Mesoscale Interplay in Lithium-Ion Batteries and Beyond

PARTHA P. MUKHERJEE,^{1,4} LEELA M. ARAVA,² and AMIT PANDEY³

1.—Department of Mechanical Engineering, Texas A&M University, College Station, TX, USA.
2.—Department of Mechanical Engineering, Wayne State University, Detroit, MI, USA. 3.—LG
Fuel Cell Systems Inc, North Canton, OH, USA. 4.—e-mail: pmukherjee@tamu.edu

Energy storage is a critical component in future vehicle electrification and grid storage. Electric vehicles (EVs) urgently demand revolutionary innovation in energy storage systems with improved performance (high energy and power density), safety, and life. Lithium-ion (Li-ion) batteries are the front runner in the race for vehicle electrification. For example, sulfur (S) and oxygen (O₂) are promising active materials, which can dramatically increase the theoretical specific capacity in Li–S and Li–O₂ based energy storage systems.

Energy storage systems consist of multiphase functional materials and experience, during cycling (charge/discharge), a multitude of coupled physicochemical processes involving mass and charge transport in electrode and electrolyte phases, phase transition with volume change, electrochemical reaction at the electrode–electrolyte interface, side reactions leading to chemical degradation, and diffusion-induced mechanical stress generation in the porous electrode microstructures. These inherently coupled, physicochemical interactions happen at different length and time scales, which ultimately determine the cell performance, life, and safety. To this end, energy storage research necessitates spanning a wide array of computation approaches and characterization techniques reflecting the fundamental need to probe at different scales. In this special topic, interactions at the mesoscale are highlighted, including mechano-electrochemical coupling, microstructure–property–performance interplay, mesoscale modeling, and characterization.

Horrocks et al. reviewed x-ray spectroscopy and imaging techniques to probe electronic and atomic structures in cathode material exhibiting phase transition during lithium intercalation. Ausderau et al. employed x-ray nanotomography (XNT) and x-ray absorption near-edge structure (XANES)

imaging to study elemental and chemical mapping of high-capacity, Cu₆Sn₅-based intermetallic alloying anode for Li-ion batteries.

Jianlin Li and co-workers reviewed different strategies for low-cost manufacturing of high-performance Li-ion batteries. Mullaivananathan et al. showed that a carbon-coated Co₂SnO₄/SnO₂ composite anode can achieve much higher specific capacity as a result of an ability of the carbon to accommodate the volume change during alloying/de-alloying. Lin et al. synthesized a Li₄Ti₅O₁₂/Ti₃O₅ composite anode using a ball milling method and showed that higher specific capacity than the conventional Li₄Ti₅O₁₂ anode can be achieved, which can be beneficial in reducing the capacity loss. Kalaga et al. investigated the effect of the temperature on V₂O₅ phase transition induced by Li intercalation. Intawin et al. presented a biosynthesis strategy for deriving hierarchical three-dimensional (3D) carbon-based architectures for supercapacitor application. It is interesting that the macroporous carbon derived from sunflower seed shows high specific capacitance with good cycling stability.

Hardin et al. developed a finite element model to investigate the interfacial fracture of nanowire electrodes for Li-ion batteries. It is found that the plastic deformation in the nanowire can reduce the crack-driving force and avert interfacial fracture between nanowire electrode and current collector. Stein et al. presented a reaction–diffusion model for investigating Li intercalation in SiOC nanocomposite electrodes that exhibit phase transition.

In the cathode side, solid sulfur as the active material is expected to replace conventional transition metal oxides because sulfur can deliver a very high theoretical specific capacity as high as 1675 mAh/g and sulfur is cheap and abundant. Nevertheless, several critical challenges, including the shuttle effect and surface passivation, continue to remain as impediments in Li–S batteries. Liu et al. developed a mesoscale interfacial modeling approach to evaluate Ti₂Si nanosheets as a

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potential cathode host material for Li–S batteries. They showed that Ti_2Si nanosheets can act as a catalyst to facilitate the dissociation and reduction of soluble long-chain polysulfides, which is beneficial for mitigating the harmful shuttle effect. Yet, on the other hand, the Ti_2Si nanosheets provide strong affinity to Li_2S , which leads to a fast surface passivation and sudden death. This work highlights an important open question: Is the Li–S cell performance dominated by shuttle effect or surface passivation?

The following articles are published under the topic “Mesoscale Interplay in Lithium-Ion Batteries and Beyond” in the September 2017 issue (vol. 69, no. 9) of *JOM* and can be accessed via the *JOM* page at <http://link.springer.com/journal/11837/69/9/page/1>.

- “X-ray Spectroscopy and Imaging as Multiscale Probes of Intercalation Phenomena in Cathode Materials” by Gregory A. Horrocks, Luis R. De Jesus, Justin L. Andrews, and Sarbajit Banerjee
- “Elemental and Chemical Mapping of High Capacity Intermetallic Li-ion Anodes with Transmission X-ray Microscopy” by Logan J. Ausderau, Hernando J. Gonzalez Malabet, Joseph R. Buckley, Vincent De Andrade, Yijin Liu, and George J. Nelson
- “Toward Low-Cost, High-Energy Density, and High-Power Density Lithium-Ion Batteries” by Jianlin Li, Zhijia Du, Rose E. Ruther, Seong Jin An, Lamuel Abraham David, Kevin Hays, Marissa Wood, Nathan D. Phillip, Yangping Sheng, Chengyu Mao, Sergiy Kalnaus, Claus Daniel, and David L. Wood III
- “Synthetically Controlled Carbon-Coated $\text{Co}_2\text{SnO}_4/\text{SnO}_2$ Composite Anode for Lithium-Ion Batteries” by V. Mullaivananathan, K.R. Saravanan, and N. Kalaiselvi
- “Investigations on $\text{Li}_4\text{Ti}_5\text{O}_{12}/\text{Ti}_3\text{O}_5$ Composite as an Anode Material for Lithium-Ion Batteries” by Yuanhua Lin, Wen Xu, Xiaoyan Zhang, Lijun Wang, and Wanying Liu
- “Phase Transformations During Li-Insertion into V_2O_5 at Elevated Temperatures” by Kaushik Kalaga, Farheen N. Sayed, Marco-Tulio F. Rodrigues, and Pulickel M. Ajayan
- “Bio-Derived Hierarchical 3D Architecture from Seeds for Supercapacitor Application” by Prathana Intawin, Farheen N. Sayed, Kamonpan Pengpat, Jarin Joyner, Chandra Sekhar Tiwary, and Pulickel M. Ajayan
- “Interfacial Fracture of Nanowire Electrodes of Lithium-Ion Batteries” by G.R. Hardin, Y. Zhang, C.D. Fincher, and M. Pharr
- “A Model for Diffusion and Immobilization of Lithium in SiOC Nanocomposite Anodes” by Peter Stein, Dragoljub Vrankovic, Magdalena Graczyk-Zajac, Ralf Riedel, and Bai-Xiang Xu
- “Mesoscale Evaluation of Titanium Silicide Monolayer as a Cathode Host Material in Lithium–Sulfur Batteries” by Zhixiao Liu, Perla B. Balbuena, and Partha P. Mukherjee