

Novel Atomic and Molecular Layer Deposition Processes for Robust Battery Interfaces

Xiangbo Meng^{*a}, Kevin Velasquez Carballo^a, Kang Lu^a, and Aiyong Shao^a

^a Department of Mechanical Engineering, University of Arkansas, Fayetteville, AR 72701, USA

* Corresponding author: xbmeng@uark.edu

Adopting lithium metal (Li) as anodes and nickel (Ni)-rich lithium nickel manganese cobalt oxides ($\text{LiNi}_x\text{Mn}_y\text{Co}_z\text{O}_2$, NMCs, $x \geq 0.6$, $x + y + z = 1$) as cathodes, the resultant Li||NMC lithium metal batteries (LMBs) could be twice higher in energy (up to 500 Wh/kg) but 50% lower in cost (\$100/kWh) than that of LIBs, holding great promise to replace LIBs for the applications of portable electronics, electric vehicles, and aircrafts. Unfortunately, such a compelling technology has been hindered from commercialization due to some serious interfacial issues related to the Li anodes and NMC cathodes. Aimed at addressing these challenges, we recently have developed a series of novel coatings via atomic and molecular layer deposition (ALD and MLD). ALD and MLD share several unique merits but are complementary in their target materials. They have emerged as two new techniques of interface engineering of rechargeable batteries in the past decade.¹⁻⁵ They both could deposit conformal and uniform coatings over complex shapes of different substrates, operate at low process temperature, and accurately control coating thickness. Through adopting different precursors, ALD exclusively deposits inorganic films while MLD specially grows organic or hybrid films. For the issues of Li anodes and NMC cathodes, we particularly designed function-oriented coatings⁶⁻¹⁰ via ALD and MLD. Using our ALD and MLD processes, very encouragingly, both the surface-coated Li anodes and NMC cathodes have exhibited remarkable improvements in their electrochemical performance. Our studies have further shown that the combination of these coatings can synergistically maximize their benefits to achieve higher performance of Li||NMC LMBs, enabling a cell capacity fading 10 times slower than that of bare Li||NMC cells and a capacity retention improvement over 60% after 500 charge/discharge cycles. In this talk, we will introduce these novel coatings and their compelling effects. Particularly, we would like to explain the underlying mechanisms related to their benefits. Thus, our studies have not only opened new areas of surface coatings but also demonstrated their technical feasibility for developing high-performance LMBs.

References:

1. Meng, X.; Yang, X. Q.; Sun, X. L., Emerging applications of atomic layer deposition for lithium-ion battery studies. *Adv. Mater.* **2012**, *24* (27), 3589-3615.
2. Meng, X., Atomic layer deposition of solid-state electrolytes for next-generation lithium-ion batteries and beyond: Opportunities and challenges. *Energy Storage Materials* **2020**, *30*, 296-328.
3. Meng, X., Atomic-scale surface modifications and novel electrode designs for high-performance sodium-ion batteries via atomic layer deposition. *J. Mater. Chem. A* **2017**, *5*, 10127-10149.
4. Meng, X., An overview of molecular layer deposition for organic and organic-inorganic hybrid materials: Mechanisms, growth characteristics, and promising applications. *J. Mater. Chem. A* **2017**, *5* (35), 18326-18378.
5. Meng, X., Atomic and molecular layer deposition in pursuing better batteries. *J. Mater. Res.* **2021**, *36*, 2-25.
6. Wang, X.; Cai, J.; Ren, Y.; Benamara, M.; Zhou, X.; Li, Y.; Chen, Z.; Zhou, H.; Xiao, X.; Liu, Y.; Meng, X., High-performance $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$ cathode by nanoscale lithium sulfide coating via atomic layer deposition. *Journal of Energy Chemistry* **2022**, *69*, 531-540.
7. Ahmed, R. A.; Carballo, K. V.; Koirala, K. P.; Zhao, Q.; Gao, P.; Kim, J.-M.; Anderson, C. S.; Meng, X.; Wang, C.; Zhang, J.-G.; Xu, W., Lithicone-Protected Lithium Metal Anodes for Lithium Metal Batteries with Nickel-Rich Cathode Materials. *Small Structures* **2024**, 2400174.
8. Meng, X.; Lau, K. C.; Zhou, H.; Ghosh, S. K.; Benamara, M.; Zou, M., Molecular Layer Deposition of Crosslinked Polymeric Lithicone for Superior Lithium Metal Anodes. *Energy Material Advances* **2021**, *2021*, 9786201.
9. Wang, X.; Cai, J.; Velasquez Carballo, K.; Watanabe, F.; Meng, X., Tackling issues of lithium metal anodes with a novel polymeric lithicone coating. *Chem. Eng. J.* **2023**, *475*, 146156.
10. Wang, X.; Velasquez Carballo, K.; Shao, A.; Cai, J.; Watanabe, F.; Meng, X., A novel polymeric lithicone coating for superior lithium metal anodes. *Nano Energy* **2024**, *128*, 109840.