

Controlling lithium metal deposits using different metal substrates

March 10 2016, by Heather Zeiger



Lithium ingots with a thin layer of black oxide tarnish. Image: Wikimedia Commons

(Tech Xplore)—Cell phones, laptops, and other electronic devices that have rechargeable batteries use lithium ion batteries. Lithium batteries have a high energy density. Fewer cells are needed to power a device, which allows for a lighter device that is more portable. However, lithium batteries are not as powerful or as long-lasting as they could be. For one, lithium battery cells tend to form dendrites, lithium metal deposits that form on the lithium electrode. These dendrites spread throughout the lithium-based electrolyte and if it reaches the other electrode, can short circuit the battery.

Kai Yan, Zhenda Lu, Hyun-Wook Lee, Feng Xiong, Po-Chun Hsu, Yuzhang Li, Jie Zhao, Steven Chu, and Yi Cui of Stanford University

have found a way to control dendrite formation allowing them to create a [gold](#) nanoparticle encapsulated in an amorphous carbon shell in which lithium deposits can grow while separated from the electrolyte, thus preserving the life time and capacity of the lithium cell. Their work appears in *Nature Energy*.

Lithium metal has the highest specific anode capacity compared to other metals, but electrochemical cycling of lithium metal poses several problems, including dendrite growth. Controlling dendrite growth has proved difficult for researchers interested in optimizing lithium battery life and capacity.

In their first experiment, Yan, et al. set up a three-electrode cell to investigate the potential at which lithium deposits occur, the nucleation barrier overpotential. Copper is often used in [lithium battery](#) studies because it has a relatively high nucleation overpotential, meaning it requires a large input of energy before dendrite formation occurs.

Using [copper](#) as a baseline substrate, they gravimetrically deposited lithium onto various metal substrates to test the overpotential. These substrates included Au, Ag, Zn, Mg, Al, Pt, Si, Sn, C, Cu, and Ni. They found that while copper had a nucleation barrier at around 40 mV, gold plated copper did not have a nucleation barrier. This is surprising given that gold's crystal morphology is very similar to copper.

The difference in overpotential has to do with solubility. Lithium-gold alloys form in solution and serve to lower the nucleation barrier. Yan, et al. tested this concept with other metals whose phase diagrams indicated that they would be soluble in lithium and found that Au, Ag, Zn, and Mg exhibited zero overpotential when fully "lithiated." Aluminum and Pt are slightly soluble in lithium solution and showed a small overpotential for Li nucleation. Copper, Ni, C, Sn, and Si are insoluble in lithium solution. All five had a clear overpotential for Li metal nucleation and exhibited

comparatively higher nucleation overpotentials. This experiment showed that the nucleation overpotential is related to the formation of lithium-metal alloys. Copper and Ni, which have high overpotentials, do not form alloys.

With this information, Yan, et al. then tested whether lithium would preferentially deposit onto one metal over another. They tested various patterned surfaces of gold on copper. In each of the cases, including one with gold nanoparticles deposited on copper, lithium preferentially deposited onto the gold surface and not on the copper surface.

This then lead to the notion of making encapsulated gold nanoparticles to see if lithium would selectively grow within the carbon shell on the surface of gold nanoparticles. Amorphous carbon is conductive, and lithium ions are small enough that they can traverse through the carbon channels. Furthermore, amorphous carbon has a nucleation barrier overpotential of $\sim 14\text{mV}$, which means [lithium metal](#) should preferentially deposit onto the gold over the carbon shell. The next question was whether it will do so even when the gold is within a carbon shell.

Yan, et al. found that lithium deposits formed on the gold nanoparticles even when encapsulated by the amorphous carbon shell. This was confirmed using several studies, including ex situ TEM, melting and sublimation laser studies, as well as comparison to a control test with an empty [amorphous carbon](#) shell. The carbon shell without an enclosed gold nanoparticle resulted in lithium deposits on the outside of the shell. The shell that contained the gold nanoparticle, however, demonstrated lithium deposits forming within the shell.

The encapsulated lithium dendrites reduced direct contact with the electrolyte, thus corralling dendrite growth. Tests showed that the encapsulated gold nanoparticles operated with a 98% Columbic

efficiency in carbonate electrolyte, a corrosive electrolyte, and survived more than 300 cycles.

This research demonstrates a new way to control lithium dendrite formation by taking advantage of differences in the nucleation overpotential for different metals. It also provides a proof-of-concept example of how this can be exploited to contain lithium dendrite growth within a carbon shell. This study provides a good foundation for additional research for optimizing [lithium](#) batteries.

More information: Kai Yan et al. Selective deposition and stable encapsulation of lithium through heterogeneous seeded growth, *Nature Energy* (2016). [DOI: 10.1038/nenergy.2016.10](https://doi.org/10.1038/nenergy.2016.10)

Abstract

Lithium metal is an attractive anode material for rechargeable batteries, owing to its high theoretical specific capacity of 3,860 mAh g⁻¹. Despite extensive research efforts, there are still many fundamental challenges in using lithium metal in lithium-ion batteries. Most notably, critical information such as its nucleation and growth behaviour remains elusive. Here we explore the nucleation pattern of lithium on various metal substrates and unravel a substrate-dependent growth phenomenon that enables selective deposition of lithium metal. With the aid of binary phase diagrams, we find that no nucleation barriers are present for metals exhibiting a definite solubility in lithium, whereas appreciable nucleation barriers exist for metals with negligible solubility. We thereafter design a nanocapsule structure for lithium metal anodes consisting of hollow carbon spheres with nanoparticle seeds inside. During deposition, the lithium metal is found to predominantly grow inside the hollow carbon spheres. Such selective deposition and stable encapsulation of lithium metal eliminate dendrite formation and enable improved cycling, even in corrosive alkyl carbonate electrolytes, with 98% coulombic efficiency for more than 300 cycles.

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Citation: Controlling lithium metal deposits using different metal substrates (2016, March 10)
retrieved 24 April 2024 from
<https://techxplore.com/news/2016-03-lithium-metal-deposits-substrates.html>

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