RESEARCH HIGHLIGHT

National Science Review
0: 1–2, 2016
doi: 10.1093/nsr/nww084
Advance access publication 0 2016

MATERIALS SCIENCE

Special Topic: Energy Storage Materials

Taming lithium metal through seeded growth

Yan Yao

Recently, there has been worldwide enthusiasm in applying lithium metal as an anode material to batteries. Lithium metal battery could triple the specific energy of today's typical electric vehicle (EV) batteries from 170–200 Wh/kg to 500 Wh/kg. In fact, lithium metal has been intensively studied as an anode material since the 1970s due to its high theoretical capacity (3860 mAh/g) and lowest redox potential (–3.040 V vs. standard hydrogen electrode) [1]. However, uncontrollable dendritic Li growth and limited coulombic efficiency during repeated Li deposition/stripping have prevented practical applications. Incidents of fire followed by manufacture recalls diminished the industrial efforts in the early 1990s [2]. With the urgent need for next-generation high-energy-density battery technology, such as Li–S and Li–air batteries, lithium metal research has been revitalized and attracted regenerated interest in recent years [3]. Key issues relating to Li metal are the following: deposition of Li metal tends to occur anywhere with unpredictable shape; deposition of Li metal tends to be highly dendritic; the irreversible reaction between Li metal and electrolyte leads to low coulombic efficiency and severe capacity decay [4]. Recent progress in improving the morphology and coulombic efficiency of Li metal with novel electrolyte additive, solid electrolyte, nanoscale interface design, etc. has been reviewed by Xu *et al.* [3]. However, there is no effective approach to regulate Li deposition with control over both position and morphology.

In a recent report, Yan *et al.* showed an elegant approach to selective deposition and encapsulation of Li metal in hollow carbon shells through seeded growth, which eliminated dendrite formation and enabled improved cycling over 300 cycles [5]. After investigating 11 metal substrates and their Li-metal binary phase diagrams, the authors discovered a substrate-dependent Li-metal nucleation phenomenon: no nucleation barrier is present for metals exhibiting a finite solubility in lithium, whereas appreciable nucleation barrier exists for materials with negligible solubility. Such a discovery immediately led to a demonstration of spatially controlled Li deposition on a patterned substrate. Au strips were patterned on copper substrates as seeds for Li nucleation where the Li_xAu alloy served as a buffer layer with no nucleation barrier. A Li-metal anode made of a nanocapsule assembly consisting of hollow carbon spheres with embedded Au nanoparticle seeds was designed and tested. Amorphous carbon shells exhibit a slightly higher nucleation barrier than that of Au nanoparticles, therefore promoting Li nucleation and growth inside the carbon spheres (Fig. 1a). This hypothesis was visualized and confirmed by *in situ* TEM characterization (Fig. 1b and c). Stable cycling over 300 cycles with 98% coulombic

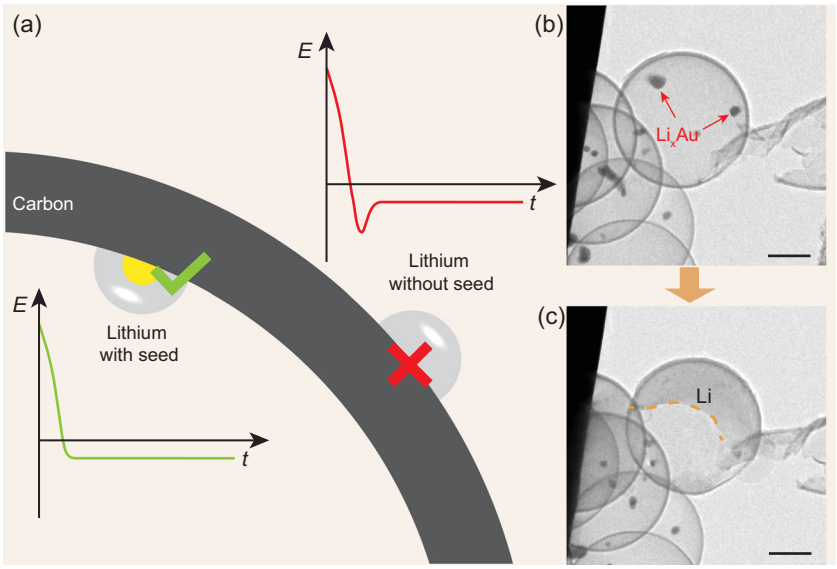

Figure 1. (a) Schematic of manipulating lithium metal deposition. Li-soluble material (Au, green curve) allows the nucleation of lithium metal without overpotential. In contrast, a lithium nucleation barrier is obvious on Li-insoluble substrate (carbon, red curve). The difference in nucleation barrier allows the selective deposition of lithium metal. (b, c) *In situ* TEM images illustrate the process of lithium metal growing inside the carbon shell. (b) The Au seeds were lithiated first before actual plating of lithium metal. (c) As the deposition process continues, lithium is exclusively formed inside the shell. Scale bar: 200 nm.

© The Author(s) 2016. Published by Oxford University Press on behalf of China Science Publishing & Media Ltd. All rights reserved. For permissions, please e-mail: journals.permissions@oup.com

efficiency was demonstrated in alkyl carbonate electrolytes. To be practical, Li-metal anodes with coulombic efficiency of at least 99.9% are necessary because irreversible reaction between Li and electrolyte will lead to the accumulation of solid electrolyte interphase and capacity decay.

This work presents an unexploited approach to precisely control Li-metal nucleation during electrochemical plating, which is not only timely, but also thought-provoking. Although there are still many obstacles to overcome, the advancements in nanomaterial and nanostructure synthesis, *operando* characterization techniques, as well as modeling tools, which did not exist in early 1990s, make us optimistic that the 'holy-grail' lithium-metal challenges could be successfully tackled in the near future.

Yan Yao
Department of Electrical and Computer Engineering, University of Houston, USA.
E-mail: yyao4@Central.UH.EDU

REFERENCES

1. Whittingham MS. *Science* 1976; **192**: 1126–7.
2. Xu K. *Chem Rev* 2004; **104**: 4303–418.
3. Xu W, Wang J and Ding F *et al. Energy Environ Sci* 2014; **7**: 513–37.
4. Huggins R. *Advanced Batteries*. Springer, 2009.
5. Yan K, Lu Z and Lee HW *et al. Nature Energy* 2016; **1**: 16010.