

The Failure of Solid Electrolyte Interphase on Li Metal Anode: Structural Uniformity or Mechanical Strength?

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Practical application of lithium (Li) metal anodes has been hindered by Li dendrite growth, which renders a low Coulombic efficiency and short lifespan of working Li metal batteries. A stable solid electrolyte interphase (SEI) is crucial in suppressing the formation of Li dendrites. Herein the local stress and deformation evolvement status of a SEI layer during Li electrodeposition are investigated through a quantitative electrochemical–mechanical model based on a finite element method. Furthermore, the impacts of structural uniformity and mechanical strength on the stability of the SEI under different working conditions are investigated. Improving the structural uniformity of SEI is the most effective way to enhance the stability of SEI, which regulates ion transportation. In addition, pursuing extremely high mechanical strength is shown to be pointless, and a moderate elastic modulus of 3.0 GPa is suggested. This work affords an insight into the rational design of stable SEI layers and sheds light on a possible pathway toward practical applications of Li metal anodes.

1. Introduction

Rechargeable batteries, powering portable electronic devices and electric vehicles, play a pivotal role in sustainable development. The burgeoning demand for electric energy storage constantly propels the upgrading of batteries toward high energy densities. Currently, lithium (Li) ion batteries (LIBs) are almost approaching to its ceiling of theoretical energy densities, and then the research of post-LIBs with high energy densities becomes urgent.^[1] Li metal has been spotlighted as a promising anode due to its ultra-high theoretical capacity (3860 mAh g^{-1}) and an extremely low redox potential (-3.04 V vs standard hydrogen electrode).^[2] A series of battery systems based on

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Li metal anodes (such as Li–sulfur batteries: 2600 Wh kg⁻¹ and Li–O₂ batteries: 3505 Wh kg⁻¹) have demonstrated a huge improvement on energy densities in comparison with commercial LIBs.^[3] However, the capacity loss, short life-span, and safety hazards caused by Li dendrite growth and unstable solid electrolyte interphase (SEI) layer in Li metal anodes have yet to be thoroughly solved after nearly four decades of research.^[4] Tremendous efforts have been devoted to reveal the dendrite growth and SEI layer transformation mechanisms.^[5]

SEI plays the most essential role in the surface morphology evolution of working Li metal anodes during repeated electrodeposition due to its passivating feature.^[6] Because of the high reactivity of Li metal, SEI primarily forms once Li

is in contact with electrolytes, and grows continuously in subsequent electrochemical cycling. A stable SEI layer without rupture and regeneration is necessary for regulating the ionic flux and protecting Li from electrolytes, further achieving a uniform and stable electrodeposition. Previously, the regulation of SEI layer mainly focused on its physical and chemical properties, including structural uniformity, chemical homogenousness, mechanical strength, electric/ionic conductivity, etc. Such regulation in experimental methods is mainly achieved by electrolyte additives and/or artificial SEI layer designs, such as fluorine/nitrogen/sulfur-containing electrolyte additives (e.g., fluoroethylene carbonate, LiNO₃, and lithium polysulfides).^[7] Furthermore, X-ray photoelectron spectroscopy, atomic force microscope (AFM), and cryo-electron microscopy can visually reveal the component and structure evolutions of the particularly designed SEI.^[8] Besides, a prolonged lifespan and enhanced Coulombic efficiency demonstrates the effectiveness of these experimental strategies.^[9] Thanks to the development of interfacial chemistry, material technology, and upscale characterization, great progresses have been achieved on the modification of SEI.^[10] However, a fundamental understanding of how SEI regulates Li dendrite growth and the key parameter of designing effective SEI is still not clear. If such an understanding can be achieved, a rational design of SEI can be used to effectively stabilize electrolyte-anode interface and to resist Li dendrite growth in practical applications of LIBs.

The structural uniformity and mechanical strength of SEI can significantly and directly affect Li-ion transportation and





Figure 1. Schematic illustration of the morphology evolution in Li metal anodes during electrodepositing. A) A structural uniform SEI achieves uniform Li deposition. B) A nonuniform SEI renders irregular Li deposition. Cracks and deformations of SEI occur under low and high mechanical strength, respectively.

the following Li deposition. These two factors are thus mainly discussed in the following. Specifically, structural uniformity is a key factor that regulates the fluctuation of ionic flux. As shown in Figure 1A, an ideal SEI layer with a perfect structural uniformity can induce uniform deposition without regard to the dendritic growth nature of Li. However, structural defects such as nonuniform thickness and small disparity of structure in SEI layers can immensely disturb the distribution of Li-ion flux. During Li electrodepositions, there are much more Li ions migrating through SEI layer at the sites with a thinner thickness, where the nonuniform deposition initiates (Figure 1B). In addition, the chemical inhomogeneity caused by complex reactions and formation processes in actual SEI layer, such as Li₂CO₃, LiF, Li₂O, and ROCO₂Li,^[11] also results in nonuniform ionic conductivity in SEI layers, which aggravates the influence of structural uniformity in Li plating process. From the perspective of ionic diffusion resistance, the influence of chemical inhomogeneity can be quantitatively analyzed by the structural nonuniform model.

Except structural uniformity, the mechanical strength of SEI also plays an important role.^[12] An SEI layer with a low mechanical strength is unable to accommodate the high local stress, which arises from an uneven Li-ion distribution and the following spot accumulation, and will rupture inevitably (Figure 1B). Consequently, fresh Li is exposed to electrolytes again and new SEI generates, inducing the loss of active Li and electrolytes. Meanwhile, a flood of Li ions preferentially rushes

into these narrow cracks and forms dendrites. The repeated disintegration and regeneration of SEI form a terrible positive feedback loop, leading to a low Coulombic efficiency and dendrite growth in Li metal anodes. A SEI layer with a high mechanical strength, which can undergo high stress without fracture, is considered to reduce exchange current density and render plastic deformation on dendritic protrusions, and further leads to a dendrite-free morphology.^[13]

In this contribution, how SEI regulates Li dendrite growth was proved through an electrochemical-mechanical model (coupling stress, diffusion, electric fields, and electrochemical reactions) based on finite element method (FEM). The stress variation in SEI and the failure mechanism of SEI during electrodeposition under different structural uniformity and mechanical strength conditions were quantitatively tracked. The failure time of SEI was quantified with reasonable assumptions and simplification to make accurate evaluation of the stability of SEI. It was found that improving the structural uniformity is the most important factor for stabilizing SEI. Furthermore, enhancing the strength modulus is very useful under a low current density and a moderate elastic modulus above 3.0 GPa is suggested. This work affords a profound understanding of the relationship between SEI properties and Li dendrite growth. A rational design strategy of constructing a stable SEI was also proposed, providing a guidance for further experimental studies and design of LIBs with high performance.

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2. Results and Discussions

2.1. Modeling of Li | Li Symmetric Cells

Li | Li symmetric cell was employed as the research system and a constant current was applied. The ion transport in the electrolyte (filling in the separator, denoted as Ely/Sep) and SEI, electrochemical reactions on electrode surface, and overall force balance of cell are three basic processes in the current electrochemicalmechanical models. As the electrodeposition progresses, both electric and concentration fields change accordingly. Simultaneously, the interface between Li and SEI moves upward, and further renders stress evolution of SEI. It should be noted that the plastic deformation of Li metal induced by the reaction in SEI was not considered. Although the yield strength of bulk Li is less than 1.0 MPa, the yield strength of a single Li dendrite can be two orders of magnitude higher at room temperature.^[14] The local stress involved in this work does not reach the yield strength of the Li dendrites. Consequently, the plastic deformation of Li can be ignored rationally. The details of multiphysics coupling methods are given in Computational Methods.

The schematic illustration of our model is showed in **Figure 2**A, where the SEI thickness (h_{SEI}) is up to 200 nm and the electrolyte/separator domain is in dimension of 20 μ m × 5 μ m.^[15]



Figure 2. Schematic illustration and calculated field distribution of the Li | Li symmetric cell. A) The geometry of the model. B,C) The distribution of B) electric potential and C) Li ion concentration in SEI and electrolyte when $p_d = 0$ and t = 600 s.

The red dashed box in Figure 2A is the main observation domain. The SEI defect on the working electrode side is circularly shaped with a variable depth (h_d) . In order to quantitatively analyze the role of structural uniformity and mechanical strength on the stability of SEI, two variables are introduced in the electrochemical-mechanical model. A dimensionless parameter p_d (i.e., normalized defect depth ratio), which is defined as the ratio of the depth of defect and overall thickness of SEI (h_d/h_{SEI}) , is used to describe the structural uniformity. It varies from 0 to 1, representing the variation from ideal structural uniform SEI to completely failed SEI. Different local defect depth ratios bring various local ionic transport resistances, which is the major factor in the effect of structural nonuniform SEI layer. As a composite film with both amorphous and crystalline components, the failure behaviors and fracture morphologies of SEI layer are mainly controlled by the normal stress.^[16] The elastic modulus of SEI (E_{SEI}) is adapted to judge the mechanical strength. It is assumed that the SEI satisfies ideal elastic-plastic model, and its yield strength is defined as 1% of elastic modulus ($\sigma_{\text{vield}} = 0.01 E_{\text{SEI}}$).

At first, structural uniform SEI was employed to investigate the distribution of electric potential and Li-ion concentration in a cell. All parameters used in this model are listed in Table S1 in the Supporting Information. If there is no special statement, the simulation is conducted under the galvanostatic condition of 3.0 mA cm⁻². The external polarization voltage of Li | Li symmetric cell is around 0.125 V (Figure S1, Supporting Information), which is consistent with the experiments.^[17] After a 600 s electrodeposition, there is an obvious drop of both electric potential and Li-ion concentration emerging in the SEI region as a result of both high electronic and ionic resistance of SEI (Figure 2B,C). There is no obvious change on stress field after electrodeposition (Figure S2A,B, Supporting Information). In terms of theoretical analysis, no stress will arise after ideally uniform depositing on uniform substrate in initial nonstressed electrochemical systems. The change in stress within 1.0 Pa appearing on the Li-SEI interface in Figure S2B in the Supporting Information is related to the meshing accuracy of FEM simulations.

2.2. Impact of Structural Uniformity

In order to judge the failure of SEI, the von Mises criterion was applied in our current model. The SEI layer is considered to be broken and failed when the von Mises stress exceeds the yield strength. The failure time of SEI is denoted as $t_{\rm f}$. The elastic modulus of SEI in 1.0 M lithium bis(fluorosulfonyl) imide (LiFSI) in 1,2-dimethoxy ethane (DME) is measured as 3.5 GPa.^[18] In our simulations, the elastic modulus of SEI is fixed as 3.0 GPa to probe the effect of structural uniformity. Figure 3A-C shows the simulated von Mises stress contours of SEI with different defect depth ratios p_d at obtained $t_{\rm f}$ (the upper subfigure) and 100 s (the lower subfigure). The gray domain represents metallic Li and the dashed line represents the initial position of Li metal surface. As the ion flux (white arrows) through SEI defect is larger than that through the defect-free region, the deposition rate at SEI defect is considerably large, rendering the formation of Li protuberance



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Figure 3. Simulated results of SEI with various defect depth ratios. Distributions of von Mises stress in SEI with A) $p_d = 0.9$, B) $p_d = 0.5$, C) $p_d = 0.1$. The arrows in snapshots represent the distribution of ion flux and the length is proportional to the values. Scale bar on the bottom left is 200 nm. D) The thickness of deposited Li at t = 100 s. E) Von Mises stress of the middle point of Li surface with different p_d . F) Failure time varies with different p_d .

(Figure 3D). For instance, the thickness of deposited Li at the center of defect ($x = 0 \ \mu m$) is 0.4246, 0.4110, 0.4044 μm under a p_d of 0.9, 0.5, 0.1, respectively. But the electroplated thickness at defect-free region is up to 0.4041 μ m. Furthermore, the protuberance squeezes the SEI, resulting in the generation of local stress and the deformation of SEI (Video S1, Supporting Information). The highest stress appears at the defect center, gradually increases with the time and eventually reaches up to the yield strength (Figure 3E). Thereafter, the yielding area (corresponding to the red area in the contour map) continues to expand but von Mises stress no longer increases. Detailed stress components are shown in Figure S3 in the Supporting Information, including shear stress (along the horizontal direction) and normal stress (along the vertical direction). It is seen that the center of the SEI defect is subjected to vertical compressive stress due to extrusion of vertical Li protuberance. In the horizontal direction, the SEI exhibits tensile stress to compressive stress from the center to the sides, respectively. Simultaneous extrusion damage and shear damage are fatal to the stability of SEI. Herein the SEI defect is built as circular shape for avoiding stress concentration at one point. In contrast, wedge-shaped crack was also calculated (Figure S4, Supporting Information). The result shows that the failure time depends on the fillet radius due to the stress concentration. The simulation of driving force on sharp cracks and cleavages needs more precise model based on fracture mechanics. It can be further investigated in the future research.

Statistically, it is found that the failure time of SEI reduces nearly inverse proportionally with increasing defect depth ratio from 0 to 1 (Figure 3F, the fitting curve is $t_f = 8.753 p_d^{-0.974}$). The tangential slope of the fitting curve is calculated in order to make a clear quantitative analysis on its decay speed. As the defect depth ratio increases, the failure time of SEI is reduced. The t_f decreases from 70 to 9.2 s, while p_d increases from 0.1 to 0.9. Particularly, when p_d increases from 0.1 to 0.2, t_f decays rapidly from 70 to 42 s, which implies that the stability of the SEI can be greatly weaken once the defect occurs. When the SEI is already with a very uneven structure ($p_d > 0.5$), the fluctuation of $t_{\rm f}$ is minor. It indicates that for enhancing the stability of SEI, a perfectly structural uniform rather than a relatively structural uniform SEI is required. Toward real operating conditions (e.g., 3.0 mA cm⁻², 3.0 mAh cm⁻²) of Li metal anodes, an SEI layer with $p_d < 0.001$ is appealed for stable cycling, which is estimated from the fitting curve. Notably, this requirement is



targeted to single plating/stripping cycle. Each cycle will cause the SEI to be slightly deformed, i.e., the defect depth ratio will increase incrementally. Therefore, maintaining the SEI stability under long cycles may require ppm-level uniformity. In this respect, strategies proceeded from chemically formed SEI to ameliorate the issue are tough, such as electrolyte additives and pretreatment of Li surface. Spontaneous reaction on Li surface is uncontrollable, hence the nonuniform structure and inhomogeneity are hard to be eliminated. The best approach to construct a structural uniform SEI is the employment of artificial protective layer, which can be realized by polymer coating, magnetron sputtering, atomic layer deposition, and other controllable techniques.^[19]

2.3. Impact of Mechanical Strength

High mechanical strength is considerably necessary for suppressing Li dendrite growth and has become an indicator of the merits of SEI. Great efforts have been devoted to improve the ratio of high modulus components in SEI, such as LiF (70 GPa), LiCl (32 GPa).^[20] To analyze the impact of mechanical strength quantitatively, the SEI with various elastic modulus from 0.05 to 100 GPa was simulated. This range can be considered as the range of organic SEI to inorganic SEI. For instance, the elastic modulus of poly(ethylene oxide), obtained by experiment, is about 70 MPa.^[21] For simplification, the separators were treated as a routine polypropylene separator with an isotropic modulus of 0.5 GPa (E_{sep}),^[22] which ignores the microstructure and anisotropy. In this section, the defect depth ratio p_d is fixed at a constant value of 0.9.

Three typical scenarios are discussed, where E_{SEI} is set as 0.1, 0.5, and 10 GPa, respectively (Figure 4). Comparing the stress distribution in Figure 4A–C, the stress is mainly concentrated in the high modulus region. In the case of the elastic modulus of SEI (0.1 GPa) is less than that of separator, the stress induced by an uneven deposition continues to diffuse toward the separator domain after reaching the yield strength of SEI, corresponding to breaking of SEI (Figure 4A,D). Then safety hazard will emerge when Li protuberances grow under a routine microporous separator. Therefore, a dense and ionic conductive material is essential to fill the microporous separator and suppress the uneven deposition, such as Al-doped Li_{6.75}La₃Zr_{1.75}Ta_{0.25}O₁₂ (LLZTO).^[23]

The failure time of SEI with different E_{SEI} and different p_d is presented in Figure 4F. With the increasing of E_{SEI} from 0.05 to 100 GPa, the failure time of SEI at $p_d = 0.9$ increases from 2.2 to 9.9 s, while at $p_d = 0.1$ increases from 67.3 to 70.5 s. Enhancing the mechanical strength is effective for SEI stabilization. It is noteworthy in Figure 4F that there is an obvious transition point corresponding to E_{SEI} of about 3.0 GPa. When the E_{SEI} increases from 0.05 to 3.0 GPa, the failure time of SEI t_f logarithmically increases with E_{SEI} . However, the failure time of SEI has no significant change when E_{SEI} is higher than 3.0 GPa. Hence, an extremely high elastic modulus is unnecessary, considering the difficulty in improving the integral elastic modulus of SEI. A moderate elastic modulus, just above 3.0 GPa, is suggested and it is easy to implement in practical batteries.

2.4. Failure Time under Different Operating Conditions

The stability of SEI is highly dependent on the structural uniformity and mechanical strength. We incorporate them into one diagram (**Figure 5**A) to compare the influence of the two factors on the stability of the SEI. The impact of structural uniformity is stronger than that of mechanical strength, which exhibits a wider range of color gradient in Figure 5A. Furthermore, the area affected by the structural uniformity is mainly in the bottom of Figure 5A (i.e., $p_d < 0.5$), which represents relative smooth SEI, and the area affected by the mechanical strength is mainly in the upper left corner (i.e., $p_d > 0.8$, $E_{SEI} < 1.0$ GPa), which means rough and high organic content SEI. For thinner SEI (such as 20 nm), the control of structural uniformity is more difficult in experimental regulation. In this case, it is a more effective way to increase its elastic modulus.

In order to investigate the applicability of above regularities, the battery system under a small applied current density of 1.0 mA cm⁻² was computed. Not surprisingly, $t_{\rm f}$ presents inverse proportional fall with the increase of p_d (Figure S5A, Supporting Information) and t_f increases logarithmically with the increase of E_{SEI} to 3.0 GPa (Figure S5B, Supporting Information), which are the same as that under a large applied current density of 3.0 mA cm⁻². Notably, the failure time of SEI is significantly prolonged under low current densities. Combined effects are exhibited in Figure 5B. Similarly, structural nonuniformity-induced instability is more severe than that induced by a low mechanical strength. The former causes the change of $t_{\rm f}$ from infinity to 0 s, while the latter causes the change within 100 s. For relative smooth SEI, color changes from black to orange at 3.0 mA cm⁻² while it changes from black to light blue at 1.0 mA cm⁻² from the perspective of vertical, which means the impact of structural uniformity is weakened. For rough and high organic content SEI, color changes from orange to red at 3.0 mA cm⁻² while it changes from yellowgreen to red at 1.0 mA cm⁻² from the perspective of horizontal, which means the impact of mechanical strength is enhanced. Based on these phase diagrams, we can predict the most important influential factors and suit the remedy to various cases.

3. Conclusions

An ideal SEI layer is very difficult to optimize and even design to achieve uniform and stable Li electroplating and stripping morphology based on empirical summaries obtained from experimental results so far. Therefore, a theoretical model is appealed to achieve more targeted optimization of SEI. We have proposed a quantitative mechanical model of SEI, which is coupled with the electrodeposition process in a Li | Li symmetric cell. In the model, the failure process of SEI layer is quantitatively described. The corresponding failure time t_f is calculated to serve as an indicator of stability of SEI, and subsequently the impact of the properties of SEI (structural uniformity and mechanical strength) and operating conditions of batteries on the stability of SEI is predicted.

First, SEI can be significantly stabilized with the improvement of structural uniformity when reducing the defect depth ratio p_d of an SEI layer. We found that the failure time of SEI t_f www.advancedsciencenews.com



Figure 4. Simulated results of SEI with various elastic moduli. Distributions of von Mises stress in SEI with A) $E_{SEI} = 0.1$ GPa, B) $E_{SEI} = 0.5$ GPa, C) $E_{SEI} = 10.0$ GPa. Scale bar on the bottom left is 200 nm. D) The thickness of deposited Li. E) Von Mises stress of the center of Li surface with different E_{SEI} . F) Failure time varies with different E_{SEI} and p_d , where t_{fmax} represents the maximum failure time under each p_d .

reduces nearly inverse proportionally with increasing p_d from 0 to 1. Then such improvement is much more effective when $p_d < 0.5$, while the increase of failure time t_f is minor when $p_d > 0.5$. And it can be inferred that an SEI layer with ppm-level defect depth ratio is required for practical conditions (3.0 mA cm⁻², 3.0 mAh cm⁻²) and long cycles. Constructing an artificial SEI layer without uncontrolled liquid phase reaction, such as coating and sputtering, is highly recommended to achieve high structural uniformity with a small p_d . Second, an elastic modulus of 3.0 GPa is strong enough for stabilizing an SEI layer. When the elastic modulus of SEI E_{SEI} increases from 0.05 to 3.0 GPa, the failure time of SEI t_f logarithmically increases with E_{SEI} . However, there is no obvious change of t_f when increasing E_{SEI} lager than 3.0 GPa. It implies that an extremely high elastic

modulus is unnecessary in practical applications. The SEI layer with an elastic modulus of 3.0 GPa is easy to realize in practical batteries. For example, the elastic modulus of SEI in 1.0 M LiFSI in DME is up to 3.5 GPa.^[18]

Our current model provides a fundamental and quantitative understanding of how SEI layer regulates Li deposition in Li metal batteries. The impact of structural uniformity and mechanical strength is quantitatively studied. A rational design guidance for constructing stable SEI is also summarized. Although it is very difficult to obtain the whole properties of SEI, both surface roughness and elastic modulus can be characterized by the AFM.^[11b,24] Hence, further experimental verification of this model is feasible. Simulation methods are effective in exploring the mechanisms in Li metal batteries.





Figure 5. Influence of structural uniformity and mechanical strength on SEI failure time under different operating conditions, A) 3.0 mA cm⁻², B) 1.0 mA cm⁻².

Recently, a series of numerical models has been developed to investigate the mechanism of dendrite growth, including phase field model, Monte Carlo model, coarse-grained mesoscale model.^[25] The influence of operation conditions (temperature, current density, and external pressure) and battery compositions (electrolytes and electrode hosts) was quantitatively discussed. But a few models considered SEI.^[15] More attentions are appealed to pay on the modeling of SEI in the future. The model developed here is not comprehensive, but it offers a basic framework which can be further extended in more realistic conditions, such as the offset of equilibrium potential induced by stress, the growth of SEI during electrodeposition, and the complicated failure criteria of SEI.^[16,26] The combination of simulation and experimental results is promising in explaining unspecified mechanism issues in Li metal batteries such as lithium dendrite growth and SEI formation, with which a rational design of high-energy-density batteries can be achieved.

4. Computational Methods

COMSOL Multiphysics 5.4 based on FEMs was used to perform the modeling of failure of SEI on Li metal anode. In the modeling, the current distribution, mass transport, electrochemical reactions, and stress distribution were coupled together. Five modules (secondary current distribution, transport of diluted species, solid mechanics, deformed geometry, and boundary ordinary differential equation) in COMSOL were used to solve the electrochemical-mechanical modeling. The main governing equations are described as below.

Li ions in electrolytes and SEI were controlled by the Nernst– Plank equation

$$J_i = -D_i \nabla c_i - z_i u_i F c_i \nabla \phi \tag{1}$$

where subscript *i* represents ionic species *i*, J_i the total flux, D_i the diffusion coefficient, c_i the concentration, z_i the charge number, *F* the Faraday's constant, φ the electric potential, and u_i the ionic mobility which can be computed by the

Nernst–Einstein relation. $-D_i \nabla c_i$ is the diffusion flux. The other part $-z_i u_i F c_i \nabla \varphi_i$ is the electromigration flux.

The reaction of Li ions with Li metal at the electrode surface (Li/SEI interface) was implemented by the concentrationdependent Butler–Volmer equation

$$i_{\rm loc} = i_0 \left(C_{\rm R} \, \exp\!\left(\frac{\alpha F \eta}{RT}\right) - C_{\rm O} \, \exp\!\left(\frac{\beta F \eta}{RT}\right) \right) \tag{2}$$

where $i_{\rm loc}$ denotes the local current density, i_0 the exchange current density, η the overpotential, α and β the transfer coefficients, $C_{\rm R}$ and $C_{\rm O}$ the concentration of reduced species and oxidized species, *R* the ideal gas constant, and *T* the operating temperature.

An elastic–perfectly plastic model was used to characterize the deformation of the Li | Li symmetric cell. It was assumed that no slip generated between SEI and Li surface and no stress existed in the initial state. The deformation of SEI is under the plane-strain condition. The von Mises stress to characterize the failure of SEI is given by

$$\sigma_{\text{Mises}} = \sqrt{\frac{(\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2}{2}}$$
(3)

where $\sigma_1, \, \sigma_2, \, \text{and} \, \sigma_3$ is the first, second, and third principal stress, respectively.

On account of that elastic relaxation is much faster than diffusion, quasi-static was adopted, which ignores the secondorder time derivatives. The state of stress was solved by the mechanical equilibrium equation

$$\nabla \cdot \boldsymbol{\sigma} = \boldsymbol{0} \tag{4}$$

where σ is the Cauchy stress tensor.

The electrolyte–SEI boundary was set as a free boundary. The SEI–Li interface was set as a prescribed displacement boundary. The plating/stripping thickness of Li was used as the displacement in the normal direction. Table S1 in the Supporting Information lists the values of all parameters used in simulations based on literature values.

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Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

finite element method, Li metal anodes, mechanical strength, solid electrolyte interphase, structural uniformity

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