Analysis of the fracture behavior of active materials for lithium-ion batteries

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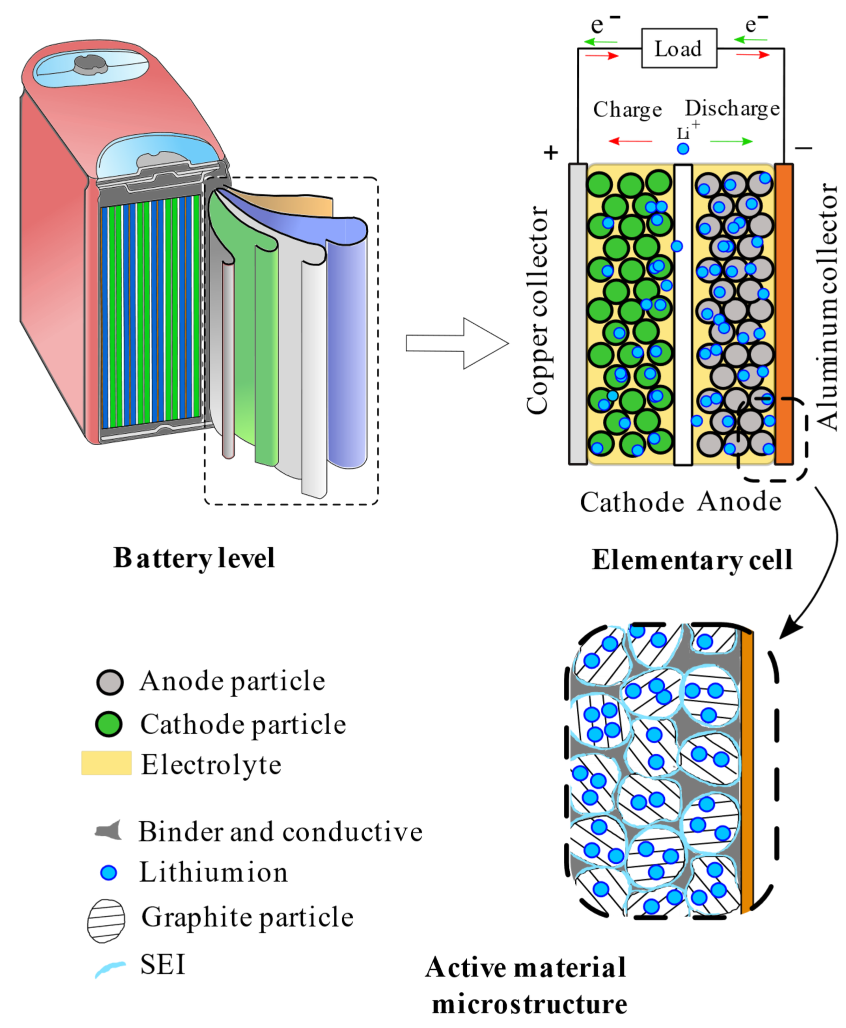
Introduction

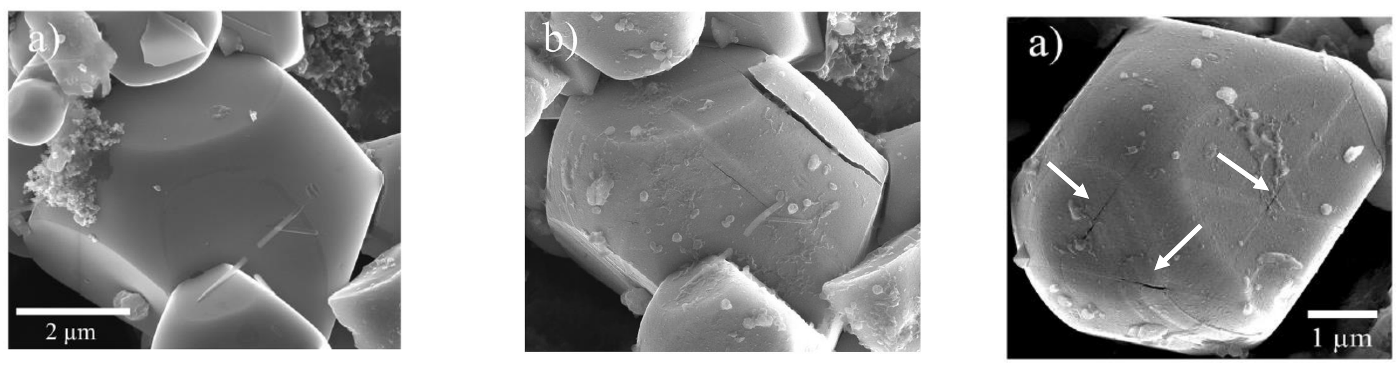
Lithium-ion batteries are currently the most widespread energy storage system with a wide field of applications. Nowadays, their durability is a crucial topic among the research community, since batteries performances, such as capacity and power, decrease during life cycle.

It is highlighted that performances decrease is linked to a progressive damage of the micro-structure of active materials, caused by their interaction with lithium ions. Indeed, lithium ions are inserted and extracted in the host structure of active material during battery operation. At micro-structure level, these processes result in the diffusion of lithium ions in the particles which make up the active material. The consequently uneven distribution of lithium ions in the particles causes the rise of mechanical stress, which ultimately results in damage and fracture propagation in the active material microstructure.

Crack propagation is studied with a finite element model in Ansys APDL to predict the influence of micro-structure geometry and current delivered by the battery on fracture and damage. Active material micro-structure of LMO is simulated as spherical particles with micrometric radius.



SEM images of active material micro-structure by D. Chen, "Chemomechanical fatigue of LiMn1.95Al0.05O4 electrodes for lithium-ion batteries», *Electrochimica Acta*, 2018



Methods

Analytical Model

- Diffusive Model

$$\frac{\partial C}{\partial t} = \nabla \cdot (D \nabla C)$$

$$\begin{cases} C(r,0) = C_0, & \text{for } 0 \le r \le R \\ \left.\frac{\partial C(r,t)}{\partial r}\right|_{r=R} = \frac{I}{FD}, & \text{for } t \ge 0 \\ \left.\frac{\partial C(r,t)}{\partial r}\right|_{r=0} = 0, & \text{for } t \ge 0 \end{cases}$$

- Concentration

$$c(r,t) = c_0 + \frac{IR}{F_a D}\left[3\tau + \frac{1}{2}\left(\frac{r}{R}\right)^2 - \frac{3}{10} - 2\frac{R}{r}\sum_{n=1}^{\infty}\left(\frac{sin(\lambda_n r/R)}{\lambda_n^2 sin(\lambda_n)}e^{-\lambda_n^2 \tau}\right)\right]$$

- Mechanical model

Equilibrium equations

$$\frac{d\sigma_r}{dr} + \frac{2}{r}\left(\sigma_r - \sigma_c\right) = 0$$

Constitutive equations

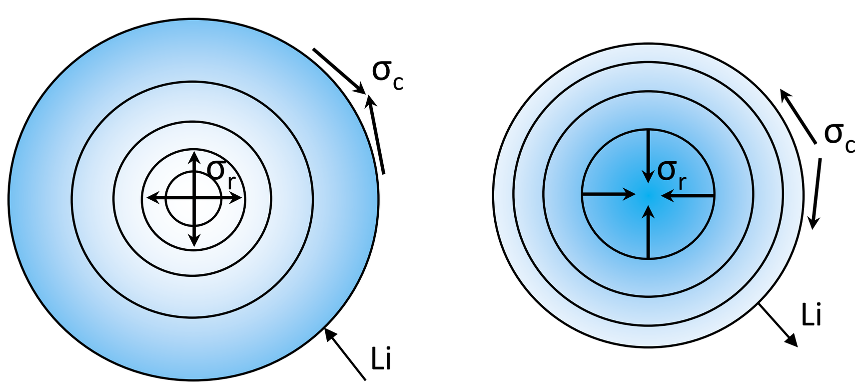
$$\begin{cases} \sigma_r = \frac{E\left[\left(\varepsilon_r - \frac{\Omega c}{3}\right)(1-\nu) + 2\nu\left(\varepsilon_c - \frac{\Omega c}{3}\right)\right]}{(1+\nu)(1-2\nu)} \\ \sigma_c = \frac{E\left[\left(\varepsilon_c - \frac{\Omega c}{3}\right) + \nu\left(\varepsilon_r - \frac{\Omega c}{3}\right)\right]}{(1+\nu)(1-2\nu)} \end{cases}$$

Congruence equations

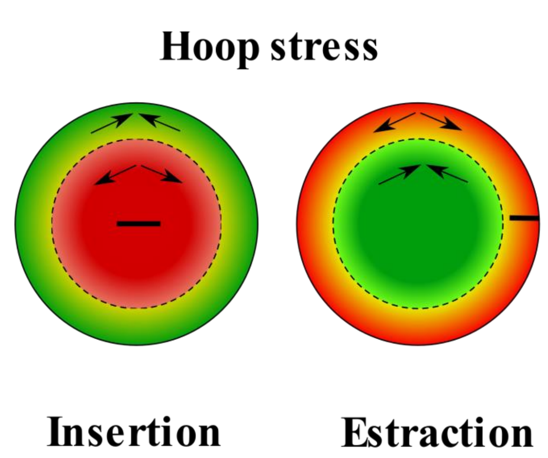
$$\begin{cases} \varepsilon_r = \frac{du}{dr} \\ \varepsilon_c = \frac{u}{r} \end{cases}$$

- Stress

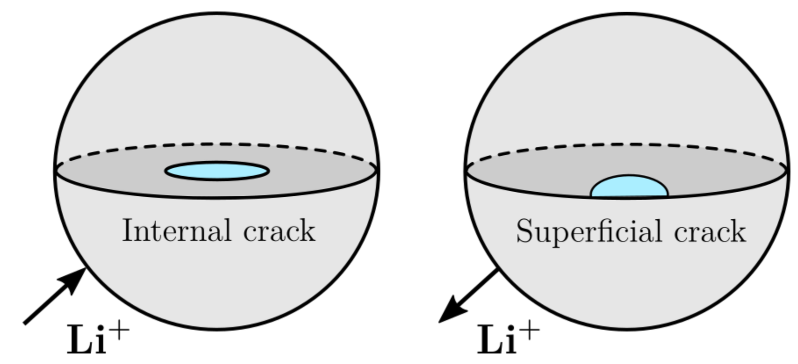
$$\begin{cases} \sigma_r(r) = \frac{2\Omega}{3}\frac{E}{1-\nu}\left[\frac{1}{R^3}\int_0^R c(r)r^2\,dr - \frac{1}{r^3}\int_0^r c(r)r^2\,dr\right] \\ \sigma_c(r) = \frac{\Omega}{3}\frac{E}{1-\nu}\left[\frac{2}{R^3}\int_0^R c(r)r^2\,dr + \frac{1}{r^3}\int_0^r c(r)r^2\,dr - c(r)\right] \end{cases}$$



Simulation procedure



Cracks propagate when hoop stress is tensile, then **superficial cracks** propagate during **extraction** and **internal cracks** propagate during **insertion**.

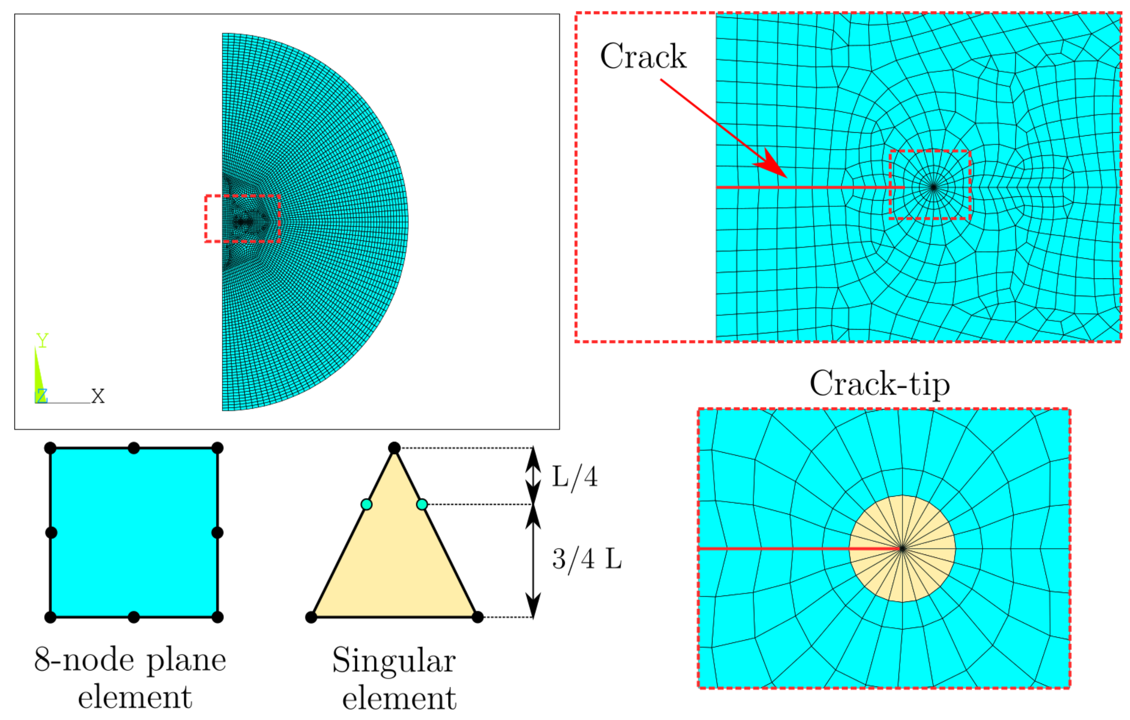


- The **multiphysics mechanical-diffusive** problem is reformulated with an **equivalent mechanical-thermal** problem:
  - The lithium **concentration** distribution is computed with the analytical model
  - The **equivalent temperature** is mapped on the FE model nodes

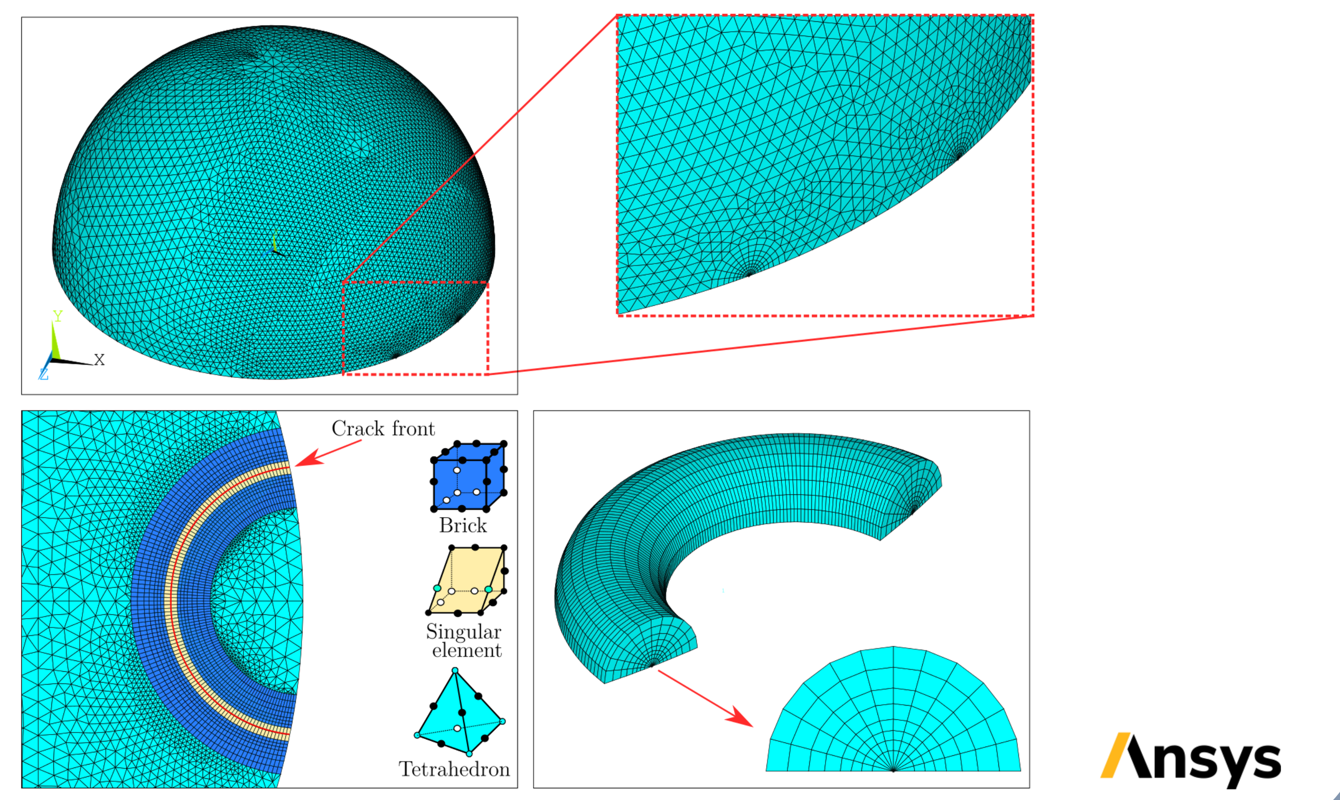
$$T = T_{ref} + \frac{\Omega}{3\alpha}\left(C - C_{ref}\right)$$

FEM Model

- **Particle with central crack**: 2D Model with PLANE183 elements

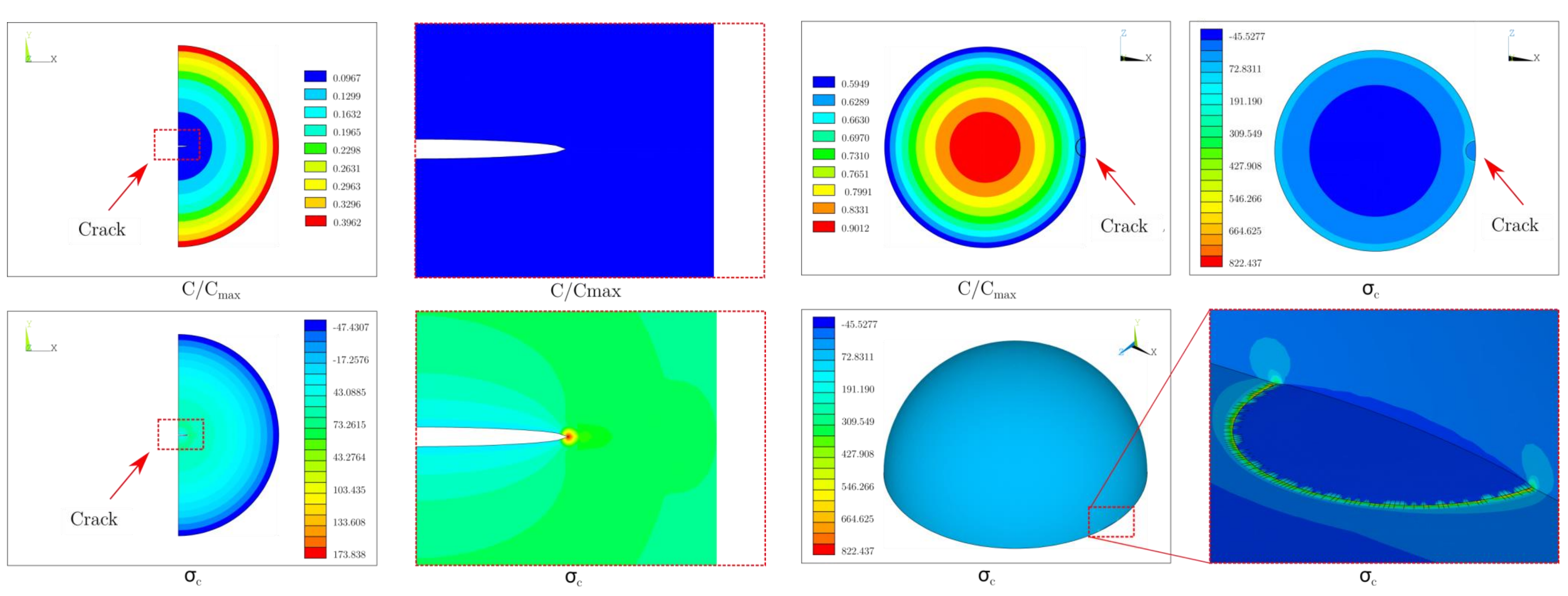


- **Particle with elliptical superficial crack**: 3D Model with SOLID186 elements



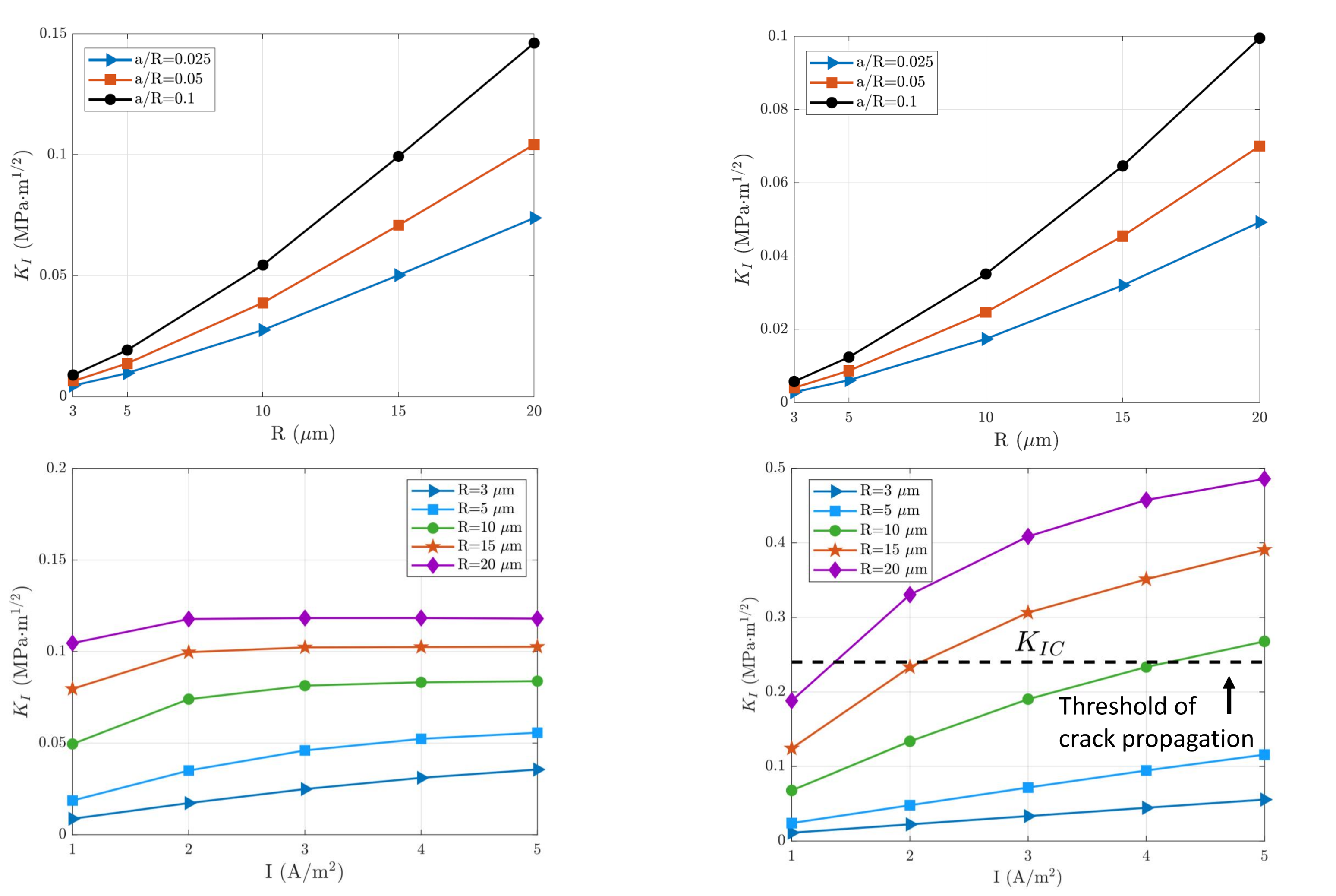
Results

Normalized concentration and Hoop Stress [MPa]



Central crack – Galvanostatic Insertion | Superficial crack – Galvanostatic Extraction

Stress Intensity Factor KI as a function of particle radius and delivered current



Central crack – Galvanostatic Insertion | Superficial crack – Galvanostatic Extraction

References

- Davide Clerici, Francesco Mocera, and Aurelio Somà. "Analytical Solution for Coupled Diffusion Induced Stress Model for Lithium-Ion Battery." *Energies* 13.7 (2020): 1717.
- Davide Clerici and Francesco Mocera. "Micro-scale modeling of Lithium-ion battery." *IOP Conference Series: Materials Science and Engineering*. (2021) Vol. 1038. No. 1. IOP Publishing.
- *D. Chen, "Chemomechanical fatigue of LiMn1.95Al0.05O4 electrodes for lithium-ion batteries», Electrochimica Acta, 2018*