



# Solid Electrolyte Interphase Growth in Lithium Metal Cells With Normal Electrolyte Flow

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In high energy density lithium metal batteries (LMBs), dendrite and solid electrolyte interphase (SEI) growth reduce safety and longevity, respectively. A stable SEI layer enables high efficiency cycling but continued SEI growth can lead to reduced capacity and coulombic efficiency. In this paper, we develop a steady-state model that predicts the effect of small advective electrolyte flow towards the lithium metal electrode on SEI growth during charging. For a fixed current density, increasing the electrolyte flow rate improves the coulombic efficiency and decreases SEI layer growth rate. Decreasing the charging current density at a constant flow rate also decreases the SEI layer growth rate. Low flow rates ( $\mu\text{m/s}$ ) can increase coulombic efficiency by up to 6%. The sensitivity of the coulombic efficiency to plating and SEI layer reaction rates is also explored.

**Keywords:** solid electrolyte interphase, lithium metal cells, normal electrolyte flow, coulombic efficiency, dendrite

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## 1 INTRODUCTION

Lithium metal batteries are promising, energy-dense next generation batteries (Whittingham, 2012). Electroplating during charging, however can lead to dendrite growth on the metal electrodes. Moreover, due to their high reactivity, metal electrodes often react instantaneously with the electrolyte to form a solid electrolyte interphase (SEI) (Peled, 1979). Stripping Li from under the SEI during discharging can lead to the formation of nanovoids. Aggregation of nanovoids at high discharge rates can lead to the collapse of the SEI layer (pitting), exposing fresh Li surfaces and consuming further Li (Shi et al., 2018) in SEI generation. These inhomogeneities in the Li metal surface formed due to repeated stripping and pitting produce surface irregularities that promote dendrite growth. Discharging at high current can lead to rapid SEI growth and increased impedance, thus making fast charging difficult. This mechanism is the root cause of failure in Li metal batteries (Lu et al., 2015). Cracking of the SEI around dendrite tips due to excessive stress also promotes dendrite growth (Liu and Lu, 2017). A mechanically tough SEI may withstand stress, reduce dendrite growth, and prevent consumption of Li during repeated cycling as fresh Li surfaces are not exposed to the electrolyte if the SEI layer does not break (Aurbach, 2000). SEI layer growth has been studied extensively in theoretical, computational, first-principles modeling, and experimental studies (Jiang et al., 2020).

Modeling plays a key role in understanding the SEI growth mechanism. The model based on electron-tunneling proposed by Peled (1979) is one of the pioneering works in this field. Broussely et al. (2001) propose a growth model limited by electronic conductivity of the SEI layer and link SEI layer growth to capacity degradation and aging in Li ion batteries. Ploehn et al. (2004) propose a growth mechanism dominated by solvent diffusion through the SEI layer. Christensen and Newman (2004) derive a mathematical model for SEI layer growth and investigate the dependence of SEI layer

growth on battery voltage. They argue that the characteristics and chemical composition of SEI layers on lithium and graphite electrodes are similar. Pinson and Bazant (2012) derive the square root dependence of long term SEI layer growth on time using single particle and porous electrode models and a solvent diffusion limited growth mechanism. Their models fit the experimental data obtained by Broussely et al. (2001), Smith et al. (2011). The long term square root dependence may also be derived by using growth mechanisms associated with electronic conduction or neutral lithium interstitial diffusion through the SEI layer Single et al., 2018. A linear dependence of SEI growth with time results from linearization of the single particle model and SEI growth rate kinetics (Tanim and Rahn, 2015). Single et al. (2018) show that a growth mechanism limited by diffusion of lithium interstitials through the SEI can reproduce the potential dependence of long term SEI growth. With increasing SEI porosity, the SEI growth mechanism transitions from electronic conduction limited growth to solvent diffusion limited growth. Attia et al. (2019) experimentally show that even within the same charge-discharge cycle the SEI layer growth is faster while charging than during discharging. Das et al. (2019) build a theoretical model based on a mixed ionic electronic conductor SEI to explain these experimental results. The SEI may act as an electronic conductor, an ionic conductor, or a mixed electronic-ionic conductor or allow lithium interstitials to diffuse through or allows all of these phenomena. The mechanistic origin of the limiting mechanism of SEI growth is unclear.

Experimentally SEI evolution has mostly been studied via electrochemical impedance spectroscopy (Lu et al., 2014). Researchers have also studied SEI chemistry using various techniques such as XPS and FT-IR (Verma et al., 2010; Fan et al., 2018a). It is difficult to determine the SEI composition because it contains many compounds and functional groups. The composition also depends on the surface finish, electrolyte, cycling rates, temperatures, etc., complicating experimental assessment of the SEI (Single et al., 2017). Calendar aging of lithium batteries occurs due to the chemical SEI formation and the effect of temperature and SOC on calendar aging has been studied experimentally by Keil et al. (2016), Keil and Jossen (2016). SEI also grows electrochemically during cell cycling. Factors such as potential, cycle number, and charging/discharging rate have been experimentally shown to govern the electrochemical SEI formation. Even at the same voltage, SEI formation rate is greater for the charging half cycle than for the discharging half cycle Attia et al., 2019.

Kim et al. (2011) study the effect of electrolyte composition on structure and evolution of SEI via molecular dynamics simulations. Bertolini and Balbuena (2018) use reactive molecular dynamics simulations to study the initial stages of formation of SEI on Li metal electrodes in the absence of a bias potential. They identify a porous phase in contact with the Li metal electrode, a nest phase (amorphous matrix of Li atoms separated by nanochannels which allow electrolyte molecules and decomposition products to diffuse through), and a disperse phase in the SEI (layer in contact with the electrolyte) and propose that uneven Li distribution in the nest and dispersed

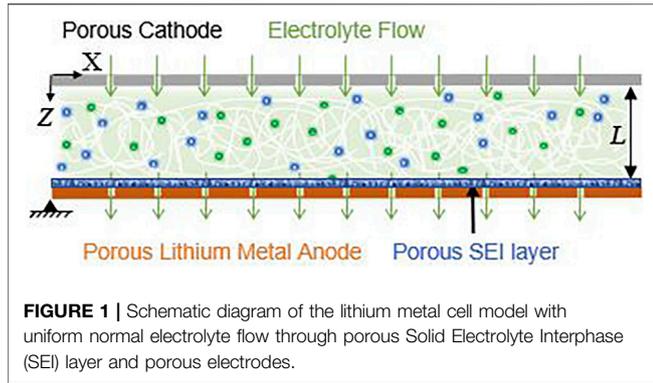
phases may be responsible for uneven electric field and dendrite growth.

The SEI layer often contains both organic and inorganic products (Aurbach, 2000; Michan et al., 2016) and its composition depends upon electrolyte composition (Matsuda, 1993; Aurbach and Zaban, 1993), temperature (Ishikawa et al., 2001) and charge-discharge current (Dolle et al., 2001). A stable SEI is thought to prevent dendrite growth, so researchers use SEI forming additives such as vinylene carbonate (Ota et al., 2004; Aurbach et al., 2002; El Ouatani et al., 2008) triacetoxymethylsilane (Lee et al., 2007), fluoroethylene carbonate (Heine et al., 2015), organosulphides (Li et al., 2017), aluminium fluoride (for cathode electrolyte interphase) (Zhao et al., 2020) in the electrolyte and grow artificial SEI layers *in-situ* (Li et al., 2016; Pathak et al., 2020) and *ex-situ* (Wang et al., 2019; Gao et al., 2019; Ju et al., 2020). Indirect strategies to stabilize the SEI include changing electrolyte concentration (Fan X. et al., 2018; Wang et al., 2018) and modifying the host (Cui et al., 2020; Liu et al., 2020) and separator (Huo et al., 2020; Liang et al., 2020) materials. Reported coulombic efficiencies range from 92% (Ma et al., 2017) to 99.1% (Gao et al., 2019).

The emerging area of flowing electrolyte metal batteries (FEMBs) has demonstrated dendrite suppression both theoretically (Parekh et al., 2020; Parekh and Rahn, 2020a; Parekh and Rahn, 2020b) and experimentally (Huang et al., 2020). In Parekh et al. (2020), Parekh M. and Rahn C. (2020), creeping flow normal to the lithium metal electrode and above a critical speed can theoretically eliminate dendrite growth. Creeping flow parallel to the lithium metal electrode can also reduce dendrite growth (Parekh M. N. and Rahn C. D., 2020). Dendrite and SEI layer growth in LMBs are inherently coupled. Our earlier efforts (Parekh et al., 2020; Parekh M. N. and Rahn C. D., 2020; Parekh M. and Rahn C., 2020) focused on effects of advection on dendrite growth and neglected SEI growth. This paper develops the first steady-state model of a lithium metal electrode with normal electrolyte flow that includes solvent diffusion and SEI layer growth kinetics. The model predicts the current distribution between plating and SEI layer growth and also identifies the reaction parameters that may be required to achieve high coulombic efficiencies. To the best of authors' knowledge, there has been no previous experimental study on the effect of electrolyte flow on SEI growth and the authors intend to explore that in the future.

## 2 GOVERNING EQUATIONS

**Figure 1** shows a schematic diagram of the Li metal cell with electrolyte flow normal to the lithium metal electrode. We assume that a charging current density  $J_{tot}$  leads to a flux  $N_c$  which transports  $Li^+$  ions from the positive electrode at  $Z = 0$  to the Li metal electrode at  $Z = L$  where they may either plate or be consumed to generate SEI. We also assume the electrodes to be perforated or porous (e.g., metal foam) with sufficiently fine scaled pore structure to allow uniform normal electrolyte flow. The SEI layer on the porous electrodes is also assumed to be porous and much thinner than the inter-electrode gap. This



means that we do not account for the temporal evolution of SEI thickness and also do not account for self passivating behavior of the SEI (*i.e.*, neglect solvent, ion and lithium interstitial diffusion and electronic conduction through SEI). With a flowing electrolyte and a porous SEI, advective transport in the bulk electrolyte is assumed to be the dominant mechanism governing SEI growth.

Within the electrolyte (neglecting the double layer region)

$$0 = -\nabla \cdot \mathbf{N}_c, \quad (1)$$

$$0 = D_c \frac{\partial^2 C_c}{\partial Z^2} + \mu_c F C_c \frac{\partial^2 \Phi}{\partial Z^2} + \mu_c F \frac{\partial C_c}{\partial Z} \frac{\partial \Phi}{\partial Z} - v \frac{\partial C_c}{\partial Z}, \quad (2)$$

$$0 = D_a \frac{\partial^2 C_a}{\partial Z^2} - \mu_a F C_a \frac{\partial^2 \Phi}{\partial Z^2} - \mu_a F \frac{\partial C_a}{\partial Z} \frac{\partial \Phi}{\partial Z} - v \frac{\partial C_a}{\partial Z}, \quad (3)$$

where  $F$  is Faraday's constant,  $D$  is diffusivity,  $\mu$  is electric mobility,  $\mathbf{N}_c$  is the solute flux,  $C(Z)$  is solute (ion) concentration,  $\mathbf{v} = v\hat{\mathbf{k}}$  is the electrolyte velocity, and  $\Phi(Z)$  is electrostatic potential. The subscripts  $c$  and  $a$  indicate cation and anion respectively. Electroneutrality requires

$$C_c = C_a = C. \quad (4)$$

Using Eqs 2–4 gives

$$0 = D \frac{\partial^2 C}{\partial Z^2} - v \frac{\partial C}{\partial Z}, \quad (5)$$

where  $D = \frac{2D_c D_a}{D_c + D_a}$  is the ambipolar diffusivity. Comparing Eqs 1, 5 gives

$$\mathbf{N}_c = \left( -D \frac{\partial C}{\partial Z} + vC \right) \hat{\mathbf{k}}. \quad (6)$$

Solvent transport is governed by

$$0 = -\nabla \cdot \mathbf{N}_s, \quad (7)$$

$$\mathbf{N}_s = \left( -D_s \frac{\partial C_s}{\partial Z} + vC_s \right) \hat{\mathbf{k}} \quad (8)$$

where  $\mathbf{N}_s$  is the solvent flux,  $C_s$  is the concentration of the solvent, and  $D_s$  is the solvent diffusivity. The total charging current density is divided between plating current density  $J_p$  and SEI formation current density  $J_{sei}$ , so

$$J_{tot} = J_{sei} + J_p, \quad (9)$$

where the current densities are given by the Butler-Volmer equations

$$J_p = FN_c(L) \cdot \hat{\mathbf{k}} \quad (10)$$

$$J_p = K_p C_c(L, T)^{1-\alpha} \left[ \exp\left(\frac{-\alpha FN_p}{RT_0}\right) - \exp\left(\frac{\alpha FN_p}{RT_0}\right) \right] \quad (11)$$

and

$$J_{sei} = FN_s(L) \cdot \hat{\mathbf{k}} \quad (12)$$

$$J_{sei} = K_{sei} [C_c(L)C_s(L)]^{1-\alpha} \left[ \exp\left(\frac{-\alpha FN_{sei}}{RT_0}\right) - \exp\left(\frac{\alpha FN_{sei}}{RT_0}\right) \right] \quad (13)$$

where  $N_p$  and  $N_{sei}$  are the activation overpotentials for plating and SEI formation,  $K_p$  and  $K_{sei}$  are the pre-exponential factors for plating and SEI formation, respectively,  $\alpha$  is the symmetry factor, and  $T_0$  is the temperature.

Non dimensional variables are defined as,  $j_{tot} = \frac{J_{tot}L}{FD_c C_0}$ ,  $d = \frac{D}{D_c}$ ,  $d_s = \frac{D_s}{D_c}$ ,  $Pe = \frac{vL}{D_c}$ ,  $\phi = \frac{\Phi F}{RT_0}$ ,  $c_c = \frac{C_c}{C_0}$ ,  $c_a = \frac{C_a}{C_0}$ ,  $c_s = \frac{C_s}{C_0}$ ,  $z = \frac{Z}{L}$ ,  $\mathbf{n}_c = \frac{\mathbf{N}_c L}{D_c C_0}$ ,  $\mathbf{n}_s = \frac{\mathbf{N}_s L}{D_c C_0}$ ,  $j_{sei} = \frac{J_{sei} L}{FD_c C_0}$ ,  $j_p = \frac{J_p L}{FD_c C_0}$ ,  $\eta_p = \frac{FN_p}{RT_0}$ ,  $\eta_{sei} = \frac{FN_{sei}}{RT_0}$ ,  $k_p = \frac{K_p L}{FD_c C_0^{0.5}}$ , and  $k_{sei} = \frac{K_{sei} L}{FD_c}$  where  $C_0$  and  $C_{0s}$  are the average solute and solvent concentrations, respectively. Substituting these variables in Eqs 5–13 produces

$$0 = \frac{\partial^2 c_c}{\partial z^2} + c_c \frac{\partial^2 \phi}{\partial z^2} + \frac{\partial c_c}{\partial z} \frac{\partial \phi}{\partial z} - Pe \frac{\partial c_c}{\partial z} \quad (14)$$

$$0 = \frac{\partial^2 c_a}{\partial z^2} - c_a \frac{\partial^2 \phi}{\partial z^2} - \frac{\partial c_a}{\partial z} \frac{\partial \phi}{\partial z} - Pe \frac{D_c}{D_a} \frac{\partial c_a}{\partial z} \quad (15)$$

$$c_c = c_a = c, \quad (16)$$

$$0 = d \frac{\partial^2 c}{\partial z^2} - Pe \frac{\partial c}{\partial z}, \quad (17)$$

$$\mathbf{n}_c = \left( -d \frac{\partial c}{\partial z} + Pec \right) \hat{\mathbf{k}}, \quad (18)$$

$$0 = d_s \frac{\partial^2 c_s}{\partial z^2} - Pe \frac{\partial c_s}{\partial z}, \quad (19)$$

$$\mathbf{n}_s = \left( -d_s \frac{\partial c_s}{\partial z} + Pec_s \right) \hat{\mathbf{k}}, \quad (20)$$

$$j_{tot} = j_p + j_{sei}, \quad (21)$$

$$j_p = k_p c_c (1)^{1-\alpha} \left[ \exp(-\alpha \eta_p) - \exp((1-\alpha)\eta_p) \right], \quad (22)$$

$$j_{sei} = k_{sei} c_c (1)^{1-\alpha} c_s (1)^{1-\alpha} \left[ \exp(-\alpha \eta_{sei}) - \exp((1-\alpha)\eta_{sei}) \right]. \quad (23)$$

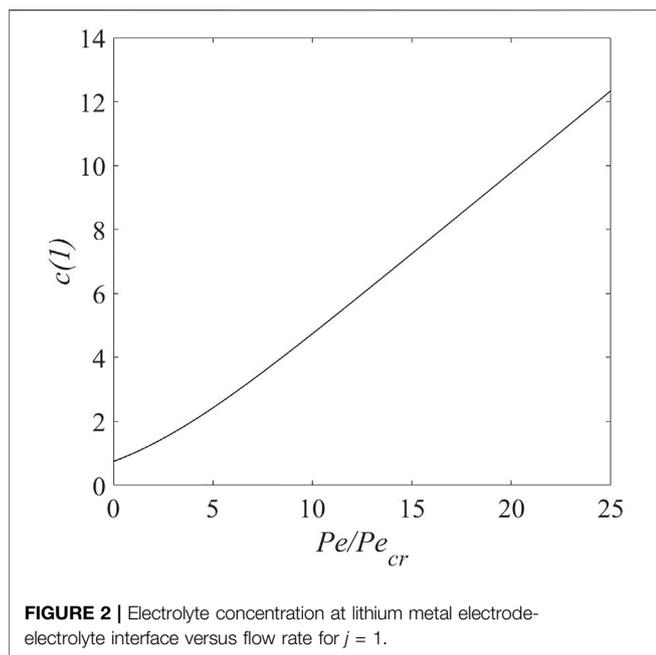
The equilibrium potential for SEI formation is assumed to be 0.8 V (Harris and Lu, 2013). Also, the plating and SEI formation reactions are assumed to be happening parallelly. So,

$$\frac{RT_0}{F} \ln(C_c(L)) + N_p = 0.8 + N_{sei}. \quad (24)$$

Non-dimensionalizing Eq. 24 gives

$$\eta_p + \ln(c(1)) + 6.909 = \eta_{sei} + 0.8F/(RT_0). \quad (25)$$

The nondimensional model given by Eqs 14–25 differs from those in previous work (Parekh et al., 2020; Parekh M. and Rahn



C., 2020; Parekh M. N. and Rahn C. D., 2020) in two important ways. The solvent transport (Eq. 19) and the SEI layer kinetics (Eq. 23) have been added.

We use Eqs 18, 20 as boundary conditions at both  $z = 0$  and  $z = 1$ . Using these boundary conditions, Eqs 17, 19, 21–23 and 25 are solved analytically to produce

$$c = \frac{j_{tot}}{Pe} + \left[ c_{a0} - \frac{j_{tot}}{Pe} \right] \exp(zPe/d), \quad (26)$$

$$c_{a0} = \frac{j_{tot}}{Pe} + \frac{\left[ 1 - \frac{j_{tot}}{Pe} \right] [Pe/d]}{\exp(Pe/d) - 1}, \quad (27)$$

$$c_s = \frac{j_{sei}}{Pe} + \left[ c_{sa0} - \frac{j_{sei}}{Pe} \right] \exp(zPe/d_s), \quad (28)$$

$$c_{sa0} = \frac{j_{sei}}{Pe} + \frac{\left[ 1 - \frac{j_{sei}}{Pe} \right] [Pe/d_s]}{\exp(Pe/d_s) - 1}. \quad (29)$$

### 3 RESULTS

The critical Peclet number,  $Pe_{cr}$ , is defined as the Peclet number at which advective flux equals the total ionic flux at the negative electrode. Our previous analysis (Parekh et al., 2020) shows that at steady state, critical flow rate or  $Pe = Pe_{cr}$  leads to an almost uniform concentration profile given by  $c = 1$  and hence we use that to calculate  $Pe_{cr}$ .

Based on our previous results (Parekh et al., 2020), we expect the concentration gradient and electrostatic potential gradient to reduce in magnitude with increasing flow rate and then the gradients change sign above the critical flow rate. Figure 2 shows that the electrolyte concentration at the lithium metal electrode surface increases with increasing flow rate. Based on Eqs 26–29, the solvent concentration is expected to follow a

**TABLE 1** | Model parameters.

Property	Value
$C_0$	1 M
$C_{0s}$	4.5 M Single et al. (2017)
$T_0$	300 K
$L$	50 $\mu\text{m}$
$D_a$	$4E - 10 \text{ m}^2\text{s}^{-1}$ Tikekar et al. (2014)
$D_c$	$10^{-11} \text{ m}^2\text{s}^{-1}$ (Akolkar (2014))
$D_s$	$10^{-10} - 10^{-8} \text{ m}^2\text{s}^{-1}$
$F$	96,500 $\text{Cmol}^{-1}$
$R$	8.314 $\text{Jmol}^{-1}\text{K}^{-1}$
$\mu_c$	$D_c/(RT_0) \text{ mols}^{-1}\text{N}^{-1}$
$\mu_a$	$D_a/(RT_0) \text{ mols}^{-1}\text{N}^{-1}$
$\alpha$	0.5 Das et al. (2019)
$K_p$	$1.1E - 2 \text{ Am}^{-2}\text{mol}^{-0.5}\text{m}^{1.5}$ Das et al. (2019)
$K_{sei}$	$2.5E - 9/C_{0s}^{0.5} \text{ Am}^{-2}\text{mol}^{-1}\text{m}^3$ Das et al. (2019)

similar pattern. It is important to note this trend because concentration of both electrolyte and solvent is expected to have an effect on SEI kinetics as per Eqs (22)–(25).

Assuming SEI layer formation and dead Li are the only Li sinks other than plating, coulombic efficiency (CE) can be defined as

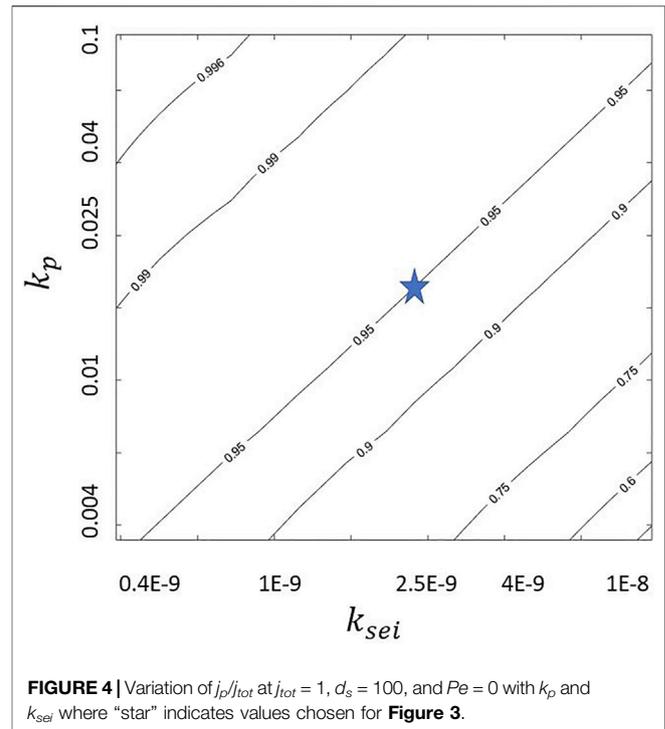
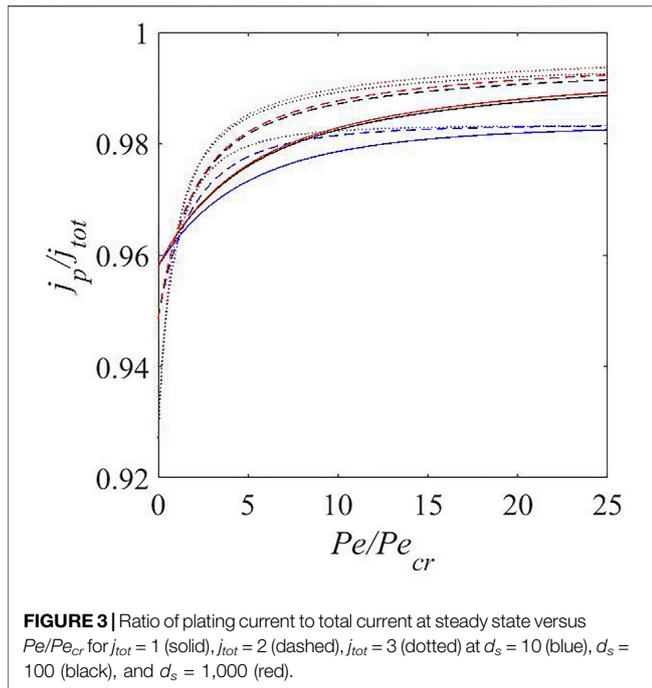
$$CE = (DC/CC) = (1 - DL/CC - SEIC/CC), \quad (30)$$

where  $DC$  is discharge capacity,  $CC$  is charge capacity,  $SEIC$  is the charge capacity lost in SEI formation, and  $DL$  is the amount of dead Li. Flow rates above  $Pe_{cr}$  eliminate dendrites (Parekh et al., 2020). So we assume that dead Li is eliminated under these conditions, and

$$CE = (DC/CC) = (1 - SEIC/CC) = j_p/j_{tot}. \quad (31)$$

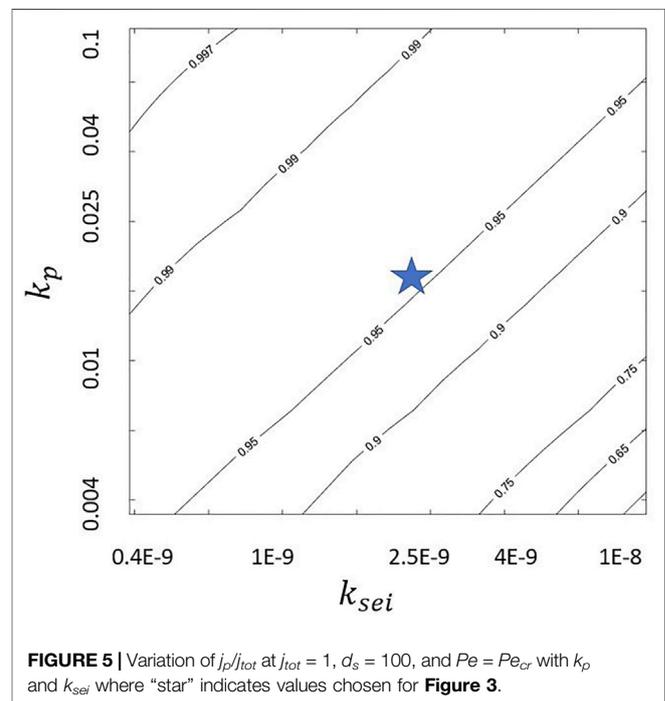
Thus, increasing  $j_p/j_{tot}$  results in increased coulombic efficiency and cycle life. For typical charging current densities,  $Pe_{cr}$  is on the order of 1, so the dimensional critical velocity is on the order of  $D_c/L$ . For  $j = 1$  and parameters in Table 1, the critical velocity is on the order of  $\mu\text{m s}^{-1}$ . Thus, the required flow rates are very low and may be practically achieved using a variety of low power microfluidic pumping systems (Iverson and Garimella, 2008) along with porous or micropatterned electrodes (Park et al., 2016; Ryou et al., 2015; Wang et al., 2017). Programmed squeezing of a battery pack composed of pouch cells is another way to achieve the desired electrolyte flow.

Figure 3 shows that increasing the flow rate increases the  $j_p/j_{tot}$  ratio. At  $Pe = 15Pe_{cr}$ ,  $d_s = 100$  and  $j = 3$ , coulombic efficiency of 99.3% is predicted. This is 6% higher than without electrolyte flow. Figure 3 shows that at low flow rates (including  $Pe = 0$ ), a high charging current density leads to a lower coulombic efficiency. This is similar to the experimental observations (Attia et al., 2019; Madani et al., 2019) and other models (Lu et al., 2015; Das et al., 2019) in literature. Most models (Ploehn et al., 2004; Pinson and Bazant, 2012; Das et al., 2019) in literature account for the SEI layer thickness evolution with time. We, however, assume the SEI layer thickness to be negligible with respect to the inter-electrode gap and hence do not account for the temporal evolution of its thickness. SEI layer thickness becomes important in cases where diffusion is the main ion

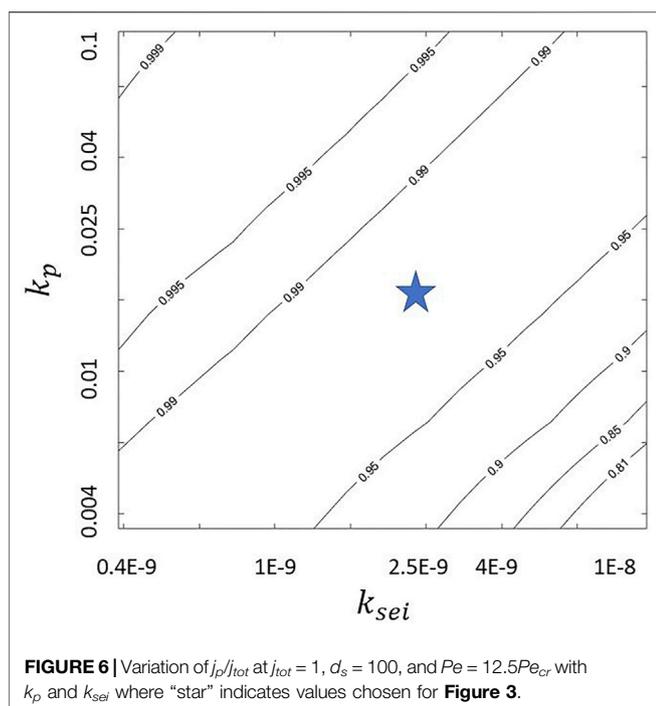


transport mechanism as a thicker SEI layer acts as a self passivating layer by reducing ion and solvent diffusivity. However, for a flowing electrolyte, the main ion transport and solvent transport mechanism is assumed to be advective transport and we do not expect our results to get affected drastically by the SEI layer thickness. At higher flow rates, the coulombic efficiencies flatten with respect to increasing charging current density and increasing flow rates. Under these conditions, lithium ions concentrate near the lithium metal electrode and increasing flow rate leads to a marginal increase in lithium ions available near the lithium metal electrode. Current literature values for coulombic efficiency in lithium metal cells mostly lie between 92% (Ma et al., 2017)—99.1% (Gao et al., 2019). Electrolyte flow accelerates both solvent and ion advective transport, so one might expect equal effects on both. Solvent diffusivity, however, is typically much higher than ionic diffusivity. Thus, ionic transport benefits more from advective transport leading to an increased CE due to an increase in  $j_p$  and reduction in  $j_{sei}$ . The fact that increasing the flow rate increases the CE is valid for all the three different  $d_s$  values used in **Figure 3**. This indicates that the increase in CE with flow rates is not an artifact of the chosen solvent diffusivity.

The predicted CE values in **Figure 3** depends on the model parameters. We explore the sensitivity of CE to plating and SEI reaction constants. The nondimensional  $k_p = 0.018$  and  $k_{sei} = 1.93E - 9$  from literature (Das et al. (2019)) are varied by a factor of 5 in both directions. **Figures 4–6** show the variation of  $j_p/j_{tot}$  with  $k_p$  and  $k_{sei}$  values. They show that a higher  $k_p$  and a lower  $k_{sei}$  imply low SEI growth rate for galvanostatic charging. This is probably because higher  $k_p$  values and lower  $k_{sei}$  values imply higher and lower reaction rates for plating and SEI formation, respectively. This is also evident from **Eqs 21–23**. Moreover, a



comparison of  $j_p/j_{tot}$  at same  $k_p$  and  $k_{sei}$  values indicates that a higher flow rate leads to higher coulombic efficiency. Finally, the top left corner of **Figure 6** shows coulombic efficiencies more than 99.9% for  $k_p \approx 0.1$  and  $k_{sei} \approx 0.4E-9$  with higher  $k_p$  values requiring higher  $k_{sei}$  values for the same coulombic efficiency. This may enable practical applications with about 225 cycles



(even when the self-passivating nature of SEI is excluded and excess lithium present in the lithium metal anode is not accounted for in the model). These figures may be used as a guideline to tune the flow rate and electrolyte/solvent (rate constants) so as to obtain practical lithium metal batteries. A comparison of **Figures 4, 6** shows that just tuning the electrolyte parameters within the explored ranges will not allow us to reach the high coulombic efficiency of 99.9%. It is necessary to tune the electrolyte/solvent and also use normal electrolyte flow to achieve high coulombic efficiencies.

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## 4 CONCLUSION

A small amount of electrolyte flow towards the metal electrode of a LMB during charging can produce remarkable results. As discussed in previous work (Parekh et al., 2020), electrostatic potential decreases and dendrites disappear, making LMBs amenable to fast charging and safe operation. This paper shows for the first time that flow reduces SEI layer formation and increases coulombic efficiency. The critical flow rate is a key parameter in the design of FEMBs. It is the lowest flow rate that suppresses dendrites. Tuning flow rates and electrolyte parameters such as rate constants can enable fast charging, safe, dendrite-free, long-lasting lithium metal batteries.

## DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

## AUTHOR CONTRIBUTIONS

MNP is the first author and he built the model, generated the results, and wrote the manuscript. CDR is the last author and he guided MNP during the project, and edited the manuscript.

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