Abstract of “Collective Behavior at the Interface of Lithium-Ion Batteries under Cyclic Lithiation” by Hsiao-Mei Wu, Ph.D., Brown University, May 2014.

This thesis presents experimental measurements and modeling of multi-scale collective behaviors characteristics of hierarchical interfaces in lithium-ion batteries (LIBs) during cycling. Two interfacial mechanisms are introduced: One is in-plane sliding between lithiated electrodes (a-$SiLi_x$) and current collectors (Cu). The other is normal contact between the internal interfaces of pouch battery cells.

To estimate the interfacial properties at a-$SiLi_x$//Cu interfaces, a new apparatus, named “Self-Adjusting Liquid Linnik Interferometer (SALLI)”, has been invented to perform *in situ* whole field deformation measurements with μm resolution in lateral direction and nm resolution in out-of-plane direction. Our result clearly demonstrates Li segregation at the interface initially which leads to 200 μm shrinkage of the Si film in the first cycle due to relaxation of residual tensile stress. A mechanical model system, plate bending distribution sensor (PBDS), which incorporates substrate bending and interfacial sliding in its calibration has been developed. By bridging the deformation estimated from PBDS and that measured from the SALLI experiment, the interfacial properties are extracted quantitatively. The critical energy release rate is estimated as 0.075 J/m^2 and 0.34 J/m^2 for the receding and growing shear crack fronts respectively. A remarkable discovery is that the interfacial shear strength of the actively segregating lithium at the interface is measured only 1.15 kPa. It is due electro-chemically active lithium-ion segregation process that allows slip processes of hopping through a series of meta-stable atomic configurations.

Finally, two sets of *in situ* experiments have been performed and a mechanical model has been developed to explain the internal contact mechanism and its relationship with interface bubble-gas evolution. Through these techniques, the degradation mechanism of the pouch cells are explained. It shows that applying 4–5 psi prestressed pressure to the cell can better control the bubble-gas formation and increase the electrode contact area. Therefore, the battery life is efficiently elongated.

It is hoped that the thesis work can contribute to optimal design of battery cells and maximize cell capacity and life of LIBs.