

Effects Of Rough Li-LLZO Interface On Electro-Chemo-Mechanical Interactions In ASSB

Investigating how Li|LLZO interfacial roughness affects mechanical contact and local current-density distribution during lithium stripping in all-solid-state batteries (ASSB) using FEM.

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Abstract

The Li|LLZO interface plays a central role in the performance and durability of all-solid-state batteries. Surface roughness produces a heterogeneous contact state, which can affect local current distribution and local mechanical response of the interface during lithium stripping. Previous studies have investigated the influence of interface geometry and mechanical stress on current-density distribution [1-3].

Building on the electro-chemo-mechanical model proposed by Nadeau et al. [1], where interfacial asperities are represented using a Gaussian profile, this work introduces self-affine roughness to investigate its influence on mechanical contact and local current-density distribution at the Li|LLZO interface.

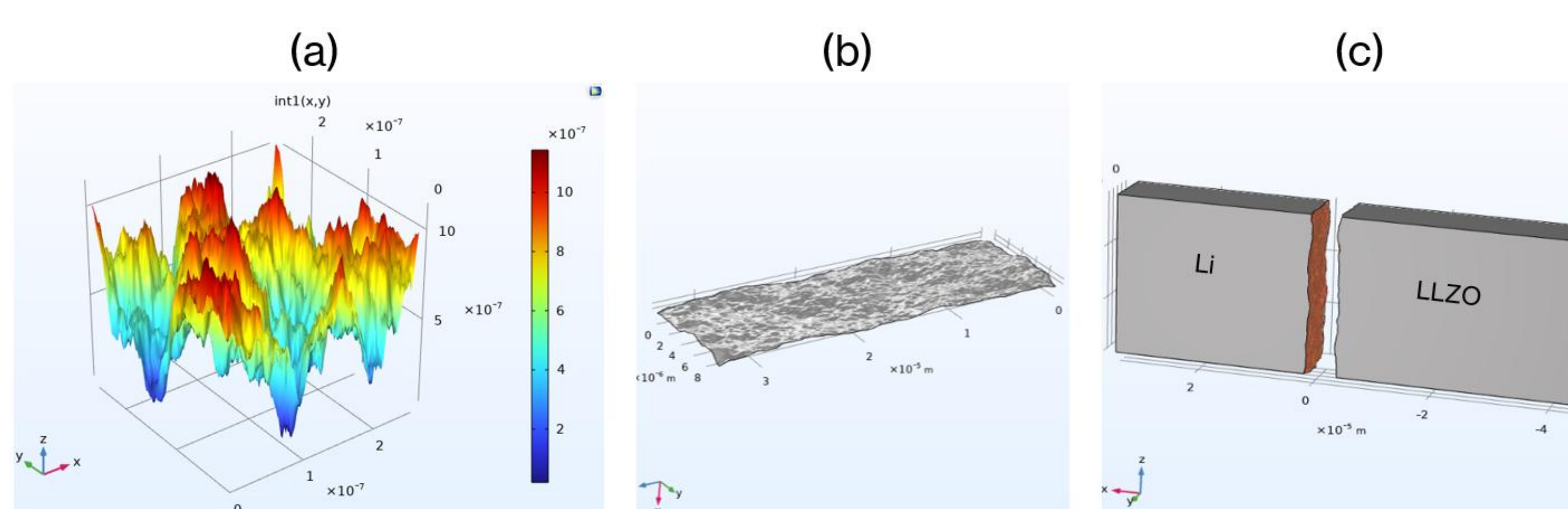


Figure 1. Generation and implementation of self-affine interfacial roughness in COMSOL Multiphysics: (a) Interpolation function representing the generated rough surface, (b) Mapping of the roughness on a parametric surface and (c) Incorporation of the resulting surfaces into the Li and LLZO geometries.

Modeling Approach

The 2D electro-chemo-mechanical model combines Secondary Current Distribution and Solid Mechanics. General Extrusion operators are used to map interfacial electrochemical variables between the opposing Li and LLZO boundaries. A gap-dependent contact function couples the mechanical contact state to the local Butler–Volmer current density.

$$\Phi_{Li}^{LLZO} \xrightarrow{\text{General Extrusion}} \Phi_{Li_on_Li} \rightarrow \eta \rightarrow j_{BV}^{Li} \xrightarrow{\text{General Extrusion}} j_{BV_on_LLZO}$$

A stationary displacement continuation is first used to establish a converged contact state, which is then used to initialize the transient stripping simulation. COMSOL LiveLink™ for MATLAB® is additionally used to extract and partition the 3D mesh and to identify a statistically representative subdomain for future 3D coupled simulations.

Results and Conclusion

The 2D results show a strongly heterogeneous electrochemical response at the Li|LLZO rough interface (Fig. 2). For an imposed current density of 1 mA cm^{-2} , the local Butler–Volmer current density is initially amplified at contacting asperities. Then, it progressively becomes more homogeneous. This evolution is consistent with a progressive redistribution of the electrochemically active contact area as the mechanically deformable Li accommodates the rough LLZO surface.

The model couples mechanical contact with Secondary Current Distribution through a gap-dependent Butler–Volmer current density under stripping conditions. Ongoing work focuses on mesh sensitivity, incorporation of a mechanics-modified overpotential, and extension to a representative 3D rough domain.

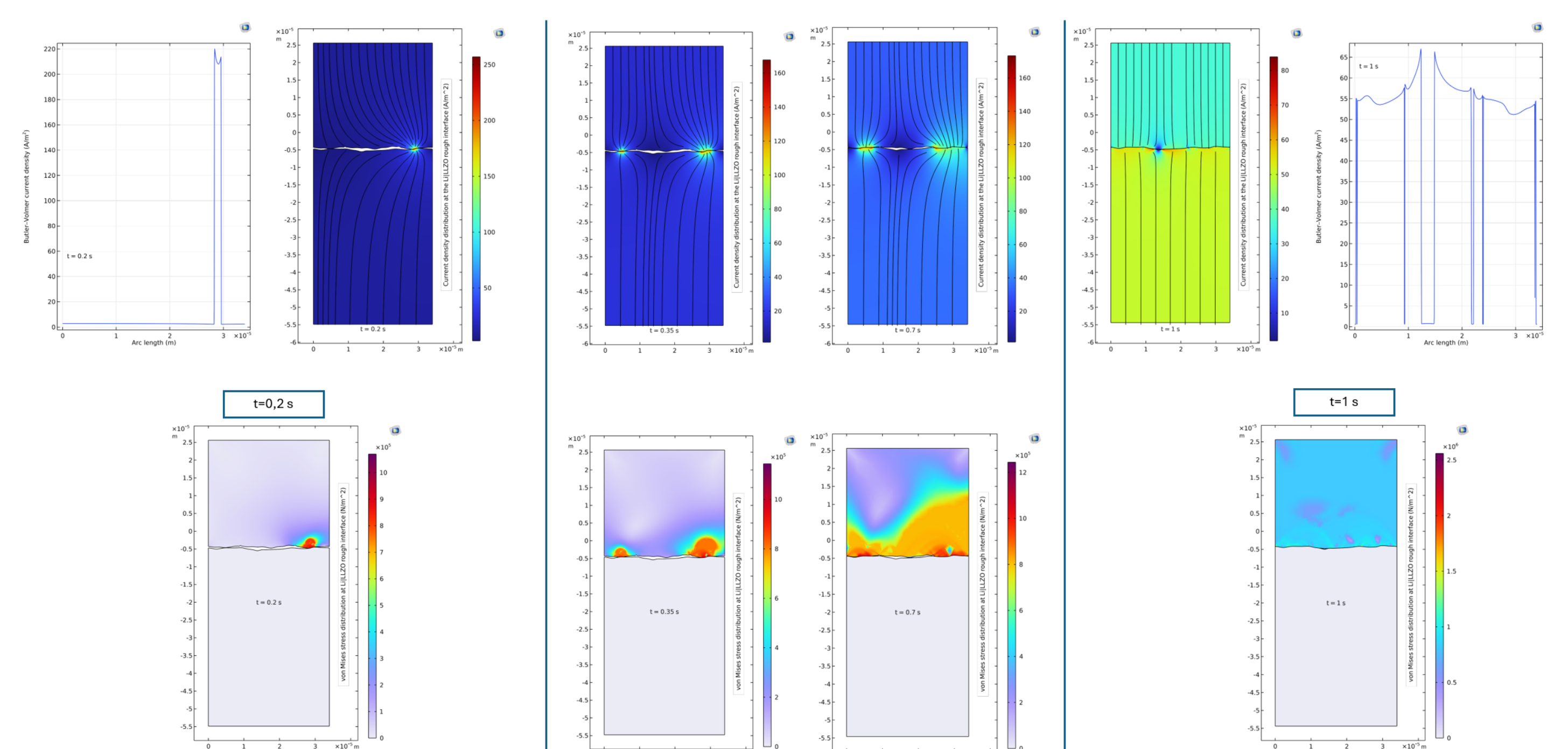


Figure 2. Interfacial electrochemical and mechanical response along the rough Li|LLZO interface at $t = 0,2\text{s}, 0,35\text{s}, 0,7\text{s}$ and 1s .

REFERENCES

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- [3] E.G. Herbert et al., *J. Mater. Res.*, 34 (2019).