Modeling of Electro-deposition and Mechanical Stability at Li Metal/Solid Electrolyte Interface during Plating in Solid-State Batteries

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Summary

Interfacial deposition stability between Li metal and a solid electrolyte (SE) is important in preventing interfacial contact loss, mechanical fracture, and dendrite growth in Li-metal solid state batteries (SSB). In this work, we investigate the deposition and mechanical stability at the Li metal/SE interface and its consequences (such as SE fracture and contact loss). A wide range of contributing factors are investigated, such as charge and mass transfer kinetics, plasticity of Li metal and fracture of the SE, and the applied stack pressure. We quantify the effect of the ionic conductivity of the SE, the exchange current density of the interfacial charge-transfer reaction and SE surface roughness on the Li deposition stability at the Li metal/SE interface. We also propose a “mechanical stability window” for the applied stack pressure that can prevent both contact loss and SE fracture, which can be extended to other metal-electrode (such as Sodium) SSB systems.

Keywords

Li-Metal solid-state battery, Continuum modeling, Interfacial deposition stability, Mechanical stability, Li plasticity, SE fracture, Contact loss
Introduction

Li-metal all-solid-state batteries (SSBs) are promising candidates to replace conventional secondary batteries, providing improved safety with the use of non-flammable solid electrolytes (SE) (1, 2) and higher energy density with the use of a Li-metal anode (3, 4). However, several bottlenecks originating from the intrinsic properties of Li metal and SEs hinder the practical application of Li-metal SSBs. First, although ceramic SEs have elastic moduli that are several times higher than that of Li metal, and are predicted to be stiff enough to suppress Li growth in the SE (5, 6), recent experiments have revealed the presence of metallic Li in the SE, which can lead to cell shorting and potential safety issues (7, 8). Second, the high interfacial overpotential limits the power density and rate performance of Li-metal SSBs (9), which has been attributed to poor mechanical contact at the Li metal/SE interface (10, 11) and the slow charge transfer kinetics stemming from the unstable interface between the SE and Li metal (12, 13).

Substantial efforts have been devoted to design a system with stable Li deposition and intimate interfacial contact, including new design of porous electrodes to suppress dendrite growth (14, 15), the use of artificial interlayers to eliminate SEI formation (16, 17), and application of mechanical pressure to reduce the interfacial resistance (18-20). However, a mechanistic understanding of the effects of the interface irregularities, the charge-transfer kinetics, and the mechanical pressure on the dendrite growth and contact loss at the Li metal/SE interface remain elusive.

Theoretical investigations are important for understanding the behavior at the Li metal/SE interface considering the difficulty in characterizing of the buried interface between Li metal and the SE. Monroe and Newman investigated the role of mechanical
stress in the growth stability of a planar interface (5), and suggested that a high shear modulus of the electrolyte (at least two times higher than that of Li metal) removes the inherent instability of a planar growth, and by extension, may limit dendrites (6). Their work has been further extended by considering the plasticity of Li metal (21) and the large shear modulus of SEs (22). In their derivations, the interfacial stress was believed to be the main factor that affects the surface overpotential, which in turn changes the current density distribution at the Li metal/SE interface. Therefore, they concluded that mechanical properties of the Li metal and the SE (such as Shear moduli, Poisson’s ratio) were dominant factors for the Li deposition stability. However, it has been demonstrated that many other factors, such as interfacial geometry (23), charge-transfer kinetics on the interface (24), ion transfer in bulk SE (25), and the externally applied stack pressure (26), can all influence the deposition stability.

In general, uneven distributions of current and stress at the Li metal/SE interface (due to geometric irregularity (23) or material inhomogeneity (27)) lead to unstable Li electrodeposition (dendrite formation) and unstable morphological evolution (interfacial contact loss). Slow ionic transport and/or slow charge transfer between Li metal and SE may increase the area-specific resistance (ASR) of the cell (28, 29) but they can also suppress dendrite formation by creating a more homogeneous current distribution. Externally applied mechanical pressure (or “stack pressure”, 1~100 MPa depending on the SE used in the cell (30, 31)) may alleviate physical contact loss through deformation and flow of Li metal (26); but can also facilitate dendrite propagation when stress intensification at specific locations of the interface (such as the tips of defects) leads to fracture of the SE (32). Investigating the combined effects of these various factors requires a model that couples Li electrodeposition and mechanical deformation together.
The objective of the current study is to provide basic models for the deposition and mechanical stability of the interface between Li metal and the SE under boundary conditions (BCs) that are relevant for practical in Li-metal SSBs. We first show that the interfacial stress caused by the stack pressure considered in this work has a much smaller effect on the Li deposition stability compared with other factors, such as the charge-transfer of \( Li^+/Li \) at the interface and the mass-transfer rate of \( Li^+ \) in the SE. Second, we investigate Li electrodeposition and plastic deformation for a wide range of BCs and material properties, including the shape of the surface irregularity, the charge-transfer kinetics at the Li metal/SE interface, the conductivity of bulk SE, the yield strength of Li metal, and the applied stack pressure. Finally, we examine the development of contact loss arising from a finite amount of Li deposition. The increase of surface overpotential as a consequence of contact loss is also discussed. In conclusion, we demonstrate that although ideal stable Li electrodeposition is impossible, the electrochemical and mechanical conditions can be optimized to reduce the electro-deposition instability and ensure mechanical stability of the Li metal/SE interface, thereby allowing control of the overpotential and suppression of dendrite formation.
Methodology

Model geometry

The model geometry used for all calculations is shown in Figure 1, where the Li metal anode is on top of a rough SE surface with perfect initial contact. The cell is reduced to a 2D geometry by assuming plane strain and isotropic transport properties in the out-of-plane z-direction. It has been demonstrated that good initial contact can be experimentally achieved by applying large pressure or heat treatment (26, 33). Here, we mainly focus on the interfacial morphology evolution with continued Li deposition at the interface. We first study the inhomogeneities of current density, interfacial overpotential and stress at the Li metal/SE interface caused by the conformal defects of the initial geometry, and then study the evolution of these inhomogeneities when the interface evolves (For example, contact loss may be developed if the stack pressure $P_0$ is not large enough to drive plastic deformation of the excess Li metal in the confined space).

In the present work, the Li deposition stability and the mechanical stability are described by the electro-deposition stability factor $\vartheta$ and the contact loss factor $\gamma$ respectively. The deposition stability factor is defined as $\vartheta = \frac{i_n(peak)}{i_n(valley)}$, where $i_n(peak)$ and $i_n(valley)$ are the current densities at the peak and valley of the Li protrusion, respectively, as defined in other work (22, 34). If the current density ($i_n$) is evenly distributed at the interface $S$, the stability factor $\vartheta$ corresponds to homogeneous deposition of metallic Li along the interface $S$. If $\vartheta \gg 1$, Li deposition at the interface $S$ is strongly inhomogeneous. The mechanical stability is defined by the degree of interfacial contact: $\gamma = \frac{A_s(t)}{A_s(0)}$, where $A_s(t)$ is the remaining contact area after $t$ seconds and $A_s(0)$ is the initial contact area. If $\gamma = 1$, the initial conformal contact is maintained. When $\gamma < 1$, interfacial contact
loss is developed, which not only causes larger uneven Li deposition but also increases the surface overpotential at interface $S$.

Both the Li electrodeposition and interfacial contact loss are affected by the charge-transfer reactions at interface $S$ (described by the Butler-Volmer relation), mass transfer in the SE (described by the Nernst-Planck relation), and interfacial contact mechanics (described by elastoplastic continuum mechanics). Equations and assumptions associated with the related physical processes are reviewed and discussed in the following subsections.

**Mechanical equations**

The continuous plating/stripping on interface $S$ generates stress fields in both the Li metal and the SE. The presence of initial defects creates inhomogeneous stress distribution, which may be sufficient to drive plastic deformation of Li metal or fracture of the SE (21). Li metal has recently been reported to exhibit different yield and creep behaviors at room temperature (35, 36). Therefore, a comprehensive approach combining transport with elasticity and plasticity of both the Li metal and SE is required. This is achieved by solving the combined equations:

Quasi-static mechanical equilibrium is assumed to both Li metal and the SE:

$$\nabla \cdot \sigma = 0 \tag{1}$$

Linear elasticity is assumed for the elastic state of both Li metal and the SE:

$$\sigma = \frac{E}{1+v} \varepsilon + \frac{vE}{(1+v)(1-2v)} \text{trace}(\varepsilon)I \tag{2}$$

Perfect plasticity without hardening is assumed for the plastic state of Li metal:
\[
\Phi(\sigma) = |d\varepsilon(\sigma)| - \sigma_y
\]  

(3)

At boundary \( S_1 \): \( \sigma n = P_0 n \).

At boundary \( S_0 \): Fixed in both x and y direction. At \( S_L \) and \( S_R \): Fixed in x direction.

Here, \( \sigma \) is the stress tensor; \( \varepsilon \) is the strain tensor; \( E \) and \( \nu \) are the Young's modulus and Poisson's ratio of Li metal or the SE, respectively; \( \sigma_y \) is the yield strength of Li metal; \( n \) is the normal vector to the interface \( S \); \( P_0 \) is the external pressure (stack pressure); and \( \nabla \cdot \), \( \text{trace}(\ast) \) and \( d\varepsilon(\ast) \) are the divergence, trace, and deviatoric operators, respectively. The stress tensor \( \sigma \) can be decomposed into two components: the hydrostatic pressure \( P \) \((P = \frac{1}{3} \text{trace}(\sigma))\), which may affect the current density \( i_n \), as described in Newman’s modified Butler-Volmer relation (5), and the deviatoric von Mises stress \( \tau \) \((\tau = |d\varepsilon(\sigma)|)\), which drives the plastic flow of metallic Li and the fracture of the SE.

In our time resolved simulations, mechanical equilibrium (equation 1) is reached at the end of each timestep for which the electrochemical problem is solved, because mechanical equilibrium occurs at the speed of sound, which is much faster than the electro-chemical kinetics in the bulk SE and at the Li metal/SE interface (37). Please refer to section SI-1 for more detailed descriptions.

**Electrochemical equations**

Since the SE is a single-ion conductor, it cannot accumulate carriers, and the conduction is therefore purely ohmic:

\[
\nabla^2 \phi_{SE} = 0, \quad i = -\sigma_{Li^+} \nabla \phi_{SE}
\]

(4)

At interface \( S \), Butler-Volmer boundary condition is employed:
\[
i_n = i_{\text{exc}} e^{\frac{(1-\alpha_a)\Delta\mu_e^{-}}{RT}} \left( e^{\frac{\alpha_aF}{RT}\eta} - e^{\frac{\alpha_cF}{RT}\eta} \right), \quad i_n = i \cdot n \tag{5}
\]

At boundary \( S_0 \), constant current flows into SE: \( i = i_0 \).

At boundary \( S_L \) and \( S_R \), no current flows out of the sample box: \( i \cdot n = 0 \).

\( \phi_{SE} \) is the electrostatic potential in the SE; \( i \) is the current density in the SE; \( n \) is the normal direction of boundaries; \( i_{\text{exc}} \) is the exchange current density of the \( Li/Li^+ \) reaction; \( \alpha_a \) and \( \alpha_c \) are the anodic and cathodic charge transfer coefficients, respectively; and \( \eta \) is the surface overpotential.

Detailed derivations are provided in section SI-2 for \( Li^+ \) conduction in the SE and in section SI-3 for charge-transfer at interface \( S \). Equation 4 implies the \( Li^+ \) diffusion in the SE caused by the \( Li^+ \) concentration gradient is negligible, which indicates that \( Li^+ \) ions are homogeneously distributed in both the bulk and on the surface of the SE without the presence of a “space charge layer”. Although some theoretical work has argued a space charge layer at the Li metal/SE interface (28), there is no experimental evidence of its existence. Equation 5 is the Butler-Volmer equation that incorporates the effect of interfacial stress in the variable \( \Delta\mu_e^{-} \) (defined as the change of the electrochemical potential of electrons) (5, 6).

**Effect of mechanical stress on deposition stability**

The effect of mechanical stress on the current density was first studied by Monroe and Newman (5, 6). In a solid-state battery, the mechanical effect will modify the original Butler-Volmer equation by a factor of \( \exp \left[ \frac{(1-\alpha_a)\Delta\mu_e^{-}}{RT} \right] \), where: \( \Delta\mu_e^{-} = V_Li\Delta P_{Li} \). (\( V_Li \) is the partial molar volume of Li in Li metal, \( \Delta P_{Li} \) is the pressure of Li metal near interface \( S \)).
The effect of pressure on the current distribution in the SE near interface $S$ is neglected because of the rigid behavior of inorganic ceramic SE materials. For a typical value of stack pressure ($P_0$) (~1 to 5 MPa for SSBs), $\Delta P_L$ developed in the Li metal near interface $S$ is no more than ~10 MPa (even considering the stress intensification at the peak of defects, as shown in the Results section). At room temperature, the term corresponding to the mechanical effect on the current, $\exp \left[ \frac{(1-\alpha_a)\Delta \mu_e}{RT} \right] = \exp \left[ \frac{(1-\alpha_a)\nabla_L \Delta P_L}{RT} \right]$, is smaller than 1.02, which implies changes of $i_n$ that are less than 2% with pressure. Therefore, the effect of stress on creating an inhomogeneous current distribution is negligible compared to the effect of inhomogeneities from pre-existing surface irregularities and the charge and mass transfer kinetics, which will be discussed later in the Results section 1-2.

**Numerical solutions and parameters**

All the partial differential equations (PDEs) are summarized in section SI-5. A self-developed code based on the finite element method (FEM) is used to solve these nonlinear PDEs. A flow chart of the code is presented in Fig.SI-1b. Default values of parameters used in this work are from reported experimental measurements of reference papers listed in the last column, as shown in Table 1.
Results

**Effect of initial surface irregularity on Li deposition stability**

As the presence of surface defects (such as cracks/voids) on the interface $S$ is common on the SE surface (7, 8), we first study the effect of such defects on Li deposition. We guide ourselves from experimental SE surface roughness data, as for example, determined through atomic force microscopy (AFM) measurements (40). Several critical parameters are needed to define the surface roughness, including the arithmetic mean length (the average length of defects on a rough surface, $l$) and mean width ($w$), area percent (the ratio of the real surface area to the sampling area), and kurtosis (the shape of defects, with a larger value indicating a sharper defect). For a typical SE material, the mean defect length and width range from 0.01 to 1 $\mu m$, and the kurtosis varies from -1 to 10 (40, 41).

Figure 2a shows the model geometry where a small interface irregularity is created at the top of the SE with varying length ($l$), width ($w$), and shape (cosine-shaped, elliptical, or circular). Perfect initial contact between Li metal anode and the SE is assumed and a constant total current density ($i_0 = 0.1 \text{ mA/cm}^2$) is applied at the bottom boundary. Small irregularities (both defect length and width are much smaller than cell size: $\frac{l}{L} \ll 1, \frac{w}{W} \ll 1$) are introduced to ensure that any perturbations of the current density induced by the defect vanish at the boundaries, as indicated by the vertical flow of current density (red arrows) when approaching the boundary $x = \pm 0.5 \mu m$. The current contour lines in Figure 2a show that current is concentrated near the defect, causing an uneven distribution of the normal current density $i_n$. Figure 2b shows the perturbations affected by defects of typical sizes ($w = 40 \text{ nm}$, $l = 100 \text{ nm}$) that are obtained after fine surface polishing of the SE (8). While the current density near the defect is highly perturbed, the current density $i_n$ at the...
interface $S$ reaches a constant value ($\sim 0.1 \, mA/cm^2$) for $|x| > 0.2 \, \mu m$, implying that the perturbation from the defect does not affect the region beyond ten times the defect width. For typical solid electrolyte materials, the surface area percent (ratio of rough surface area to sampling area) can be less than 10% with simple micro/nano polishing (40, 42). This means that most irregularities at Li metal/SE interface will only cause “local” effects to the current density and potential distribution for a well-polished SE surface.

The inset in Figure 2b shows that the peak value of the current density for the three different shaped defects is almost the same ($\sim 0.11 \, mA/cm^2$), with the cosine-shaped defect exhibiting a lower magnitude difference ($\vartheta = i_n(peak)/i_n(valley)$) than the elliptical or circular defect. Because the current differences between the three shapes are small, only cosine-shaped defects are considered for later calculation and discussion.

Figure 2c plots the charge-transfer overpotential $\eta$ (the difference between the electric potential of the SE and Li metal at interface $S$) of three defects with different lengths ($l = 10 \, nm, 100 \, nm, 200 \, nm$) and the same width ($w = 40 \, nm$). As the defect length increases, the overpotential becomes more concentrated near the defect and its maximal value grows. For a defect with length of $1 \, \mu m$ and width of $8 \, nm$ under applied current density $1 \, mA/cm^2$, the maximal overpotential can reach $3 \, mV$. If unscreened, this large overpotential could generate a stress with magnitude $25 \, MPa$ according to the thermodynamic relation: $\sigma = \frac{F}{V_{Li}} \eta$, which is sufficient to crack SE materials with low fracture toughness. (8)

A more systematic study of the geometry effect (defect width $w$ and length $l$) on the stability factor $\vartheta$ is done by calculating stability factors for defects with various widths and lengths, which is shown in Figure 2d. In general, $\vartheta$ decreases when the defect is “shallow
and wide” (top right area) and increases when the defect is “deep and narrow” (bottom left area). Therefore, to increase the depositional stability, surface engineering methods should be employed to make the SE surface as flat as possible. For example, mechanical polishing can be used to reduce the defect length, and surface corrosion and etching can be applied to increase the defect width or modify the defect shape (41). Moreover, Figure 2d shows that for a long defect, $\vartheta$ is very sensitive to $w$. Thus, for a defect length $> 10 \mu m$, a small increase of the defect width can notably homogenize the plating of metallic Li and increase the depositional stability.

The interfacial area-specific resistance can be computed as $ASR_{\text{interface}} = \eta / i_n$, where $\eta$ is the overpotential at interface S and $i_n$ is the normal current density at the same point. Notably, $ASR_{\text{interface}}$ is dependent of the interfacial contact loss and the exchange current density at the interface. In experiments, the interfacial resistance can be measured through electrochemical impedance spectroscopy (EIS), whose value is usually a combined effect of the interfacial contact loss and the interfacial charge transfer, which explains why the measured values can be very different even with the same material. For example, ASR values between 0.2 and 200 $\Omega \cdot cm^2$ have been measured for Li/LLZO/Li cells depending on the pressing pressure (20). To decouple the contact loss resistance and the charge transfer resistance, intimate interfacial contact is needed, which can be achieved either by applying large enough stack pressure (400 MPa stack pressure gives $\sim 0.2 \Omega \cdot cm^2$ for LLZO (20)), or by smooth enough surface conditions (10 nm surface roughness gives $\sim 0.5 \Omega \cdot cm^2$ for LiPON (40)). When intimate contact is assumed in our work, the interfacial resistance $ASR_{\text{interface}}$ is 0.26 $\Omega \cdot cm^2$, in good agreement with the above experiments. Effects from contact loss will be discussed in the last section.
Effect of charge transfer kinetics on deposition stability

Additional factors that affect the deposition stability are the SE ionic conductivity $\sigma_{Li^+}$ and the exchange current density $i_{exc}$, which controls the $Li^+$ mass-transfer rate at the interface, as shown in Equation 5. $Li^+$ conductivity within the SE is the limiting factor for deposition stability if $\sigma_{Li^+}$ is small, whereas the $Li^+/Li$ charge-transfer reaction is the limiting factor if $i_{exc}$ is small. Because the formation of a SEI is common between the electrode and SE (such as Li metal with LPS), substantial changes of the ionic conductivity $\sigma_{Li^+} \left(10^{-3}-10^{-1} \text{mS/cm}\right)$ (39) and exchange current density $i_{exc} \left(10^{-3} \text{mA/cm}^2\right)$ (38, 43) are considered in this study.

The effect of ionic conductivity $\sigma_{Li^+}$ and exchange current density $i_{exc}$ on Li deposition is studied with the same model shown in Figure 2a. A constant total current density ($i_0 = 0.1 \text{mA/cm}^2$) is applied at the bottom boundary. In Figure 3a, the normal current density $i_n$ at interface $S$ is plotted for three different values of $i_{exc}$ with $\sigma_{Li^+} = 0.3 \text{mS/cm}$ (39). While a small exchange current density $i_{exc}$ corresponds to a higher overpotential, it can homogenize the current distribution when it becomes the rate limiting step. Figure 3b shows the current density $i_n$ for three different values of ionic conductivity $\sigma_{Li^+}$ with $i_{exc} = 100 \text{mA/cm}^2$ (38). A larger ionic conductivity $\sigma_{Li^+}$ corresponds to faster $Li^+$ migration in the SE, which also homogenizes the current distribution. Therefore, a smaller exchange current density $i_{exc}$ or larger ionic conductivity $\sigma_{Li^+}$ is preferred for stable Li electrodeposition. The coupled effect of $\sigma_{Li^+}$ and $i_{exc}$ on the deposition stability $\theta$ is shown in Figure 3c, with ionic conductivity $\sigma_{Li^+}$ ranging from $3 \times 10^{-4}$ to $3 \times 10^{-1} \text{mS/cm}$ and exchange current density $i_{exc}$ ranging from $10$ to $10^3 \text{mA/cm}^2$. It is clear from
the figure that both the ionic conductivity and the exchange current density can have a large influence on the stability factor of deposition.

To maintain stable electrodeposition (top left area of Figure 3c) at interface $S$ without dendrite formation and contact loss, a Li metal SSB should possess a SE with high ionic conductivity $\sigma_{Li^+}$ in the bulk and low exchange current density $i_{exc}$ at the interface with Li metal. Notably, a higher $\sigma_{Li^+}$ not only ensures more stable Li deposition but also results in a lower ohmic area-specific-resistance (ASR) of the bulk SE. However, although a lower exchange current density $i_{exc}$ ensures more stable deposition, it may result in a higher charge-transfer ASR at the interface. Figure 3d plots the interfacial charge-transfer ASR as a function of the exchange current density $i_{exc}$. The charge-transfer ASR increases dramatically when $i_{exc}$ is less than $50 \, mA/cm^2$. Notably, lots of work has focused on decreasing the interfacial ASR through interface modification (18, 44) in order to prevent dendrite formation, which may appear to contradict the conclusion in this work that higher charge-transfer ASR is expected to homogenize the current density. This apparent contradiction occurs because in most experimental measurements, it is hard to separate the charge-transfer ASR from the total interfacial ASR, which includes contact loss as well as interfacial overpotential, as discussed previously. We believe that in experiments, the decrease of interfacial ASR arises mainly from better interfacial contact. Our conclusion only applies to the part of the ASR that is controlled by the charge transfer.

Conventionally, the formation of a SEI (Figure 1b) between Li metal and the SE is believed to be detrimental to cell performance. One reason is that the ionic conductivity $\sigma_{Li^+}$ within the SEI is usually much lower than that in the bulk SE, leading to a higher ohmic ASR at the SEI and lower Li deposition stability (Figure 3c). However, there are scenarios under which the effectively lowered exchange current density $i_{exc}$ through SEI
formation may enhance depositional stability. Therefore, in determining the function of the SEI in a Li metal SSB, both the negative effect on the ASR and the beneficial effect on the depositional stability should be considered.

**Effect of stack pressure and yield strength on Li metal plasticity and SE fracture**

The unstable Li deposition discussed in the previous section may cause contact loss at interface $S$. Therefore, the application of a stack pressure $P_0$ is essential during cell cycling to maintain intimate contact. In principle, $P_0$ should be large enough to drive plastic/creep deformation of Li metal to smooth out the inhomogeneities caused by the uneven deposition. A wide range of yield strengths for Li, $\sigma_y$, from 0.6 to 50 MPa have been reported in the literature (45-47). Although Li hardening and creep behaviors of Li metal have been observed in some experiments (35, 36, 45), they are not included in the current study due to the lack of reliable constitutive measurements. Instead, perfect plasticity without hardening and with different $\sigma_y$ (ranging from 0.5 to 3 MPa) is assumed.

The effects of the stack pressure $P_0$ (ranging from 1 to 5 MPa) and the yield strength of Li metal $\sigma_y$ (ranging from 0.5 to 3 MPa) on Li deformation are investigated using the model shown in Figure 2a (defect width $w = 40\, nm$ and length $l = 200\, nm$). The existence of surface irregularities will cause local intensification of both the pressure $P$ and the von Mises stress $\tau$ at the interface $S$. The latter is the relevant quantity that determines the plastic yield of Li metal. Figure 4a and 4b show the distributions of interfacial pressure $P$ and von Mises stress $\tau$ of Li metal along interface $S$ near the Li protrusion for different stack pressures $P_0$ and a yield strength ($\sigma_y = 0.8\, MPa$) before any current is applied. As observed in Figure 4a and 4b, the interfacial pressure $P$ increases as the stack pressure $P_0$
increases, reaching a maximal value of $5.8 \text{ MPa}$ when the stack pressure is 3 MPa. The interfacial von Mises stress $\tau$ also increases but is bounded by the yield strength $\sigma_y$ ($0.8 \text{ MPa}$ in this case). The peak point of the surface irregularity has the largest von Mises stress $\tau$ and therefore reaches plasticity first. When a larger stack pressure $P_0$ is applied, the plastic region of Li metal grows from the peak area.

Figure 4c shows the distribution of the elastic/plastic regions of Li metal under a stack pressure $P_0 = 3 \text{ MPa}$ and Li yield strength $\sigma_y = 0.8 \text{ MPa}$. The elastic area is colored in light gray (labeled as region III in Figure 4c), and the plastic area is colored in dark blue (labeled as regions I & II). The peak area of the Li protrusion (region I) reaches plasticity due to stress intensification, which is consistent with the results in Figure 4b. It is more difficult for the top area of the Li protrusion (region III) to reach plasticity than the area far away from the protrusion (region II). This result stems from the hydrostatic component in region III being much higher than its deviatoric component because of the confining geometry, consistent with the plastic deformation of a material in a confined space (48). The degree of plasticity $\beta$ (the ratio of the plastically deformed area to the total area of Li metal) can be used to describe the plastic flow of Li metal. Figure 4d shows the effect of the stack pressure $P_0$ and yield strength $\sigma_y$ on $\beta$, with minimum value 0 (fully elastic) and maximum value 1 (fully plastic).

From Figure 4c, we can see that the location of Li plasticity may play a role in the cell performance and has not been previously discussed in other studies. In Figure 4c, region I and II reach plasticity while region III only elastically deforms. Note that the maximal elastic deformation for metals is typically very small and for Li metal is $\approx 0.01\%$ (36). The lack of plastic flow in region III is destructive in two ways. First, any voids/defects
developed in this region may not be filled in by Li plastic flow because of the limited deformation taking place. Second, the high hydrostatic pressure developed in this region will cause higher stress intensification in the Li protrusion peak, which may cause it to fracture the SE it is in contact with.

In Figure 4d, the degree of plasticity $\beta$ increases as the stack pressure $P_0$ increases or the yield strength $\sigma_y$ decreases. Considerable debate exists in the literature as to the yield stress of Li (35, 45, 46). To maintain a high enough plasticity (such as $\beta = 0.8$ shown in Figure 4d), the stack pressure $P_0$ must be increased accordingly if the yield strength of Li metal were to be larger than the correctly accepted macroscale value (0.8 $MPa$). For example, if the yield strength of Li can reach 50 $MPa$ (46), a stack pressure of 150 $MPa$ (extrapolated from the curve $\beta = 0.8$ shown in Figure 4d) is needed to drive enough Li deformation to remove effects of inhomogeneous current deposition. However, although this high stack pressure $P_0$ can be used to promote the Li metal deformation that is needed to maintain intimate contact under inhomogeneous plating, it can also result in fracture of the SE material. Such SE fracture under cell operation conditions has been observed in experimental studies (32). A detailed analysis of the fracture mechanisms of solid electrolytes is beyond the scope of this paper. By conducting an elementary study of the SE fracture based on classical fracture mechanics (49) we can assess the effects of stack pressure $P_0$ and the relative Young’s moduli of electrolyte and Li metal ($E_r = E_{SE}/E_{Li}$) on fracture, as summarized in Figure 5.

Figure 5a shows the stress intensity factor under mode I loading condition ($K_I$) as a function of the applied stack pressure $P_0$ and the Young’s moduli ratio $E_r$ for a yield strength of Li metal $\sigma_y = 0.8$ $MPa$. SE fracture will occur near the tip of defect and
propagate if the stress intensity factor ($K_I$) is larger than the fracture toughness ($K_{IC}$) of the SE material (49). According to the G-criterion, the stress intensity factor $K_I$ and the strain energy release rate ($G$) around the defect are related by: $K_I = \sqrt{\frac{G}{1-\nu^2}}$, $\nu$ is the Poisson’s ratio of the SE (49). The strain energy release rate ($G$) can be extracted from the stress state near the defect tip through the use of J-integral (50). The maximal allowed stack pressure $P_0$ to prevent SE fracture should satisfy the relation: $K_I(P_0, E_r) \leq K_{IC}$. For example, the fracture toughness of glassy LPS is $K_{IC,LPS} = 0.25 \text{ MPa} \cdot m^{1/2}$ (black lines shown in Figure 5a), which gives the maximal allowed stack pressure $P_0$ to be $6 \text{ MPa}$ with the relative Young’s modulus $E_r = \frac{E_{SE}}{E_{Li}} = 2$. In addition, as $E_r$ increases (either a stronger SE material such LLZO or a softer metal electrode such as Na-metal), the maximal allowed stack pressure $P_{0,max}$ will increase accordingly (as for LLZO shown with the red lines of Figure 5a, with $K_{IC,LLZO} = 1.25 \text{ MPa} \cdot m^{1/2}$).

Figure 5b shows the stress intensity factor $K_I$ of the SE as a function of the stack pressure $P_0$ and the yield strength of Li metal $\sigma_y$ while keeping the relative Young’s moduli $E_r = 2.5$. When the stack pressure $P_0$ is small ($<1.5 \text{ MPa}$), the stress intensity factor $K_I$ of the SE are almost identical for the three different yield strengths of Li metal. This is because when $P_0 < 1.5 \text{ MPa}$, Li metal undergoes fully elastic deformation (refer to Figure 4d), leading to similar mechanical responses of the contact pair. However, as the stack pressure $P_0$ grows larger than a critical value of $P_0$ (points $A_1, A_2$ and $A_3$ in Figure 5b), the stress intensity factor $K_I$ decreases first within a range of $P_0$ (between points $A$ and points $B$) and increases again after another critical value of $P_0$ (points $B_1, B_2$ in Figure 5b). This is due to the transition of Li metal deformation from fully elastic state ($\beta = 0$) to fully plastic state.
\( \beta = 1 \). Starting at points \( A \), Li metal starts to deform plastically near the tip area (Figure 4c), which will cause redistribution of interfacial stress (Figure 4b). The remaining elastic area in Li metal will accommodate more load than the tip area, leading to the decrease of the stress intensity factor \( K_f \) near the defect tip. Starting at points \( B \), Li metal undergoes fully plastic deformation and the interfacial stress distribution is again dominated by the geometric irregularity, similar to the situation during fully elastic deformation. Interfacial stress accumulates at the tip area and leads to the increase of the stress intensity factor \( K_f \) (as shown the blue and red lines after point \( B \) in Figure 5b).

**Coupled effect of Li electrodeposition and Li plasticity on contact loss**

Whether contact loss occurs at an interface is determined by the competition between the irregular Li deposition and the ability to displace Li metal under plastic deformation driven by the stack pressure \( P_0 \). In this section, both the mechanical and electro-deposition effects on interfacial contact loss are discussed. To quantify the contact loss, the degree of contact \( \gamma \) is defined as \( \gamma(t) = A_s(t)/A_s(0) \), where \( A_s(t) \) is the remaining interfacial contact area after \( t \) seconds and \( A_s(0) \) is the initial contact area. For the 2D model used in this work, the "contact area" is the contact length of interface \( S \).

A similar model as that in Figure 2a is built with defect width \( w = 40 \, nm \) and length \( l = 200 \, nm \), and the degree of contact \( \gamma \) at interface \( S \) is investigated for different values of the exchange current density \( i_{exc} \) \((30 - 160 \, mA/cm^2)\), applied total current density \( i_0 \) \((0.01 - 0.2 \, mA/cm^2)\), stack pressure \( P_0 \) \((1 - 5 \, MPa)\), and yield strength \( \sigma_y \) \((0.8 - 4 \, MPa)\), with the ionic conductivity \( \sigma_{Li^+} \) maintained at \( 0.3 \, mS/cm \). The thickness of Li deposited is calculated from the Faradaic relation \( H = \frac{V_{li}}{F} i_n \Delta t \), where \( H \) is the thickness
of newly deposited Li and $\Delta t$ is the deposition time. The simulation proceeds by depositing Li metal for a discrete timestep, followed by a mechanical equilibration after each timestep, including elastic and plastic deformation. (A convergence test is done on the selection of the timestep, as described in section SI-6). The process is shown in more detail in the flowchart in Figure SI-1b. A total deposition time of 50s (with average deposition thickness $\sim 1\text{nm}$) is conducted in all calculations with timestep $\Delta t = 10s$.

Figure 6a and Figure 6b show examples of the contact loss at interface $S$ with conditions: applied total current density $i_0 = 0.1\ mA/cm^2$, exchange current density $i_{\text{exc}} = 100\ mA/cm^2$, and yield strength of Li metal $\sigma_y = 0.8\ MPa$. When the stack pressure is low (such as $P_0 = 1\ MPa$ in Figure 6a), the deformation of Li metal is not sufficient to fill in the voids developed during the Li deposition. Voids near the edges of the defect develop, preventing further Li deposition in these areas as shown by the current density in Figure 6b after the voids develop. Once the SE and Li metal anode are separated in the void area, a sharp discontinuity in the current density $i_n$ is created. Higher pressure is able to reduce the contact loss, and when $P_0$ is 3 MPa no contact loss occurs during the limited time of the simulation.

Figure 6c shows the degree of contact $\gamma$ for different applied total current density $i_0$ and exchange current density $i_{\text{exc}}$ for a Li metal yield strength of $\sigma_y = 0.8\ MPa$ and stack pressure $P_0 = 2\ MPa$. It is clear that higher charge-transfer rate and larger current density leads to more contact loss, consistent with our earlier observation that large exchange current density magnifies the inhomogeneity in the current density arising from surface irregularity. For a low charge-transfer rate ($i_{\text{exc}} = 30\ mA/cm^2$), a higher charging rate ($i_0$ up to $0.2\ mA/cm^2$) can be tolerated to maintain physical contact ($\gamma = 1$). However,
approximately 30% of the contact is lost when exchange current density $i_{exc}$ increases to 160 $mA/cm^2$ and applied total current density $i_0$ increases to 0.2 $mA/cm^2$. The contact loss shown in Figure 6c originates from uneven electrodeposition because of the original defect geometry. In general, when the applied current density $i_0$ is too large (x-axis in Figure 6c) or the exchange current density is too large (curves from top to bottom), the deposition becomes more unstable, and more void space is developed.

Figure 6d shows the degree of contact $\gamma$ for various stack pressure $P_0$ and Li metal yield strength $\sigma_y$ with applied current density $i_0 = 0.1$ $mA/cm^2$ and exchange current density $i_{exc} = 100$ $mA/cm^2$. A low yield strength and a large stack pressure both help to prevent contact loss. When Li metal is soft ($\sigma_y = 0.8$ $MPa$), a small value of the stack pressure $P_0$ (~2.5 $MPa$) is sufficient to maintain perfect contact ($\gamma = 1$). However, approximately 6% of the contact will be lost when the yield strength of Li metal is 3 $MPa$ even if the stack pressure $P_0$ increases to 5 $MPa$. Therefore, the stack pressure $P_0$ needs to be well above the yield strength of Li metal $\sigma_y$ to maintain intimate contact near surface irregularities (As shown in Figure 6d, $P_0 > 2.2$ $MPa$ is needed if $\sigma_y = 0.8$ $MPa$, and $P_0 > 3.6$ $MPa$ is needed if $\sigma_y = 1.3$ $MPa$). Moreover, the degree of contact $\gamma$ decreases more rapidly as the yield strength $\sigma_y$ increases; such as for $\sigma_y = 1.8$ $MPa$, a minimum value of the stack pressure $P_0 = 5$ $MPa$ is needed maintain contact, which is close to the fracture limit (6 $MPa$) for LPS fracture (Figure 5a). The contact loss developed in Figure 6d mainly originates from insufficient mechanical deformation. In general, when the applied stack pressure is too low (x-axis in Figure 6d), or the yield strength of Li metal is high (curves from top to bottom), deformation of Li metal is not sufficient to smooth out the uneven deposition.
Discussion

Unstable Li deposition is believed to be one of the main sources for dendrite formation/propagation and interfacial contact loss. Both effects will amplify current inhomogeneity by either focusing current on a Li domain that is growing into the SE, or by redirecting current from the contact loss area to other areas where now the local current density needs to increase. As the internal stress of irregularly deposited Li accumulates (Figure 4a and Figure 4b), two different mechanisms can be triggered to release this internal stress: a) The deposited lithium is driven into pores or GBs of the SE, where the mechanical strength of the SE is weaker, initiating dendrite propagation; b) If the stack pressure is low enough, the excess internal stress in deposited Li can also be released by pushing the Li metal anode backward, resulting in interfacial contact loss (Figure 6). Since an ideal homogeneous current density distribution (with stability factor $\vartheta = 1$) is likely to be impossible due to the geometric and/or material inhomogeneities, it is more practical to minimize its detrimental consequences through an optimization of the material’s structure (such as surface geometry and pore connectivity of SE) and boundary conditions (such as the stack pressure or applied current density).

To increase deposition stability, both the geometry and the electrochemical properties of Li metal and SE are important: a) “Shallow and wide” surface defects are preferred (Figure 2d). Specifically, a decrease in the surface roughness by ten times results in an increase in deposition stability by approximately 20%. b) A higher ionic conductivity of the SE is always preferred for both decreasing the SE bulk resistance and increasing the deposition stability. For example, interfacial deposition can be 25 times more stable (stability factor $\vartheta$ decreases from 30 to 1.2 when the ionic conductivity $\sigma_{Li^+}$ increases from
0.003 mS/cm to 0.3 mS/cm (Figure 3b). c) For a given defect geometry, a relatively slow interfacial charge-transfer kinetics (low exchange current density) is preferred (Figure 3a) though this has to be kept within what is acceptable in terms of total interfacial resistance (Figure 3d). Increasing the ionic conductivity of the SE is clearly the most efficient way to stabilize the electrodeposition, as decreasing the exchange current density (Figure 3a) or preparing a well-polished SE surface (Figure 2d) only decrease $\theta$ by around 3 times and 1.5 times respectively. Therefore, screening for solid electrolyte with a high ionic conductivity is important for the performance of SSB, not only for lower SE resistance but also for more stable interfacial deposition. The reason the ionic conductivity is so important is that it effectively acts to screen surface irregularities. When inhomogeneous current develops near a defect, then through Ohm’s law, this also corresponds to inhomogeneities in the electric potential. When the conductor has high $\sigma_{t+t^+}$ it can spread this current laterally in the conductor more easily, thereby effectively screening the defect. This can be well observed in Figure 3b where for high enough ionic conductivity $\sigma_{t+t^+}$ the effect of the defect in the current density almost becomes invisible. Hence, we want to stress that high ionic conductivity in a solid-state electrolyte is not only important to lower the impedance of the full cell, but also plays an important role in screening the current inhomogeneities from defects near the charge transfer interface. One very different option to mitigate surface irregularity is to throttle the transport at or very near the interface. As evidenced in our calculations which show that a smaller exchange current leads to more homogeneous deposition and less void formation. While it seems unlikely that much can be done to change the actual charge transfer kinetics between Li metal and the conductor, a varying exchange current density can be mimicked by using a thin buffer layer. If this thin buffer layer has lower conductivity than the SE it will dampen irregular current coming
from the SE over some length scale. But a key requirement is that this buffer layer itself has good contact with the Li metal so that even deposition can occur from the buffer layer. If the buffer layer were to have worse contact with the Li anode, then deposition inhomogeneity would actually be aggravated by its application.

To make our discussion more general, a new quantity \( l_0 = \sigma_{Li^+} \frac{RT}{i_{exc}} \) is defined to reflect the combined effect of the bulk (ionic conductivity \( \sigma_{Li^+} \)) and the surface (exchange current density \( i_{exc} \)) of the SE on the Li deposition stability. Notably, the second term \( \frac{RT}{i_{exc}} \) equals the charge transfer area-specific resistance \( ASR \), when the Butler-Volmer relation (Equation 5) is linearized (when the overpotential \( \eta \) is small: \( \eta \ll \frac{RT}{F} \)). We call \( l_0 \) (with length unit) the damping length as it quantifies the ability of the SE to dampen deposition instability caused by surface irregularities. Figure 7a shows the deposition stability as a function of the defect length \( l \) and the damping length \( l_0 \). The dark blue, sky blue and yellow colors correspond to deposition stability factor \( \vartheta \leq 10 \), \( 10 < \vartheta < 20 \) and \( \vartheta \geq 20 \), respectively. The two horizontal white lines are the damping length \( l_0 \) of LPS (28 \( \mu m \), (38)) and LLZO (2 \( \mu m \), (44)). As previously discussed, the deposition is more stable when defect length decreases (Figure 2c and 2d) and damping length increases (exchange current density decreases in Figure 3a, or ionic conductivity increases in Figure 3b). An SE with large \( l_0 \) (such as LPS) creates more stable deposition than an SE with small \( l_0 \) (such as LLZO) when their surface roughness is similar. That is to say, an SE with larger \( l_0 \) can tolerate worse surface quality. LPS allows a defect length up to 4 \( \mu m \) while LLZO can only tolerate a value less than 1.4 \( \mu m \) if the deposition stability factor \( \vartheta \) has to be kept less than 20. Results in Figure 7a make it possible to evaluate the Li deposition stability at the Li metal/SE interface just from the material and geometry information of the SE.
Accumulation of internal stress near surface irregularities (Figure 4a and 4b) provides a driving force for Li deformation. Low stack pressure may result in insufficient Li deformation (Figure 4d) and cause interfacial contact loss (Figure 6a), while large stack pressure may lead to severe stress concentration (Figure 5) and cause Li infiltration into pores/GBs or even SE fracture if the stress intensity factor is above the limit of the fracture toughness of the SE. We find that in general, the stack pressure required to cause sufficient Li flow and maintain contact is well above the Li yield stress. For example, a minimal value of $2.2 \, MPa$ is needed to maintain intimate contact when the Li yield strength is $0.8 \, MPa$ (Figure 6d) and a maximal value of $6 \, MPa$ is allowed to prevent SE fracture against LPS (Figure 5a). Therefore, a “mechanical stability window” of the stack pressure ($[2.2 \, MPa, 6 \, MPa]$) is available to prevent both contact loss and SE fracture. Notably, this window would be modified if the Li metal yield strength is larger than $0.8 \, MPa$. This is because larger stack pressure is required to maintain intimate contact at higher Li metal yield strength (Figure 6d) while larger stack pressure is allowed for SE fracture at higher Li metal yield strength (Figure 5b). Therefore, a mechanical stability window for the stack pressure must be established for a Li-metal SSB system depending on the mechanical properties of both Li metal and the SE material.

Conclusions drawn above are also applicable to other metal SSB system (such as Sodium and Magnesium) as long as the metal electrode/SE interface is chemically stable. For solid-state battery system with different combination of metal electrodes and SEs, the required mechanical stability window for stack pressure can be very different. From the available material properties of electrodes and SEs in typical SSB systems (as shown in Table SI-1), we can extract the lower and upper limits of the stack pressure from Figure 6d and Figure 5b respectively. A summary of the mechanical stability window for the stack
pressure for typical SSB systems is shown in Figure 7b. Smaller stack pressure is needed in Na-metal system than in Li-metal system to maintain intimate contact. This is because Na-metal has a lower yield strength ($\sigma_y = 0.2\ MPa$) than Li-metal ($\sigma_y = 0.8\ MPa$). Oxide-SE systems can withstand much larger stack pressure than sulfide-SE systems because of their much larger moduli and fracture toughness (Table SI-1). Notably, results shown in Figure 7b assume no hardening of metal electrode and no degradation of the mechanical properties of the SE from voids/GBs.

**Conclusion**

In this work, the continuum theory with a two-dimensional model is employed to investigate the Li electrodeposition stability and mechanical stability at the Li metal/solid electrolyte interface in Li-metal solid-state battery. Through our analysis, we find that the Li deposition can be stabilized by increasing the ionic conductivity of the SE, decreasing the exchange current density of the interfacial charge-transfer reaction, and preparing a well-polished smooth SE surface. We show that LPS can tolerate worse surface quality than LLZO at the same deposition stability because LPS has much larger “damping length” than LLZO (28 µm vs. 2 µm). We also find that a “mechanical stability window” for the applied stack pressure is important for different SSB systems in order to prevent both contact loss and SE fracture. We conclude that cell performance of SSBs can be enhanced by a more comprehensive selection of SE materials, and a more careful control of applied stack pressure.
Supplementary information

Supplemental information can be found with the article online at XXXX.

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Author contributions

Q.T. planned the project with G.C.; Q.T. derived all equations, programmed all codes and calculated all data with the help from L.B. and T.S.; The manuscript was written by Q.T. and was revised by L.B., T.S. and G.C. with the help of the other authors. All authors contributed to discussions.

Declaration of interests

The authors declare no competing interests.
Figure 1. a) Schematic model of a half SSB cell consisting of a Li metal anode $\Omega_1$ and solid electrolyte $\Omega_0$. The roughness at the interface $S$ is approximated by the periodic protrusion defined in (c). The surfaces $S_0$ and $S_1$ are assumed to be flat. The left and right boundaries of Li metal and SE are labeled as $S_L$ and $S_R$, respectively. A stack pressure $P_0$ is applied to the ends of the cell ($S_0$ and $S_1$). Li ions are extracted from the cathode (not shown) and migrate from $S_0$ toward $S$ under galvanostatic conditions: $I_0 = i_0 \cdot A_0 = \int i_n \, dS$, where $i_0$ is the applied current density on $S_0$ with constant area $A_0$, and $i_n$ is the normal current density on $S$ ($i_n = \mathbf{i} \cdot \mathbf{n}$, where $\mathbf{i}$ is the current density in the SE and $\mathbf{n}$ is the normal direction of the interface $S$). Upon reaching $S$, these ions are plated onto $\Omega_1$ via the reaction $Li^+ + e^- \leftrightarrow Li$. b) Li deposition on the interface $S$ with normal current density $i_n$. A thin layer of SEI above $S$ is shown in yellow. c) Geometry of the contact pair of Li metal protrusion and SE defect on the interface $S$, with shape $y(x,t)$, width $2w$, and length $l$. 
Figure 2. a) Current density distribution in SE for a cosine-shaped defect with defect width $w = 40 \, nm$ and defect length $l = 100 \, nm$; the magnitude is represented by the contour line, and the direction is represented by the red arrows. b) Distribution of normal current density $i_n$ at interface $S$ within range $|x| < 0.2 \, \mu m$ for different shapes with $w = 40 \, nm$ and $l = 100 \, nm$. The inset shows $i_n$ within the range $|x| < 0.05 \, \mu m$. c) Surface overpotential $\eta$ near a defect with different length $l$ but same width $w = 40 \, nm$. d) Stability factor $\vartheta$ (contour surface) as a function of length $l$ and width $w$ for cosine-shaped defect.
Figure 3. Distribution of normal current density $i_n$ at interface $S$ near the defect with width $w = 40 \text{ nm}$ and $l = 100 \text{ nm}$: a) varying exchange current density $i_{exc}$ while keeping ionic conductivity $\sigma_{Li^+}$ constant ($0.3 \text{ mS/cm}$) and b) varying $\sigma_{Li^+}$ while keeping $i_{exc}$ constant ($100 \text{ mA/cm}^2$). c) Stability factor $\theta$ (color map) as a function of the exchange current density $i_{exc}$ and $\log(\sigma_{Li^+})$. d) Charge-transfer ASR as a function of exchange current density $i_{exc}$. All data at applied current density of 0.1mA/cm$^2$. 
Figure 4. Interfacial pressure (a) and von Mises stress (b) of Li metal along interface S near the defect for a Li metal yield strength $\sigma_y = 0.8 \, MPa$. (c) Elastic/plastic region of Li metal in contact with the SE (light gray indicates the elastic region, and dark blue indicates the plastic region), with the stack pressure $P_0 = 3 \, MPa$ and the yield strength $\sigma_y = 0.8 \, MPa$. The width and length of the Li protrusion (SE defect) are $l = 200 \, nm$ and $w = 40 \, nm$, respectively. (d) Degree of plasticity $\beta$ of Li metal as a function of the yield strength $\sigma_y$ (varying from 0.5 to 2.5 $MPa$), and the stack pressure $P_0$ (varying from 0.5 to 4.5 $MPa$).
Figure 5. (a) Stress intensity factor $K_I$ of SE near the defect tip as a function of the applied stack pressure $P_0$ at two different relative Young’s moduli $E_r$ ($E_r = E_{SE}/E_{Li}$, the ratio of Young’s moduli between SE and Li metal), with the yield strength $\sigma_y$ fixed at 0.8 MPa. The horizontal dashed lines represent the fracture toughness of LPS ($K_{IC,LPS} = 0.25 \text{ MPa} \cdot \text{m}^{1/2}$) and LLZO ($K_{IC,LLZO} = 1.25 \text{ MPa} \cdot \text{m}^{1/2}$) respectively. (b) Stress intensity factor $K_I$ as a function of the applied stack pressure $P_0$ at three yield strength of Li metal $\sigma_y$, with the relative Young’s moduli $E_{Li}$ fixed at 2.5.
Figure 6. (a) Loss of interfacial contact after a finite amount of Li deposition with applied total current density $i_0 = 0.1 \text{ mA/cm}^2$, stack pressure $P_0 = 1 \text{ MPa}$, and Li metal yield strength $\sigma_y = 0.8 \text{ MPa}$. (b) Current distribution near defect for the conditions in (a) but with three different stack pressure $P_0$. (c) Degree of contact for different values of exchange current density $i_{exc}$ and applied total current density $i_0$. (d) Degree of contact for different values of Li metal yield strength $\sigma_y$ and stack pressure $P_0$. 
Figure 7. (a) the deposition stability $\vartheta$ as a function of defect length $l$ and damping length $l_0$. The two black dash lines ($\vartheta = 10$ and $\vartheta = 20$) separate the deposition stability into three regions: dark blue ($\vartheta \leq 10$), light blue ($10 < \vartheta < 20$), and yellow ($\vartheta \geq 20$). The two white lines represents the values of damping length for LPS and LLZO respectively.

(b) The mechanical stability window of the stack pressure for different SSB systems. The lower limit in each window is the minimal value of stack pressure needed to maintain intimate contact of the metal electrode and SE. The upper limit is the maximal value allowed to prevent SE fracture.
Table 1. Critical parameters used in this work

| NAME                                | SYMBOL | UNIT       | VALUE | REF  |
|-------------------------------------|--------|------------|-------|------|
| **Mechanical Parameters**            |        |            |       |      |
| Young’s modulus of Li metal          | $E_{Li}$ | GPa        | 7.8   | (36) |
| Poisson’s ratio of Li metal          | $\nu_{Li}$ | 1          | 0.38  | (36) |
| Density of Li metal                  | $\rho_{Li}$ | kg/m$^3$  | 534   | (36) |
| Yield strength of Li metal           | $\sigma_0$ | MPa        | 0.8   | (36) |
| Young’s modulus of SE                | $E_{SE}$ | GPa        | 19.5  | (31) |
| Density of SE                        | $\rho_{SE}$ | kg/m$^3$  | 1880  | (31) |
| Poisson’s ratio of SE                | $\nu_{SE}$ | 1          | 0.36  | (31) |
| Fracture toughness of SE             | $K_{IC}$ | MPa·m$^{1/2}$ | 0.25 | (31) |
| **Electrochemical Parameters**       |        |            |       |      |
| Anodic/cathodic transfer coefficients | $\alpha_a, \alpha_c$ | 1       | 0.5   | (34) |
| Exchange current density             | $i_{exc}$ | mA/cm$^2$ | 100   | (38) |
| Li$^+$ conductivity in SE            | $\sigma_{Li^+}$ | mS/cm   | 0.3   | (39) |
| Applied current density              | $i_0$ | mA/cm$^2$ | 0.1   | (8)  |
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