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Ph.D. DISSERTATION DEFENSE

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Degree:	Doctor of Philosophy
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Date:	Tuesday, April 30th, 2024
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Title:	Lithium-dendrite suppressing 3D Framework by Electrowriting for Battery Applications
Chairperson:	Dr. Jae Chul Kim, Department of Chemical Engineering and Materials Science, Charles V. Schaefer, Jr. School of Engineering & Science
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ABSTRACT

Lithium metal has gained extensive attention to overcome the theoretical limitations of intercalation-type anodes. Unlike commercial graphite anode, metallic lithium is well-known for its high energy density and low reduction potential. The anode-free configuration with zero-excess lithium is progressively suggested as a particular type of lithium metal battery for safe and facile manufacturing purposes. However, due to dendritic lithium formation, realizing poor cycling stability and Coulombic Efficiency remains a continuing challenge.

In our work, a novel solution electrowriting method was developed to accommodate reversible lithium (Li) plating/stripping processes and address the issue of uncontrollable growth of Li dendrites during the energy storage process for anode-free Li batteries. This method merges the broad applicability of electrospinning with the precision inherent in three-dimensional (3D) printing to create microporous, intricate structures (e.g., a square-grid pattern) from polyvinylidene fluoride (PVDF) fibers on copper substrates. The periodic polymer grid successfully confines Li nucleation for the anode by tuning the electrical conductivity and lithiophilicity of PVDF fibers. It steers the lithium growth on a microscale, delaying the Li dendrite formation in the early stage. The key to our extended cyclability of anode lies in the formation of dense and smooth SEIs that are well integrated with the polymer framework during Li plating and stripping. The mechanically flexible framework can effectively dissipate strain energy associated with volume change and reinforce SEIs, further suppressing Li dendrite growth. As a result, we demonstrate the dendrite-free operation of reversible Li plating and stripping (1 mAh cm-1) for more than 150 cycles (1 mA cm-1 rate) in a half-cell and 100 cycles (C/5 rate) in an anode-free full-cell, substantially outperforming the conventional planar anode systems for energy storage.

Applying the electrowriting technique to battery applications, we have established a robust and adaptable processing technique to tailor the anode geometry of advanced batteries that utilize metallic lithium in the anode, such as Li metal batteries, anode-free batteries, solid-state batteries, etc. This not only paves the way for the development of dendrite-free energy storage but also orchestrates electro-chemo-mechanical properties at both a material and cell level. This provides a tangible blueprint for the future of energy storage, inspiring and exciting the scientific community about the potential of our research.