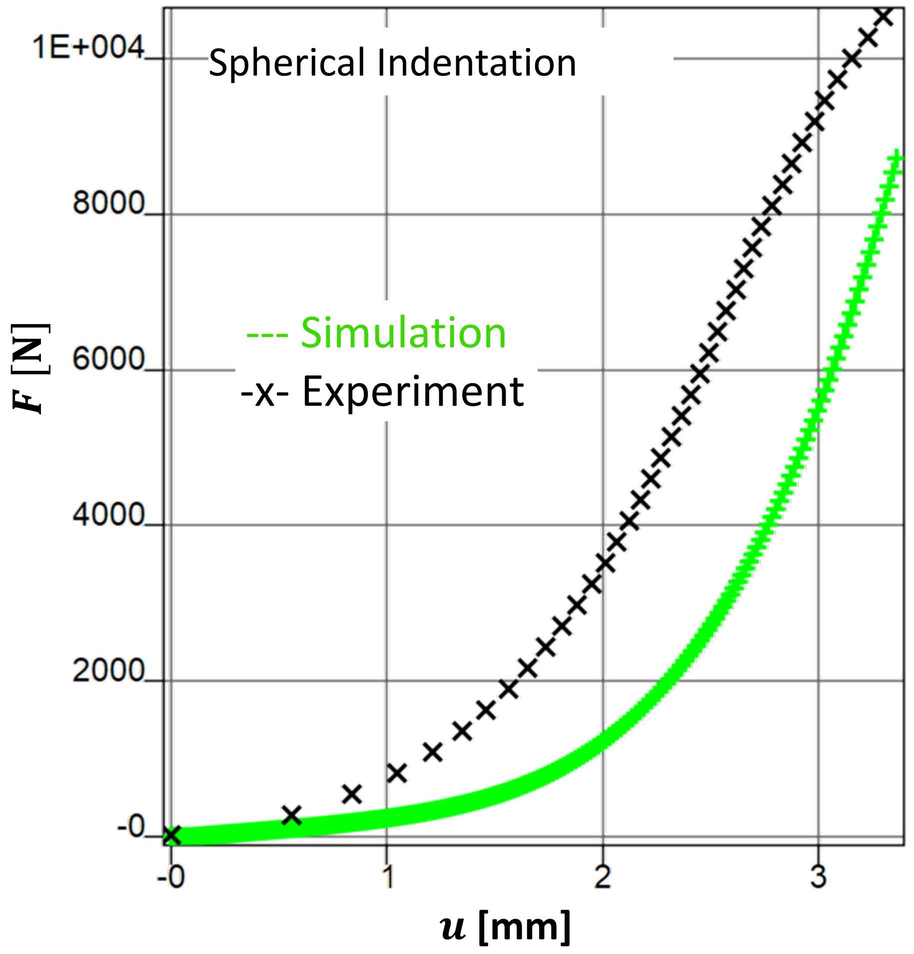
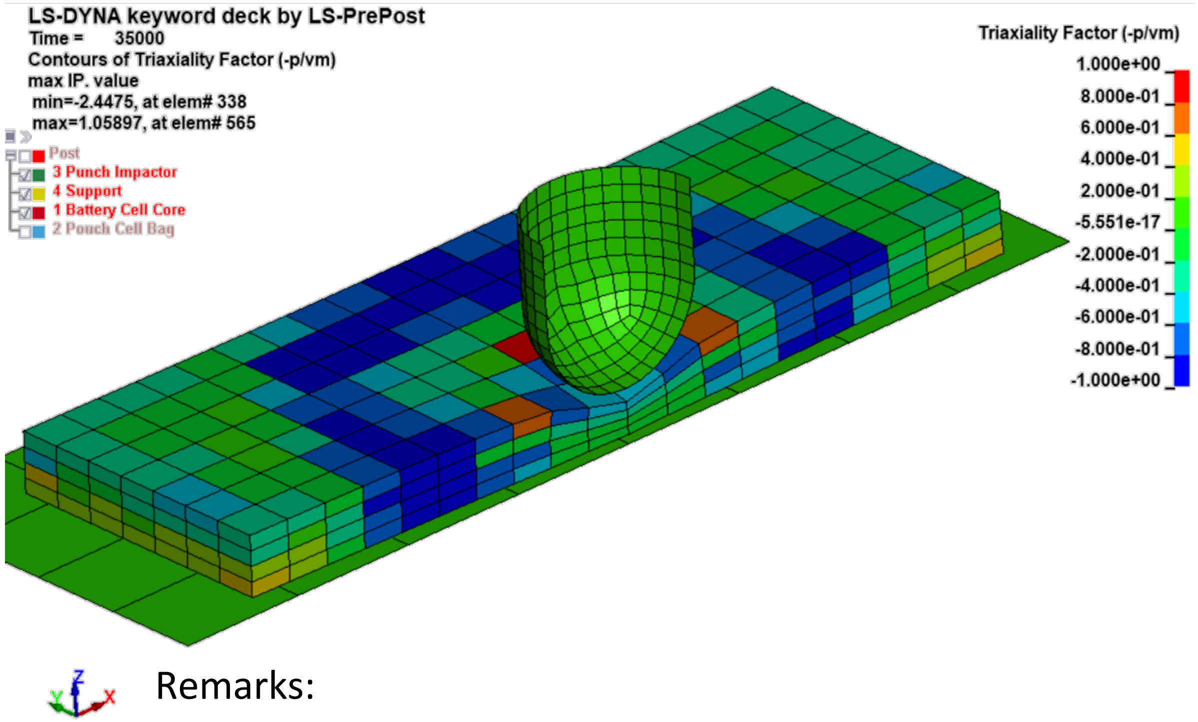
Indentation test with hemispherical indenter

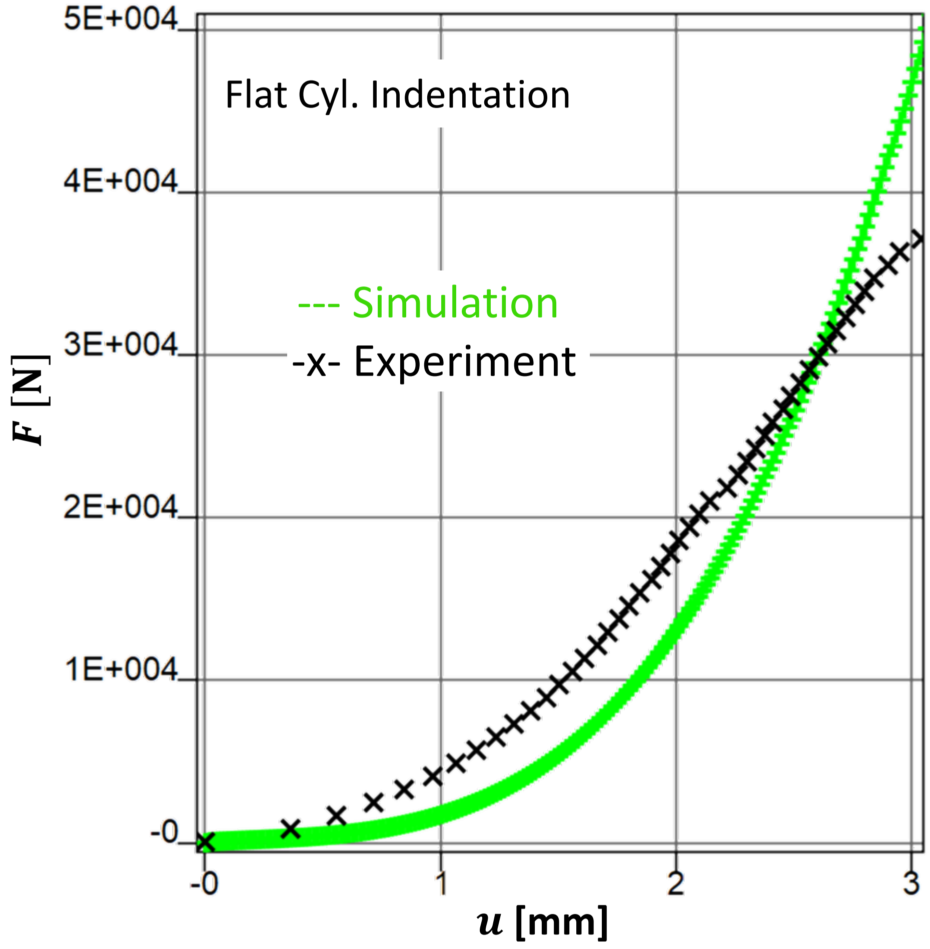


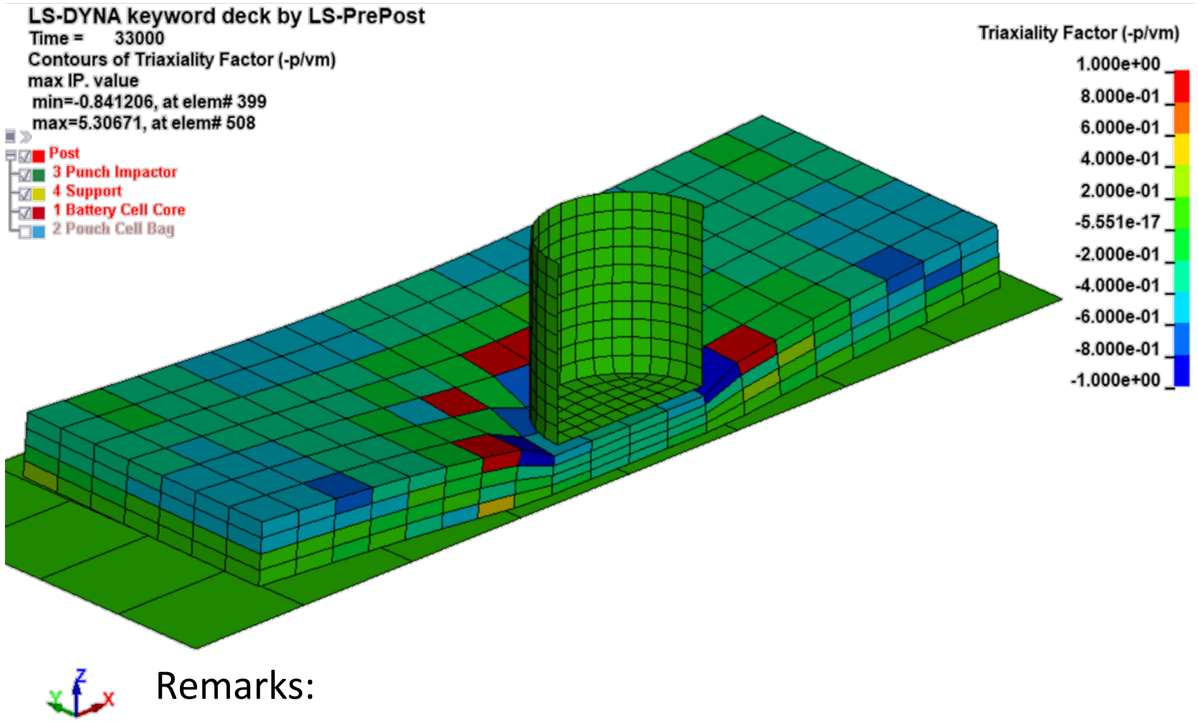


Remarks:

- Value of stress-triaxiality of -0.6 near contact zone
- Smaller stress-triaxiality (~-0.3) in lower regions influenced of indenter
- Stronger variation of stress-triaxality in thickness
- Global F-u characteristic softer in relation to experiments

Indentation test with flat indenter

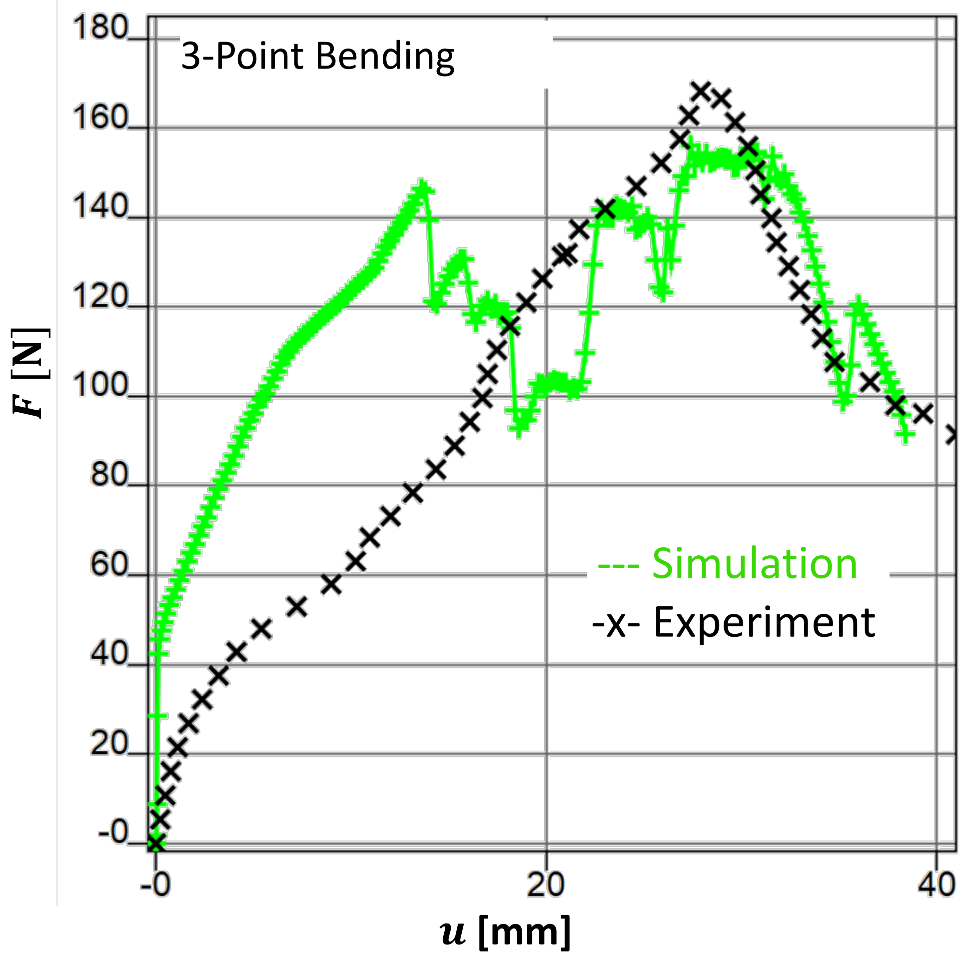


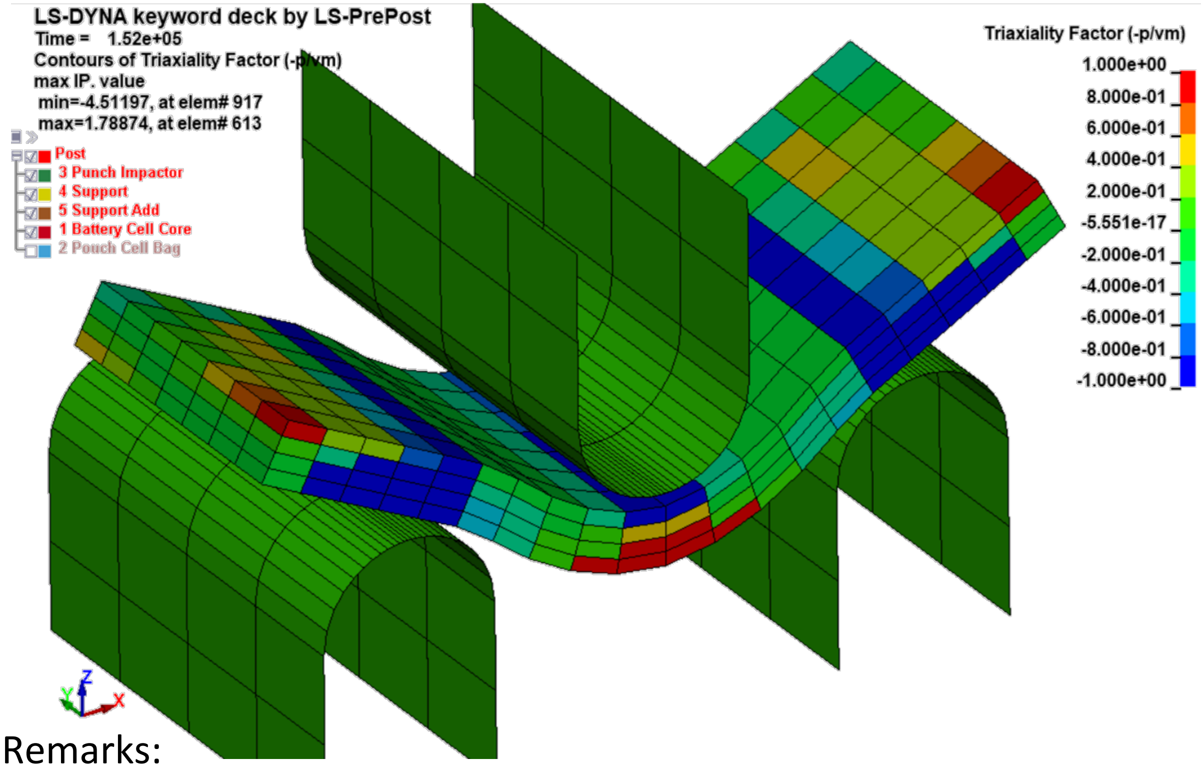


Remarks:

- Value of stress-triaxiality of -0.4 near contact zone in
- Relative constant stress-triaxiality in lower and contact region of indenter
- Less variation of stress-triaxiality in thickness
- Global F-u characteristic fits well to experiments

3-point bending test





Remarks:

- Value of stress-triaxiality of -1 (also lower) near contact zones in
- Due to bending conditions strong variation of stress-triaxiality in thickness in middle of cell
- Global F-u characteristic: stiffness too high for very low u, stiffness similar for medium u, upper force level fits okay

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Summary

- Presentation of simple and performant analogous model for Li-ion cell
- Suggestion of a (new) simplified homogenized constitutive continuum approach to model anisotropic behavior for Li-ion cells
- Suggestion for simplified analogous cell model by using a homogenized core with solid and an outer hull with shell elements (approach should work for pouch, but also for prismatic or cylindric cells)
- Model calibration based on experiments for representative crash scenarios like: compression normal and parallel to the inner layered structure, indentations normal to layers and 3-point bending test
- Models were calibrated in relation to all force-displacement curves for all load cases simultaneously
- The suggested anisotropic model shows over all load-cases good agreements to the experiments (especially the in- and out-of-plane loadings can be controlled by the weight parameter h)
- The moderate smaller deviations (spherical indentation) explainable due to the coarse discretization
- …
- Next steps: better calibration using deviatoric part; further model improvement motivate by the intrinsic structural layered cell setup; implementation of electrical short circuit criterion based on intrinsic mechanical quantities, …