

Improving Thermomechanical Reliability of Li-Ion Batteries to Withstand Freeze-Thaw Process (Thermal Cycling). N. Rolston¹, S. Smas¹, and J. Rice¹, ¹Arizona State University, 781 S Terrace Rd, Room 632, Tempe, AZ, 85287. (Contact: nicholas.rolston@asu.edu)

Introduction: Mechanical properties are often overlooked but excellent predictors for device resilience to environmental stressors that accelerate the evolution of internal defects and cause fracture and device failure. Photovoltaics, power electronics, and batteries often incorporate layered structures, for which the understanding of mechanical properties is crucial to designing durable and cyclable devices. These devices can accumulate significant film stresses in operation. In the lunar environment, the extreme variation in temperature from the lunar day to night is another source of significant film stress. The combination of stresses resulting from mechanical and environmental effects induce fracture processes either at the interfaces of adjacent device layers or through a single layer, the effect of which decreases performance, acts as a pathway for accelerated diffusion of environmental species, and ultimately causes device failure.

Environmental stressors (e.g.- thermal cycling, high vacuum, radiation, and lunar regolith) are used to probe the operational robustness of power devices and systems. The objective of this work is to study the viability of hibernation on power systems for both polar and equatorial lunar missions using thermomechanical properties to determine design criteria for batteries and power devices capable of recovery and continued operation at lunar dawn. The answer is pursued by studying deformation and reaction/degradation in lunar environments that together target a mechanistic understanding of how these factors affect functionality of power system components.

Technical approach: Adhesive or cohesive failure that leads to device degradation and lack of operation may result from either residual or applied mechanical stresses introduced during device processing, handling, packaging, and operation. Mechanical stresses provide the driving force for damage and can vary markedly with device application. The mechanical driving force for failure processes like film cracking or interface debonding can be quantified in terms of a strain energy release rate, G (J/m^2), which is a function of the aforementioned mechanical stresses and the film thickness and modulus. Since the mechanical driving force for damage scales as the square of the film stress, σ , an understanding of thermomechanical properties is critical for these devices to enable durability. Fracture occurs when G meets or exceeds the critical value of the strain energy release rate for a material, G_c where G_c is also known as fracture energy since it involves the energy needed to cause bond

breakage—related to bond type and density—and by other energy dissipation processes such as molecular deformation and flow. Measuring G_c does not require any information about the mechanical properties or stresses in the thin film layers, which are often unknown. G_c is a key metric of thermomechanical reliability, yet it is not a well-understood quantity.

Objective 1: *Characterize chemo- and thermomechanical properties of Li-ion batteries and power system components across cell manufacturers, formats, chemistry, and state of charge.*

We have observed a sharp, knee-like degradation feature as a drop in 18650 Li-ion battery capacity with cycle number that precedes device failure (Figure 3), the source of which has not yet been identified. The degradation is accelerated under fast charging, for which strain rates increase and result in larger stresses. The large values of film stress during cycling and resulting driving forces for damage could be a mechanism for the characteristic degradation.

Objective 2: *Design battery architecture and power system to establish safe protocols for cell hibernation cycles and state of charge guidelines with a lightweight form factor.*

The findings in objective 1 further informs the design criteria that will be implemented to engineer more reliable and thermomechanically robust batteries and power electronics. Based on identifying the failure point of weak graphite adhesion, preliminary work has been performed with a strategy to improve interfacial bonding using an organothiol through a self-assembled monolayer approach. The use of ethanethiol and benzimidazole were highly effective in reducing the defective copper oxide and functionalizing the copper surface for improved adhesion. G_c of the graphite/Cu interface increased to $> 1 \text{ J}/\text{m}^2$ for both treatments, and nearly $5 \text{ J}/\text{m}^2$ was achieved in the case of ethanethiol. Surface chemistry from X-ray photoelectron spectroscopy also showed a complete elimination in the oxide after the organothiol monolayer deposition with improved cyclability.

Additional work will focus on studying stability of additional battery components (such as the seal crimp and pressure vent) and investigating chemical degradation mechanisms during the freeze-thaw process.

Acknowledgements: Funding for this project was enabled by ASU NewSpace, which heads the integration of academic and commercial space enterprises using ASU's core strengths in space science, engineering, and education.