

Flexible lithium metal anode featuring ultrahigh current density stability with uniform deposition and dissolution

Ahmed M. Hafez, Jinzhi Sheng, Daxian Cao, Yong Chen, Hongli Zhu*

Department of Mechanical and Industrial Engineering, Northeastern University, Boston, MA 02115, United States.

E-mail: h.zhu@neu.edu

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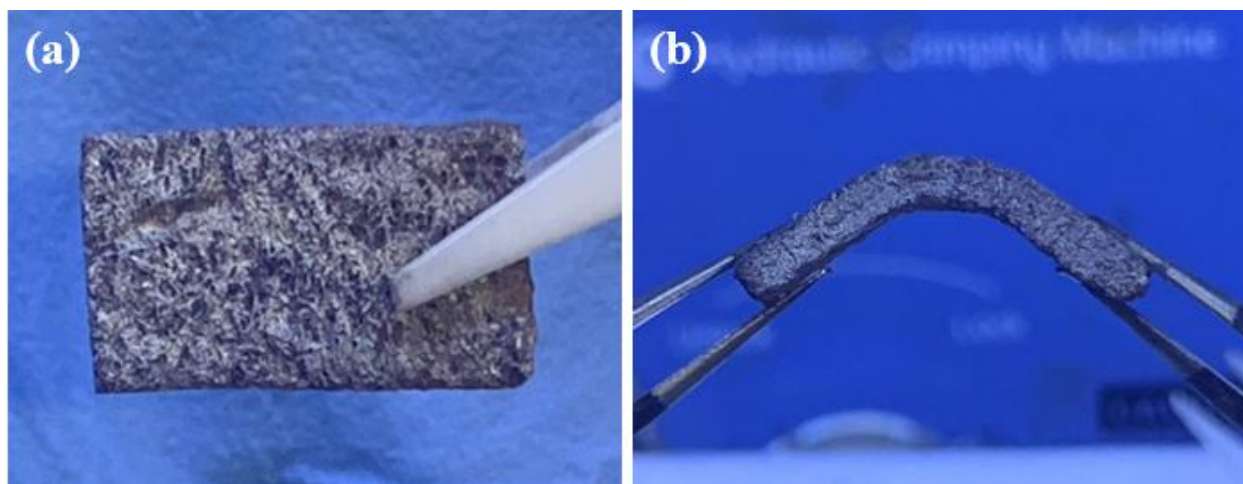


Figure S1. Snapshots for the Li felt after Li infiltration. (a) Snapshot for the Li felt surface. (b) Snapshot for the side view of bended Li felt.

1- The Multiphysics model definition:

The Multiphysics model demonstrated in this study is defined by balancing Li^+ cations and TFSI anions species through the electrolyte assuming electroneutrality conditions. Which gives

three dependent variables throughout the simulation, these are, Li ions concentration, TFSI ions concentration, and the change in ionic potential. These variables are tracked with time to probe the Li growth and dissolution at the electrode surface as well as the morphological deformations at the surface. Herein, we assumed there is no parasitic side reactions occur at the electrodes, and only Li deposition and dissolution takes place at the cathode and anode sides, respectively. On the other hand, the convection through the electrolyte due to the electrolyte density variations with cycling is assumed to be small and neglected. In this model also, we used a 2D cross-section structure to represent the coin cell, with an electrode strip of 90 μm in length (x-axis) for both the anode and the cathode, and 25 μm separation between electrodes, which is close to the real separation dimension using Celgard separators. For simplicity, we have represented the Li felt cross-section by set of spherical Li metal spheres with 14 μm diameter each, which together form an electrode with equivalent length to their Li foil electrode counterpart.

2- Computational model governing equations:

The flux for each of the ions in the electrolyte is given by the Nernst-Planck equation:

$$N_i = -D_i \nabla c_i - z_i u_i F c_i \nabla \phi_l$$

Where N_i is the transport vector, c_i is the species concentration in the electrolyte, z_i is the charge of the ionic species, u_i is the mobility of the charged species, F is Faraday's constant, and ϕ_l is the electrolyte potential.

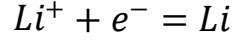
The electroneutrality is governed through the balancing equation:

$$\frac{\partial c_i}{\partial t} + \nabla \cdot N_i = 0$$

We have two species one for Li^+ cation and other for TFSI anion then:

$$\sum_i z_i c_i = 0$$

The boundary conditions for the cathode and anode and cathode are governed by Butler-Volmer equation for Li metal deposition, assuming the following reaction.



The local exchange current density is computed using Tafel-equation:

$$i_{ct} = i_0 \left(\exp\left(\frac{0.75F\eta}{RT}\right) - \frac{c_{Li^+}}{c_{Li^+,ref}} \exp\left(-\frac{0.5F\eta}{RT}\right) \right)$$

Where η is the interface overpotential, defined by:

$$\eta = \phi_{s,0} - \phi_l - \Delta\phi_{eq}$$

Where $\phi_{s,0}$, is the electrode applied potential, and ϕ_{eq} is the equilibrium potential.

3- Input parameters used in the model:

Initial concentration	1 M
System temperature	298 K
Exchange current density	27 mA/cm ²
Relative equilibrium potential	0V
Anode potential	0.02V
Cathode potential	-0.02V
Anode symmetry factor	0.75
Cathode symmetry factor	0.25
Li diffusivity	10 ⁻⁶ m ² /s
TFSI diffusivity	10 ⁻⁶ m ² /s