# Finite element analysis of diffusion-induced stresses and phase segregation in Li-ion battery electrode

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## 1 Motivation

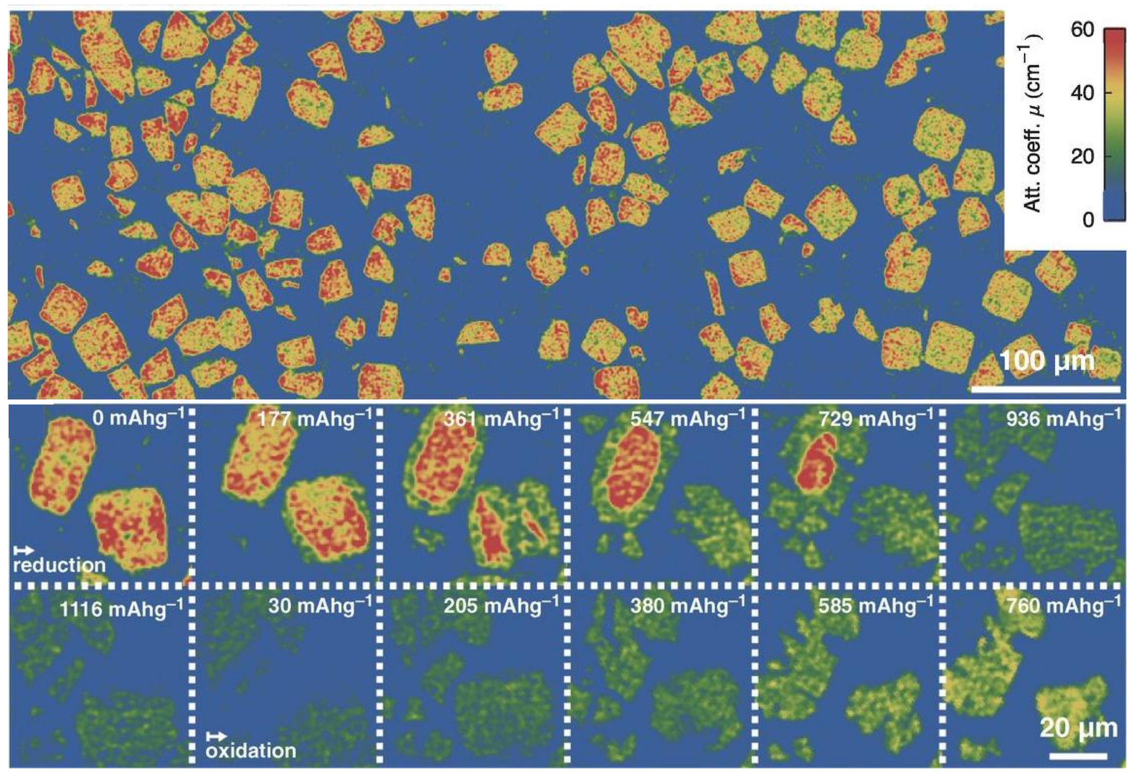


FIGURE 1: SnO phase segregation *(M. Ebner et al., 2013)*

- Phase segregation occurs in alloy anodes such as Si, Sb and Sn, as well as in cathode materials (FIG. 1).
- Diffusion-induced stresses (DIS) will lead to the degradation of electrode particles in Li-ion batteries.
- Cahn-Hilliard (CH) phase field model is coupled with mechanical stresses in this research.
- Isogeometric finite element analysis is employed due to a numerical requirement of $C^1$-continuity.

## 2 Theoretical background

Local mass balance:

$$\dot{\bar{c}} = \nabla \cdot \left[ M_0 \bar{c}(1-\bar{c}) \nabla \mu \right]$$

Local force balance:

$$\mathrm{Div}\,\boldsymbol{\sigma} + \mathbf{b} = \mathbf{0}$$

Constitutive relation:

$$\mu = \delta_c \Psi(c, \nabla c, \boldsymbol{\epsilon}), \qquad \boldsymbol{\sigma} = \delta_{\epsilon} \Psi(c, \nabla c, \boldsymbol{\epsilon}),$$

in which

$$\Psi(c, \nabla c, \boldsymbol{\epsilon}) = \int_B \psi(c, \nabla c, \boldsymbol{\epsilon}) dv,$$

$$\psi(c, \nabla c, \boldsymbol{\epsilon}) = \psi_c(c) + \psi_s(\nabla c) + \psi_e(\boldsymbol{\epsilon}, c),$$

$$\psi_c(c) = \mu_0 c + R\theta c_{max} \chi \bar{c}(1-\bar{c}) + R\theta c_{max}\left[(1-\bar{c})\ln(1-\bar{c}) + \bar{c}\ln\bar{c}\right],$$

$$\psi_s(\nabla c) = \frac{1}{2}\kappa |\nabla c|^2,$$

$$\psi_e(\boldsymbol{\epsilon}, c) = \frac{1}{2}\left[\boldsymbol{\epsilon} - \frac{\Omega c}{3}\mathbf{1}\right] : \mathbb{C} : \left[\boldsymbol{\epsilon} - \frac{\Omega c}{3}\mathbf{1}\right].$$

Here,

- $\bar{c} = \dfrac{c}{c_{max}}$, denotes normalized concentration.
- $\psi_c$, $\psi_s$ and $\psi_e$ represent the homogeneous, interface and elastic free energy, respectively.
- $\mathbb{C}$ is the fourth-order elasticity tensor.
- $\boldsymbol{\epsilon}$ is the small strain tensor.

## 3 Simulation Results

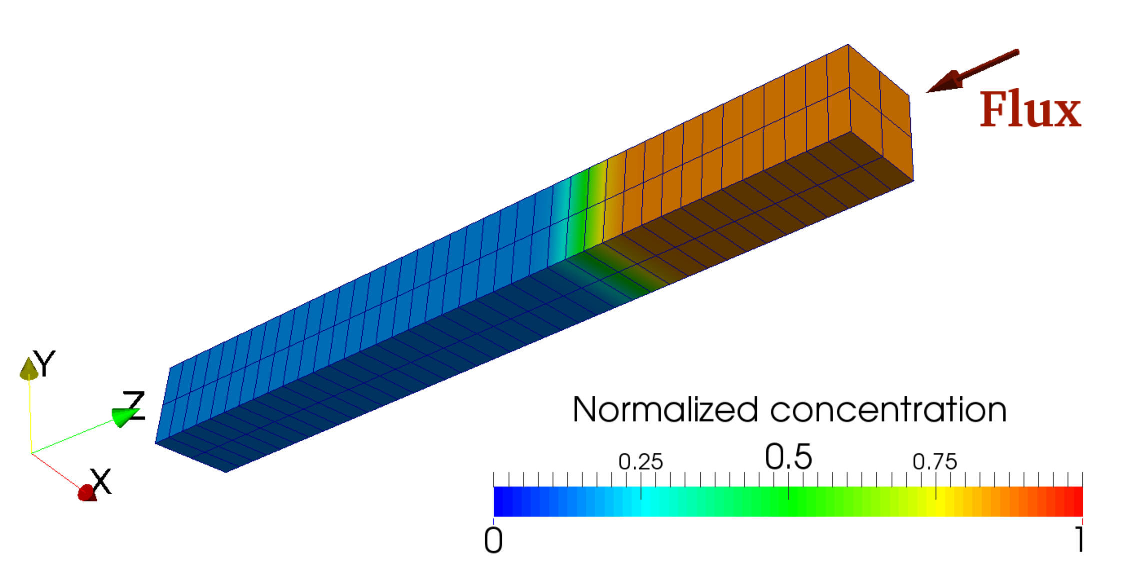


FIGURE 2: Distribution of normalized concentration at equilibrium state after a certain amount of homogeneous flux is given at the end of the bar. The other 5 sides are kept flux-free. For the mechanical part, normal displacements are fixed on the four sides as well as on the bottom; free-end and fixed-end are discussed as two cases (FIG. 6). In FIG. 3 and FIG. 4, the mechanical coupling is not considered.

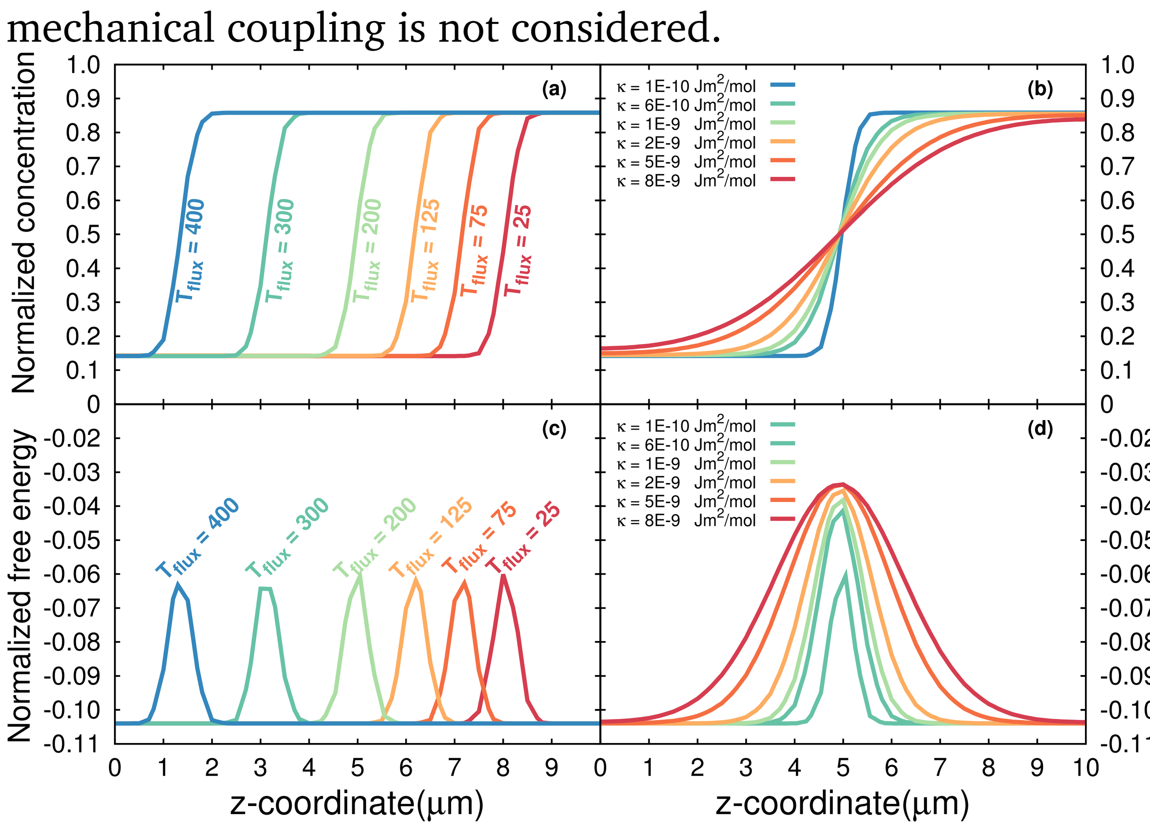


FIGURE 3: Local concentration (a,c) and local free energy (b,d) at equilibrium state. In particular, (a) and (c) show a moving interface due to different amount of incoming flux; (b) and (d) show a dependence of the domain wall free energy and thickness on the interface parameter $\kappa$.

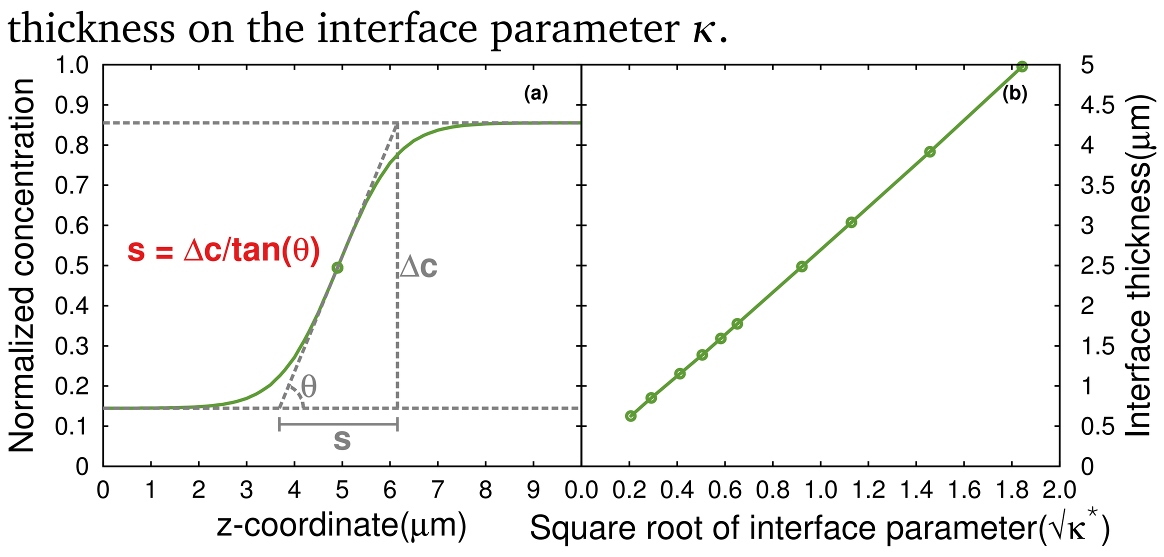


FIGURE 4: Determination of the interface thickness. If the interface thickness is defined as $s$ in (a), a linear relationship $s \propto \sqrt{\kappa}$ can be obtained (b). $\kappa^* = \kappa/\kappa_0$, and $\kappa_0$ is a normalization parameter.

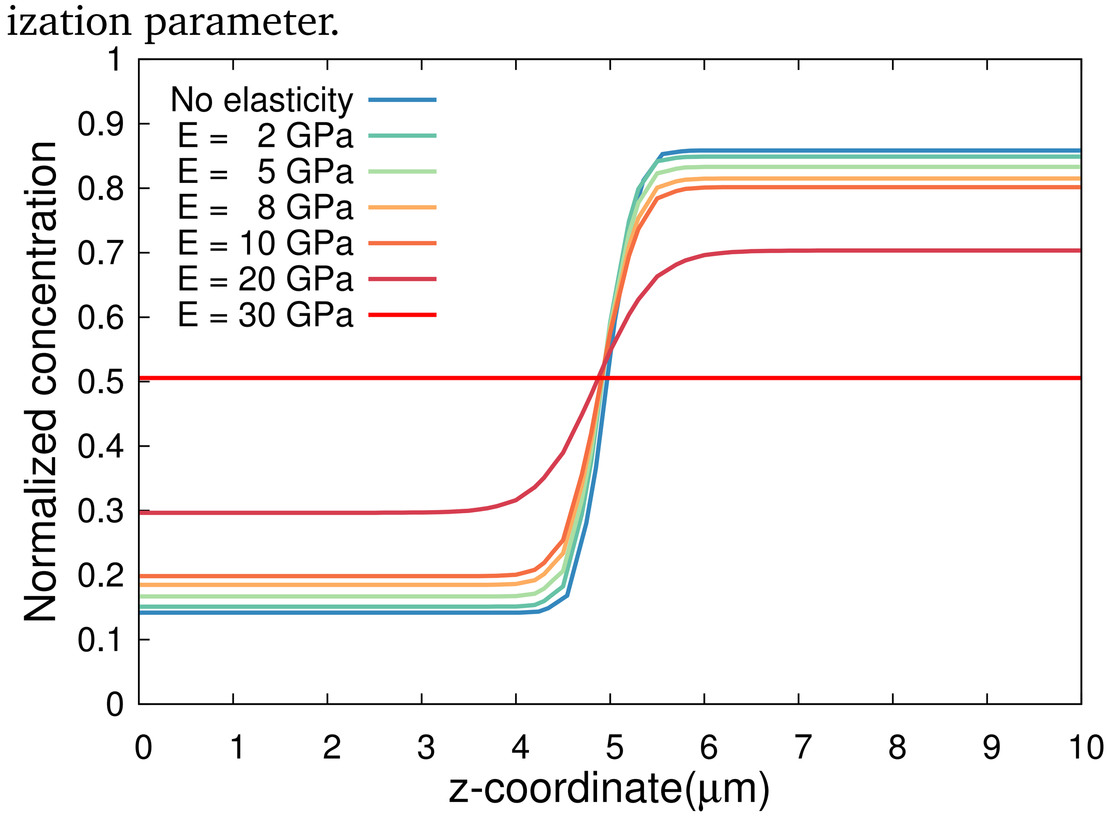


FIGURE 5: The suppression of phase segregation by elasticity.

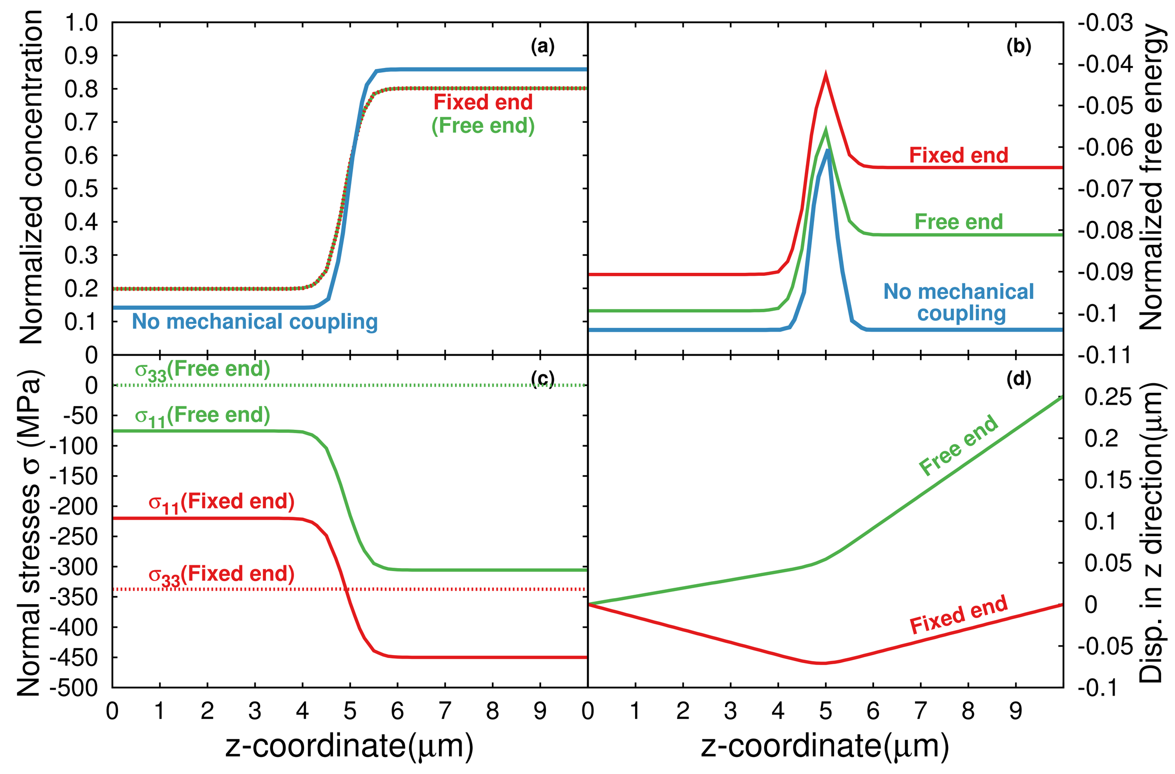


FIGURE 6: Results from a chemo-mechanical coupling model: (a) local concentration, (b) local free energy, (c) normal stresses, (d) displacement in z-direction along the longitudinal axis of the bar.

## 4 Conclusion & outlook

**Conclusions:**

- Two phases with a diffusive interface form after certain amount of flux is provided.
- Interface sits at different locations when different amount of flux is applied.
- Interface thickness is proportional to square root of the interface parameter $\kappa$.
- Interface free energy peaks at the middle of the interface, where the largest concentration gradient exists.
- Phase segregation can be suppressed by mechanical effect.

**Outlooks:**

- Simulation of spherical or ellipsoidal particles.
- Extention into a large deformation model.
- Consideration of the plastic deformation.

## 5 Acknowledgement

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## 6 References

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