**Interface Chemistry and Engineering Approaches for**

**Stable, High-energy Li-metal Batteries**

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Li-metal batteries (LMBs) have been revisited to surpass the theoretical energy density limit of current Li-ions. However, its practical uses in rechargeable batteries have long been plagued for several decades due to Li dendrite growth and low Coulombic eﬃciency (CE), resulting in safety hazards and poor cycling performances. Moreover, Li metal anode suffers from the dynamic evolution of interphase structures due to uncontrolled Li plating/stripping processes, leading to severe Li pulverization and chemical failures of cell components. Therefore, a highly stable Li-electrolyte interface structure with rational cell design is highly required to regulate Li deposition and minimize the Li loss, thereby enabling higher-energy, highly safe, longer-cycling LMBs.

In this talk, the physicochemical features and structural evolution of the Li-electrolyte interface will be discussed. Critical parameters of cell design have been redefined to obtain a high-energy-density in practice, and their effects on cycling performances have been investigated [1]. Through this “top-down” approach to elucidate more realistic challenges in practical LMBs, a prototypical 300 Wh kg−1 Li-metal pouch cell (1.0 Ah) has been successfully demonstrated by combining with much thinner Li-metal, LiNi0.6Mn0.2Co0.2O2 cathode and, lean electrolyte [2]. A key issue inherent to Li-metal pouch cells is excessive cell swelling triggered by parasitic reactions with electrolytes and a highly porous passivation layer buildup. It has been demonstrated that the cell swelling can be suppressed by more than 100% with 1.2 M LiFSI/(TEP:BTFE) electrolytes and uniform external pressure and thereby resulting in more than 200 stable cycles with 86% capacity retention. Based on the findings, multi-scale engineering approaches on the battery materials and cell interior/exterior designs will be presented.

**Reference**

[1] S. Chen†, C. Niu†, H. Lee†, Q. Li, L. Yu, W. Xu, J.-G. Zhang, E. J. Dufek, M. S. Whittingham, S. Meng, J. Xiao, J. Liu, *Joule*, 3 (4) 1094-1105 (2019).

[2] C. Niu, H. Lee, S. Chen, Q. Li, J. Du, W. Xu, J.-G. Zhang, M. S. Whittingham, J. Xiao, J. Liu, *Nat. Energy* 4, 551–559 (2019).