

## **Physicochemical Interaction in Electrode Processing for Energy Storage**

Zhixiao Liu, Malcolm Stein IV, Partha P. Mukherjee\*

Department of Mechanical Engineering, Texas A&M University, College Station, TX

\*pmukherjee@tamu.edu

### **ABSTRACT**

Energy storage, such as lithium-ion batteries, is a key enabler for grid energy storage and vehicle electrification. The performance of lithium-ion batteries is dependent on the electrode microstructure. Processing attributes play an important role in the electrode microstructure formation and resulting influence on the properties and performance. In this work, an integrated study combining experiments and mesoscale modeling is presented in order to elucidate the impact of evaporation in electrode processing. Our experimental study demonstrates that a low evaporation rate can produce an electrode with active material particles more uniformly coated with conductive additive/binder composite. The electrochemical performance tests demonstrate that the electrode processed via a low evaporation rate has lower resistance than that processed by higher evaporation rate. In addition, the former one is beneficial for mitigating capacity fading during the cycling. To fundamentally understand the experimentally observed microstructure evolution affected by solvent evaporation, a morphology-detailed mesoscale model is developed. The simulations predict that a high evaporation rate cuts off the connecting pathways between active materials and conductive additives due to the fast growth of gas bubbles in the electrode slurry. However, an appropriate evaporation rate is beneficial for conductive additive coating on the active material. The mesoscale model also predicts that small-sized active material nanoparticles are beneficial for conductive additive coating due to the high specific surface area. In addition, a stepwise mixing sequence during the slurry evaporation facilitates electrode microstructure formation with improved properties performance.