FY24 Aging & Lifetimes Final Report Template – length no more than 4 pages

Proposal Name and Technical Area: Rapid Predictions of Part Lifetimes in Corrosive Environments, Corrosion
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Work Category: Aging of the legacy stockpile, aging diagnostics, predictive aging models and capabilities,
Primary material/component aging risk addressed, benefitting components and systems: Benefits future systems (SLCM-N), legacy systems using COTS electronics, and surveillance
Single year, multi-year, or continuation: FY23-FY25
Budget for FY24: 1290k

Technical issue addressed: Corrosion challenges persist throughout SNL's mission areas. The primary difficulty lies in the fact that corrosion typically manifests as isolated, rare events, making preemptive identification exceedingly difficult. Our current strategy for addressing corrosion issues, such as anomalies and SFIs, is similarly isolated and reactive. This method is costly, time-consuming, heavily dependent on a limited number of experts, and offers minimal understanding of the overall damage distribution within the stockpile. This technical challenge is not unique to corrosion but is also prevalent in other material aging phenomena, such as tin-whisker growth in lead-free solder and fatigue failure of springs.

Technical Approach and Results: This project investigates a new method for predicting rare materials degradation phenomena by developing a deep-learning model, complemented by SME expertise, to quickly predict damage distribution across parts under various environmental conditions. The degradation of galvanically coupled Al/Au wire bonds in COTS electronics due to corrosion was chosen as a representative case due to its broad relevance to SNL's mission. To implement this predictive approach, the project consolidated multiple existing A&L corrosion projects under a single umbrella, divided into three main thrusts: 1) examining corrosion mechanisms and rates at the nanoscale, 2) evaluating materials degradation rates due to corrosion at the continuum scale, and 3) accelerating corrosion predictions using data-driven methods, including machine learning. From the outset, the project aimed to maintain relevance to stakeholders in future systems, surveillance, and COTS electronics. Quarterly meetings provided technical updates and gather feedback from stakeholders.

FY24 Impacts Accomplishments:
2 Publications, 1 manuscript submitted*: de Oca Zapiain, David Montes, et al. "Accelerating fem-based corrosion predictions using machine learning." Journal of The Electrochemical Society (2024)*
Venkatraman, Aditya, et al. "An active learning framework for the rapid assessment of galvanic corrosion." npj Materials Degradation (2024)
Venkatraman, Aditya, et al. "Accelerating charge estimation in molecular dynamics simulations using physics-informed neural networks: corrosion applications" ***Submitted to*** *npj Computational Materials (2024)*
7 Presentations: *2 at GRC on Corrosion, TMS 2024, MRS Spring Meeting, March APS Meeting, AMMP Spring Conference, ECS Fall Conference*
Active Learning Framework: Developed an active learning framework to optimize additional simulations and experiments, significantly enhancing model performance and predictive accuracy. This framework is widely applicable to various aging problems at Sandia, providing a versatile tool for future research.

Section 1: *FE and atomistic modelling advances:* We created continuum-scale models, based in Sierra solid-mechanics, that account for corrosion, species transport, moving interfaces, and resistance calculations. These complex multi-physics models were run under similar atmospheric corrosive conditions as the wire

bond experiments described in **Section 2**. Qualitative comparisons showed agreement between the model and experiment, particularly in the steep rise in resistance observed during the corrosion process, see Figure 1. Additional physics were incorporated into the FEM (finite element model), with corrosion current density scaled by the local stress field, marking a first at Sandia. These models created a new FEM capability by coupling stress, corrosion, species transport, moving interfaces, and resistance calculations. A key outcome of this effort was demonstrating that resistance change is not sensitive to corrosion damage – 95% of the interface was destroyed by corrosion before a substantial change in resistance occurred. Validation of the model was also performed using experimental data from literature.

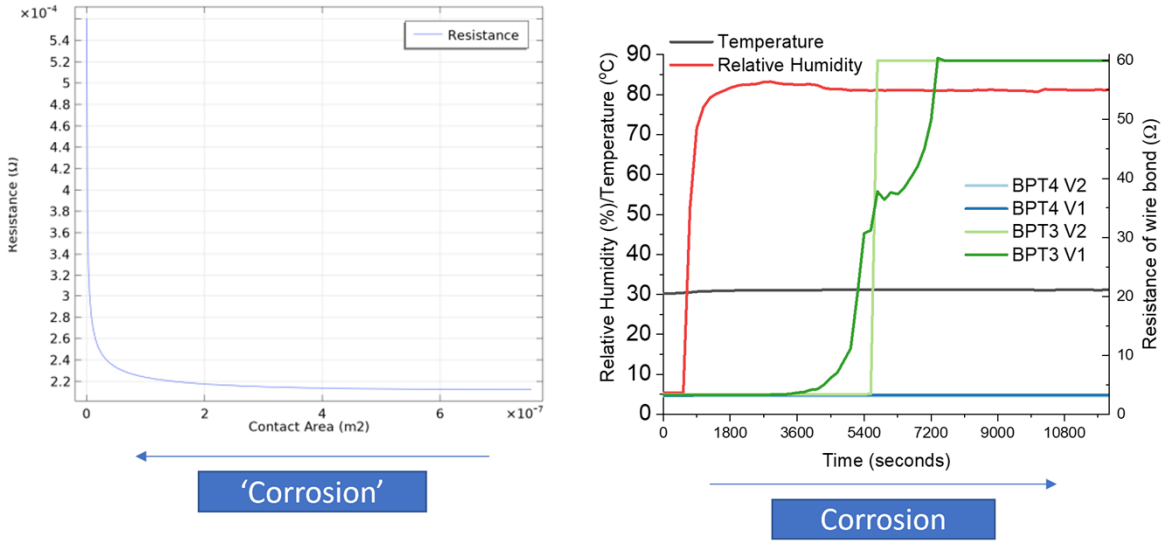

Figure 1. ***(Top-left) Resistance vs. contact area (and time) for FEM corrosion models. It is noted that as contact area decreases, corrosion is occurring noting an increase in resistance. (Top-right) Resistance and experimental conditions vs. time for experimental measurements.***

During FY24, we also developed a simulation method to quantify the energetics and reactivity associated with atomic-scale corrosion events. This research was focused on atomistic length scales, particularly examining the boundary between the specimen and electrolyte. Under normal operating conditions, an oxide layer rapidly forms on the anode, preventing further degradation. This oxide layer undergoes quasi-static depassivation, exchanging ions with the electrolyte. However, under severe conditions, the depassivation rates increase significantly, allowing the electrolyte to penetrate into the base metal, leading to mass loss. Understanding the kinetics at the interface between the oxide layer and electrolyte is crucial for assessing depassivation rates. To study these chemical reactions, which involve significant charge transfer, we used Molecular Dynamics (MD) with reactive force fields (ReaxFF). This allowed us to perform "steered" molecular dynamics (SMD) to bias and drive corrosion events to occur at a metal/electrolyte interface. These events consist of metal ions desorbing from the surface and then traversing into the electrolyte. During these simulations, we traced multiple properties of the given ions: the charge evolution, the force, and the energy necessary to liberate the given ion. Data from the charge evolution simulations were used to validate and demonstrate the capabilities of the charge equilibration ML surrogate model (described in **Section 3**), while the force and energy data provide an avenue to calculate activation energies and rates of corrosion events. Given the near-infinite structural variability of an atomic-scale interface with an electrolyte, we developed a sampling method to calculate these values as an average over multiple trajectories. This simulation approach required wrapping the LAMMPS MD code in Python to perform hundreds of replicate simulations to determine one sample trace.

Section 2: *Experimental validation – resistance change, mass change, and material characterization:*
Prior work by Rob Sorensen suggested that changes in electrical resistance of chips during corrosion provided useful insights into the rate of corrosion. This year, we expanded on this work by exposing "ACT" chips, fabricated ≈20 years ago, to various relative humidity (RH) and temperature conditions. NaCl salt was printed onto the chip surfaces with low (3 μg/cm²) loading densities. Exposures were conducted in RH and temperature-controlled chambers set to 25°C and 25%, 40%, 60%, and 80% RH. Resistance measurements were taken periodically every 200 hours. After ~3000 hours (~3 months), failures were observed at 25% and 40% RH. Increasing the temperature to 50°C yielded no additional failures after 1000 hours. Elevating the 80% RH chip to 85°C for another 1000 hours resulted in six wirebond failures. These failures will be analyzed next year to determine the mechanisms, hypothesized to be related to corrosion and thermal expansion mismatch.

A key insight from the resistance change measurements was that damage below the deliquescence point of NaCl (<76% RH) occurs too slowly to be observed by any technique except, perhaps, QCM (quartz crystal microbalance), though it was unclear if mass changes measured using QCM were caused by corrosion or the formation of an oxide film. Consequently, we shifted our focus to studying conditions above the deliquescence point. This shift had a significant impact, as it essentially rendered QCM useless for experiments above 76% RH due to signal interference from water droplet formation on the sensor surfaces. To address this, we used a high-precision ProUmid microbalance for measurements above the deliquescence point. Experiments on aluminum coupons and microwire chips with varying salt deposition levels were conducted under controlled conditions (50°C/80% RH and 30°C/76% RH). Optical microscopy revealed more corrosion in samples with higher NaCl loading, particularly along the gold-aluminum edges. However, temperature and humidity effects on corrosion rates were inconclusive due to non-uniform salt deposition.

Due to the limited supply of old "ACT" chips, new chips were designed and fabricated. We developed an in-situ testing capability to measure resistance while taking time-lapse images and tracking RH and temperature. This approach addresses the limitations of periodic resistance measurements, which often showed minor fluctuations that were likely measurement artifacts. The in-situ method allows continuous tracking of resistance and electrolyte dynamics. Initial experiments in aggressive environments (80% RH, 40 μg/cm², and 30 or 46°C) showed rapid failures (1-2 hrs.). We are now testing lower salt loading (5 μg/cm²) chips to observe a more gradual failure response. However, resistance alone is relatively insensitive to changes in contact area between the wire and the bondpad. This can be seen in the example resistance *versus* time dataset in Figure 1. Therefore, we are also testing electrochemical impedance spectroscopy (EIS) to detect the earlier onset of corrosion. Our first EIS test was unsuccessful due to facility issues, but we continue to refine the method.

Transmission Electron Microscopy (TEM) was used to investigate the formation of intermetallics at the Au-Al interfaces in wire-bonds. Both new chips and ≈20-year-old ACT chips were characterized. TEM analysis of freshly bonded Al-Au samples showed a consistent Al oxide layer thickness of ~5-15 nm, with no rupturing in the boundary layer. Energy Dispersive X-ray Spectroscopy (EDS) revealed local concentrations of Al on the Au side, indicating the formation of Al-Au intermetallic particles. For the ACT samples, TEM revealed significant attack on the Al layer. Secondary Electron (SE) Scanning Electron Microscopy (SEM) images showed dissolution of the Al layer, and EDS confirmed complete conversion to Al oxide. These long-term exposure results highlight the progressive nature of corrosion and oxidation at the Au-Al interface, providing crucial insights into degradation mechanisms over extended periods.

Section 3: Multi-Modal Data Fusion and Active Learning: We developed an efficient surrogate model using machine learning (ML) to accelerate corrosion assessments. The model performed searches over the multi-dimensional input space to identify corrosion hotspots, achieving speeds 5 orders of magnitude faster than traditional finite element simulations. This allowed us to quickly pinpoint corrosion areas, facilitating targeted experimental investigations. These results were published in the *Journal of Electrochemical Society* (IF: 3.1). An overview of this workflow is provided in Figure 2.

Next, we synthesized heterogeneous data sources, including RDE experiments and simulations, to establish active learning protocols. Using model fusion, we drove active learning cycles to gather additional experimental data efficiently. Low-fidelity simulation models were used as a risk-qualification measure before

pursuing higher-fidelity alternatives. The expected information gain acquisition function provided rapid improvements in experimental assessments, exploring variables like temperature, salt concentration, and specimen geometry. This demonstrated how probabilistic ML could handle multi-modality and deliver agile predictions of materials degradation. This work was published in *npj Materials Degradation* (IF: 5.1). This workflow is illustrated in Figure 3.

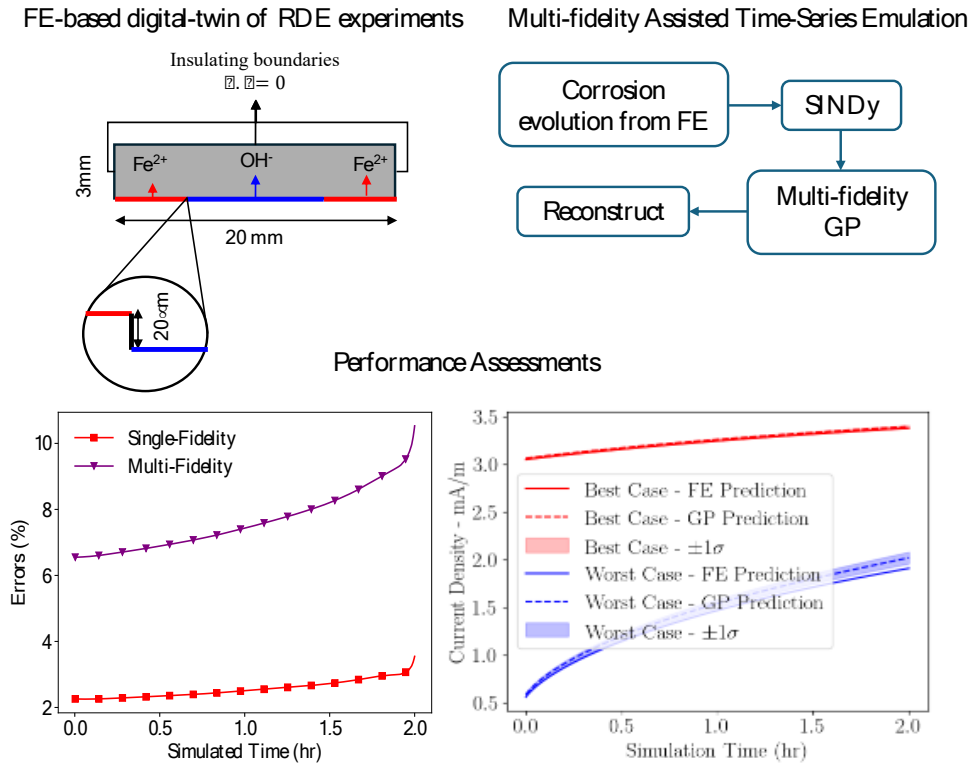

Figure 2. ***(Top-left) Digital-twin of RDE experiments, leveraging a hierarchy of physics-based corrosion models, (Top-Right) ML protocol for accelerating multi-fidelity, time-series current predictions from FE models, (Bottom-left) Error progression across simulated time, showing significant improvements with multi-fidelity framework in comparison to single-fidelity case (i.e. only using high-fidelity data), (bottom-right) Best and worst-case scenarios are qualitatively very similar, illustrating robustness of the workflow***

To further accelerate transient corrosion predictions, we used multi-fidelity simulation data. Five fidelities were considered, with the highest fidelity being 50 times more computationally expensive. We represented the time-evolution of corrosion activity using Sparse Identification of Nonlinear Dynamics (SINDy) and built multi-fidelity Gaussian Process (GP) linkages between basis coefficients at different fidelities. This approach drastically reduced discrepancies between high and low-fidelity models, expediting high-fidelity predictions by over six orders of magnitude. We are preparing a manuscript based on this work for submission to the *CMAME* journal (IF: 7.3).

The MD simulations described in Section 1 incur high computational costs due to charge equilibration, requiring expensive all-to-all matrix inversions. To address this, we developed a Physics-Informed ML approach to expedite charge estimation in ReaxFF-MD simulations. We created quick surrogates to investigate chemical kinetics at the oxide-electrolyte interface, representing atomic environments using spherical harmonics and compressing them with Principal Component Analysis (PCA). Bayesian Long Short-Term Memory (LSTM) networks were used to construct probabilistic, time-dependent surrogates of partial charge. Our ML objective function included physical constraints like electroneutrality and electronegativity equivalence. The probabilistic LSTM generated candidates for charge predictions, screened by applying hard

thresholds on physical constraints, resulting in physics-aware predictions. We demonstrated remarkable accuracy in charge predictions, even for extrapolation. We are expanding this formulation to consider grain boundary effects and coarse-graining/upscaling kinetics to mesoscopic scales using the Mori-Zwanzig formulation. This work is undergoing peer review at *npj Computational Materials* (IF: 9.7).

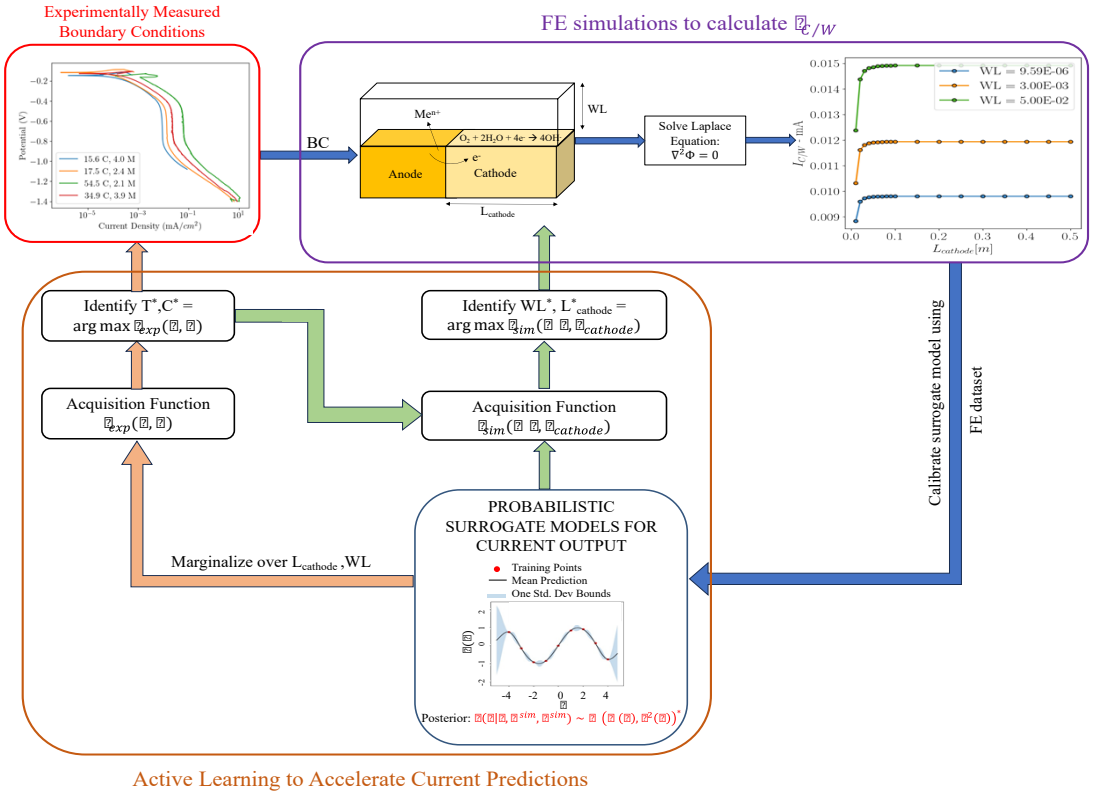

Figure 3. ***Illustration of the model fusion protocol utilized in this study. The blue arrows indicate the exploration step where additional data is acquired with FE simulations. The green and orange arrows represent the exploitation step where the acquisition function is used to identify optimal input configurations for further exploration.***

WPA Deliverables/Date Completed	
1. Establish efficient ML surrogate model for FE-based model that is orders of magnitude faster (published in the *Journal of the Electrochemical Society*)	12-23
2. Use active learning to optimize additional simulations and experiments for model performance (published in *npj Materials Degradation)*	3-24
3. Accelerate transient corrosion predictions using multi-fidelity Gaussian Process (GP) linkages, reducing discrepancies between high and low-fidelity models. (Manuscript in preparation for *CMAME*)	6-24
4. Develop simulation method for atomic-scale corrosion energetics and reactivity using ReaxFF and SMD. (manuscript submitted to *npj Materials Degradation*)	8-24

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