Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

Process Image Analysis using Big Data, Machine Learning, and Computer Vision

This project had 2 objectives: The first was the development of machine learning algorithms to identify the presence of corrosion from a very large set of images generated by laser confocal microscope scanning of 3013 canister used to store Pu oxides. The second component of the LDRD consisted of the development of machine learning algorithms that obtain molecular mechanics force-fields from ab-initio Density Functional Theory (DFT) calculations for corrosive attack by chlorides on 304L or 316L stainless steel.

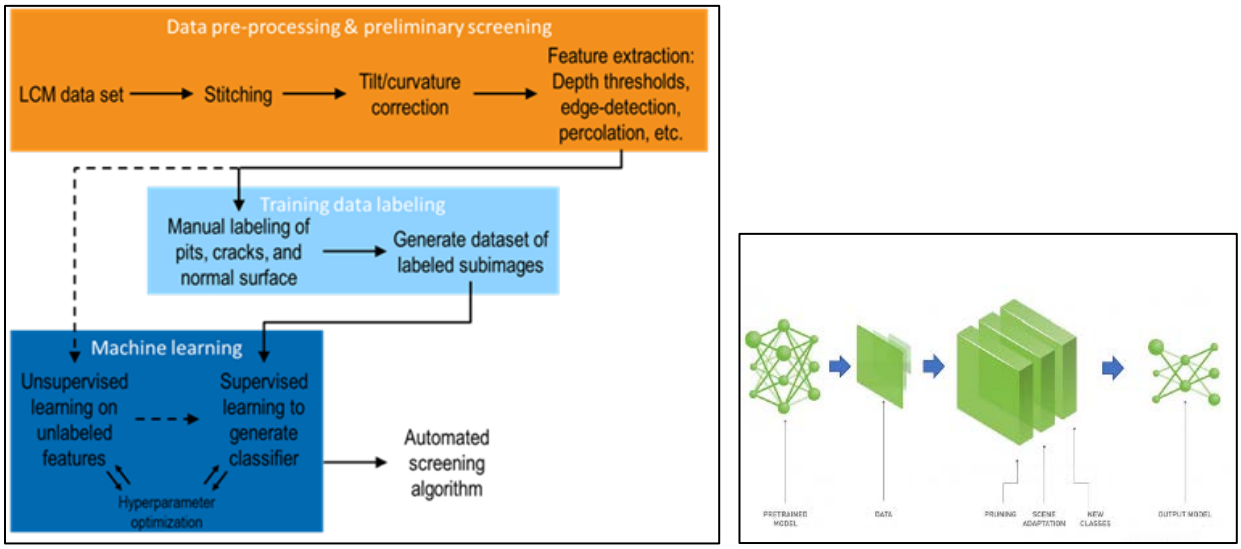

Figure 1. The left figure shows the flowchart for data processing and application of machine learning for identification of corrosion and cracking in image data. The figure on the right shows the flowchart for application of machine learning to development of molecular mechanics force-fields.

Awards and Recognition

None

Intellectual Property Review

This report has been reviewed by SRNL Legal Counsel for intellectual property considerations and is approved to be publicly published in its current form.

SRNL Legal Signature

Signature **Date**

Process Image Analysis using Big Data, Machine Learning, and Computer Vision

Project Team: Bruce Hardy (PI), Anna d'Entremont, Michael Martinez-Rodriguez, Brenda Garcia-Diaz, Lindsay Roy,
Subcontractor: USC:Jason Bakos, Taylor Clingenpeel, , Phil Moore, Ben Torkian, R. Doran
GT: AJ Medford, Suvadip Das
Thrust Area: SEM
Project Start Date: October 1, 2018

The development of algorithms for machine learning and data analysis for the 3013 MIS corrosion surveillance program is a collaborative effort by SRNL, USC and GT. For corrosion detection, LCM image data is extracted from large binary files, with software written to convert the data to physical attributes (i.e. height, color and grayscale values; all as functions of a location in a plane projection). The user interface for the software permits selective downloading of binary data and interrogation of attributes. User input thresholds are used to flag attributes of interest. Machine learning algorithms, developed for this application, are used to determine whether the features are the result of corrosion. To address the fundamental mechanisms of corrosion, machine learning algorithms are being developed to derive interatomic potential force-fields from ab-initio DFT calculations. The goal is to apply molecular modeling on a large enough scale to guide the design of resistant materials.

FY2020 Objectives

- Develop machine learning methods, based on computer vision, to analyze imaging data for corrosion.
- Develop machine learning methods for molecular modeling of corrosion processes.
- Identify 3013 data sets, and numerical methods, suitable for near-term development.
- Determine preliminary set of attributes for training supervised ML algorithms.
- Assemble training sets, train and test ML algorithms.
- Classify features by size, quantity, density, and location.
- Utilize computer vision to reduce amount of data needing manual analysis.
- Identify additional data sets within SRNL that can be analyzed using the methodologies developed as part of this project.
- Initiate development of ML methodology for obtaining adaptive force-fields from ab-initio molecular models (MM) for corrosion (added to originally approved scope).

Introduction

Halides contained in Pu-bearing material have been found and produce corrosion in the Inner Can Closure Weld Region (ICCWR) for the 3013 canister system used throughout the DOE complex. Inspections using an Laser Confocal Microscope (LCM) produce immense amounts of image data: approximately 6000 images per can, having 786,432 pixels per image, with 8 layers of data for each pixel. There is currently a 5-year backlog of images, with approximately 5 canisters/year, that must be evaluated. Simplistic computer-aided image analysis can flag parameters, such as pit depth and cracking to guide manual examinations for corrosion. However, while this approach greatly improves the efficiency of the examination process compared to unaided manual screening, it is still excessively time consuming. A more sophisticated approach is to assess the data using machine learning algorithms to identify corrosion without manual intervention. As a complement to corrosion detection, molecular level analyses can yield a fundamental understanding of corrosion occurring in the ICCWR and guide the design of corrosion resistant materials, welding processes, and coatings. These 2 efforts comprise the research in this LDRD.

This 2-year LDRD project had 2 concurrent objectives: The first was the development of machine learning algorithms to identify the presence of corrosion from a very large set of images generated by LCM scanning of 3013 canisters used to store Pu oxides. This portion of the LDRD constituted image analysis of a metal surface for the presence of corrosion. The image processing algorithms developed for this project are suitable basis for analysis of other types of corrosion data, which can be produced in vast quantities using modern analysis technology.

The second component of the LDRD consisted of the development of machine learning methodology used to develop molecular mechanics force-fields from ab-initio Density Functional Theory (DFT) calculations for corrosive attack by chlorides on 304L or 316L stainless steel. The long-term goal of this latter component was to understand corrosion on a fundamental level.

Approach

First Component – Image Analysis

Corrosion is strongly, but not exclusively, associated with surface pitting and cracking, coloration, along with shapes and patterns of surface features. Conversely, not all pits and surface lesions are the result of corrosion: some are artifacts of fabrication, impact, scoring or other non-corrosion events. Corrosion is identified via the combined properties of pit depth, area, edge contour, color and clustering. Software was developed to extract these features from large binary files generated by the LCM. The individual images, which collectively span the ICCWR were stitched together and corrected to eliminate the effect of curvature on measurement of the local height. Various methods were applied to the data to best relate it to presence of corrosion. Mathematical operations invoked for computer vision and image interpretation included, Deep Neural Networks (DNNs), gradient methods, statistical characterization, correlations and filters[1,2]. The processed data would be input to ML algorithms; labeled data for training, and afterwards data for evaluation by the trained ML algorithm. The process is shown schematically in Figure 1.

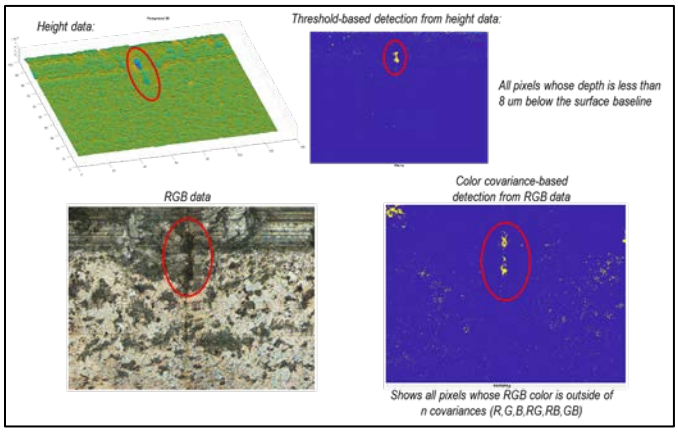

Figure 2. Crack and corrosion data from LCM images. Data channels include RGB, grayscale and height.

Second Component – ML based FF Derivation from DFT Calculations

The objective of the second part of the LDRD project is to advance the fundamental understanding of corrosion by developing novel methods to simulate the complex chemistry and physics through coupling quantum mechanical and empirical force field methods. In corrosion science, sophisticated multiscale models beginning at the *ab-initio* level provide mechanistic insight into metal-environment interactions resulting in general corrosion, intergranular corrosion, and pitting corrosion[3]. Specifically, this research focuses on designing machine-learning algorithms to develop and train adaptive force fields for the study and prediction of corrosion behavior, specifically the metal-environment interface[4]. Accurately calculating the parameters for a robust force field, however, is a much more complicated than a simple regression fit, requiring more sophisticated data analytics[5-7]. Further, the functional form of standard force fields is often insufficient to capture the complex physics of a reactive interface, especially in the case of the complex electronic structure of magnetic metal oxides. While atomistic modeling techniques are well suited to study the chemical reactions occurring at the interface between a material and its environment, modeling corrosion is computationally slow because the models must be constructed to

resolve both relevant reaction mechanisms and mass transport processes. By developing machine learning methods to obtain quantitative structure-activity relationships considering both molecular and bulk boundary conditions, researchers will be able to significantly advance knowledge by exploring more combinatorial spaces and nonlinear processes which are difficult using traditional approaches.

Results/Discussion

LCM image data taken for the MIS program was reviewed to obtain samples containing cracks, pits and other features characteristic of corrosion. The low incidence of corrosion and cracking in the actual ICCWR samples made it necessary to incorporate data augmentation schemes for proper training of the ML algorithms developed for this application. Images containing cracks were augmented by generating vertical and horizontal translations of the original labeled image. Capability for labeled image rotation was also developed. To provide an efficient means for handling large amounts of binary image data a GUI was developed to serve as an interface with the data files, manipulate and group images, label features for training the ML algorithm, group features with user defined thresholds, correct for sample tilt and curvature, stitch images, train ML algorithms, and apply the algorithms for crack and corrosion identification. Further, methods were developed to read binary WAMS data, which has recently been adopted for 3013 image interrogation. Studies conducted during FY 20 emphasized that larger views, represented by image tiles containing a larger number of pixels, improve the accuracy of crack detection by the ML algorithms.

After preprocessing, images obtained from LCM and WAMS data are partitioned into tiles (rectangular blocks of pixels). Image data used for training and testing ML algorithms is labeled. Cracks, pits and color patterns are all associated in various forms with corrosion. Pits can readily be detected using height data thresholds. Cracks, particularly “hairline” cracks do not always have a definitive height signature. Rather, crack identification is a combination of grayscale image intensity (pixel value) and height data. Initially, it was hoped that standard edge detection methods could be used with pixel values to extract crack edges. Methods considered included: erosion and dilation, blurring, Fourier and Gaussian filters, and gradient methods. Unfortunately, other surface features combined to create background noise that was similar in frequency to that associated with crack edges. To overcome this problem, DNN methods were developed as an attempt to identify cracks. Early in the development of this approach the training of the DNN algorithms suffered due to the small amount of crack data available. Training data was expanded by using augmented image data. Increasing the amount of labeled training data, adjusting the DNN algorithms, and increasing the image tile size from 64x64 to 112x112 pixels improved the precision and recall for crack identification. Examples of labeled training data for cracks, taken from LCM images are shown in Figure 3. It was found that the greatest accuracy was obtained by using a consensus drawn from an ensemble of randomly generated Convolutional Neural Network (CNN) algorithms, having the following characteristics:

- *2 channel input – grayscale contrast-adjusted peak intensity and smoothed height*
- *CNN depth of 1 to 5 layers, each layer consisting of a convolution layer using a RELU activation function and a maximum value pooling layer*
    - *Convolution layers have kernel sizes ranging from 1x1 to 3x3, with 1 to 128 feature maps*
    - *Pooling kernel size ranges from 1x1 to 3x3, with strides ranging from 1x1 to 3x3*
- *The final layer of the CNN consists of a fully connected layer of 1 to 128 neurons*

The consensus is a vote on whether or not an image contains a crack. Results from this classification method are shown in Figure 4, which compares consensus accuracy with that of a single model and shows that the ensemble reached an accuracy greater than that of any individual algorithm within it. It was found that that accuracy was improved when training sets contain an approximately an equal number of tiles with and without cracks.

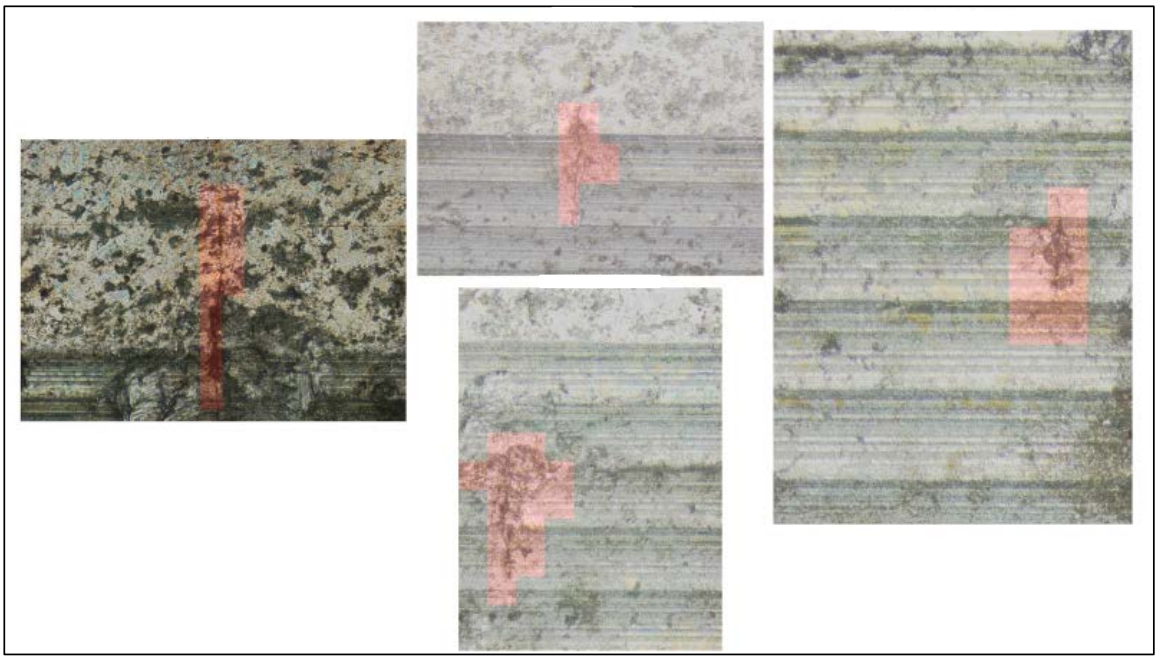

Figure 3. Examples of labeled data consisting of 112x112 pixel tiles (blocks). The training process used bootstrapped data samples that were augmented using horizontal and vertical translation. Highlighted regions are tiles that contain cracks.

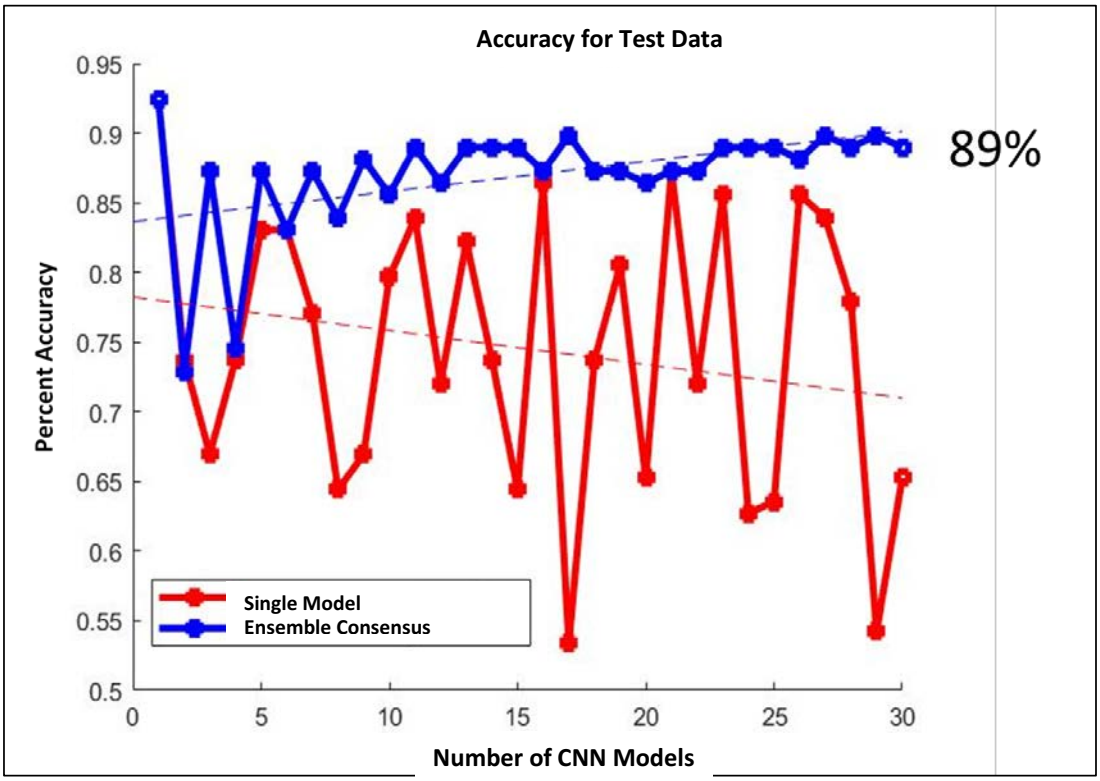

Figure 4. Comparison of the accuracy of crack identification from an ensemble consensus of CNN algorithms with the accuracy of a single model.

The second half of this project focused on establishing a pipeline for generating adaptive force fields that can be dynamically updated to match the results of quantum-mechanical calculations as closely as possible for corrosion on iron surfaces in water. There were two research routes established:

- Generating data with ReaxFF force field for iron/water;
- Establishing a workflow for force field construction of iron oxide systems based on a set of diverse atomic environments and force data.

The use of ReaxFF for generating initial sampling configurations provides an opportunity to sample a wide range of chemically relevant space by running molecular dynamics simulations at elevated temperatures or with enhanced sampling techniques such as metadynamics. In this work, a nearest-neighbor-based sampling technique is applied to identify a "maximum diversity" subset of simulations (Figure 5). The approach draws a uniform distribution from the high-dimensional fingerprint space, and the resulting subset can be used to improve the generality of neural-network models by ensuring that "rare" configurations from the tails of the distribution are included in training at all steps. Preliminary results indicate that the number of atoms that must be simulated can be decreased by 2-3 orders of magnitude using sub-sampling.

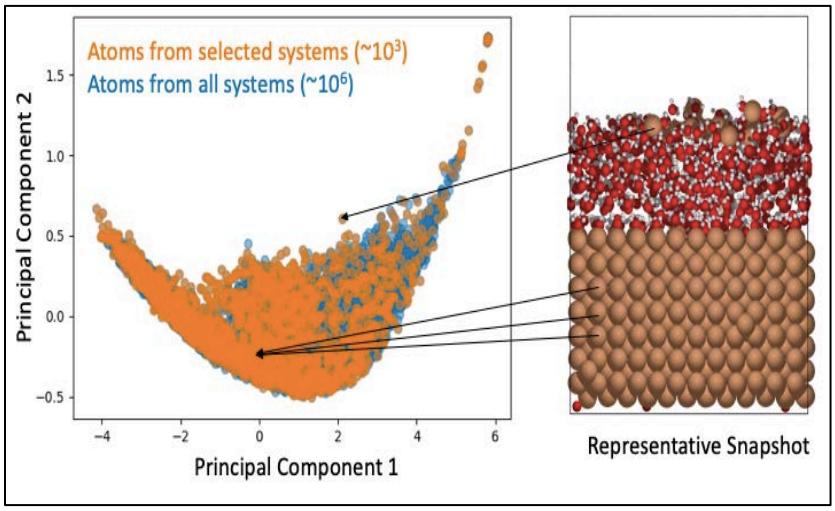

Figure 5. Illustration of sub-sampling algorithm applied to a collection of ReaxFF data for the iron/water interface. Individual atomic environments are displayed in PCA space (left) and representative atoms are qualitatively identified in a single snapshot of a molecular dynamics simulation (right).

The results of the initial ReaxFF Fe-O structures were then further analyzed to enable a wide range of training systems, including those that are explicitly similar to systems of interest. Since atomic forces are purely dependent upon the local environment, the construction of adaptive force fields is possible for any system where reliable quantum mechanical calculations can be performed. The first step requires that the reference data set for crystalline materials be representative of a variety of chemical environments, or defects (e.g. defect-free bulk, surfaces, point defects; see Figure 6) in both equilibrium

and non-equilibrium states. Calculation of the force component on each atom at equilibrium was then determined using DFT.

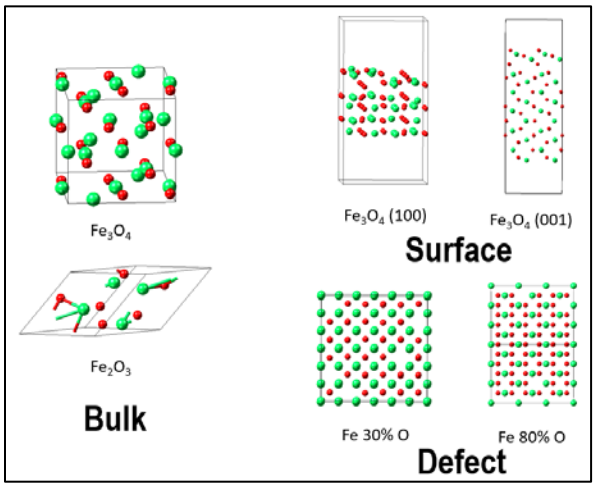

Figure 6. Reference configurations used to sample the Fe-O atomic environment for training and testing of force fields.

In a suitable representation for an atom and its environment, the force component must conform to any arbitrary direction and be invariant to basic atomic transformation operations (i.e., translation, rotation, permutation). One potential representation, or fingerprint, is:

$$V_i^u(\eta) = \sum_{i \neq j} \frac{r_{ij}^u}{r_{ij}} \cdot e^{-(\frac{r_{ij}}{\eta})^2} \cdot f_d(r_{ij})$$

where r_{ij} is the distance between atoms *i* and *j*, r^u_{ij} is a scalar projection of the distance along direction u, η is the gaussian function width, and f_d is a damping function for atoms within a cutoff distance. This fingerprint was used to identify the atomic forces for >4600 unique environments. The results of ReaxFF and DFT data can then be combined using techniques from transfer learning to adaptively generate force fields that approach DFT accuracy while minimizing the required number of DFT simulations. Finally, the approaches will be combined with global optimization schemes to help automate the process of identifying the most accurate neural network architectures.

FY2020 Accomplishments

- The functions developed for image analysis are now available through a user-friendly GUI that was developed specifically for this application. This includes:
    - Data input.
    - Data labeling, flagging of features of interest, and other diagnostics. Includes zoom capability.
    - 3D surface imaging (including color and height data).
    - Convolutional Neural Network (CNN) Machine Learning (ML).
        - Training methods can be implemented through the GUI.
    - ML algorithms are not yet in final form, but GUI access is modular so that updated algorithms can readily be imported, replacing the current ones.
- ML algorithms are not yet in final form, but GUI access is modular so that updated algorithms can readily be imported, replacing the current ones.
    - Tested different forms of ML algorithms.
        - Algorithms tested included well regarded CNN's: such as ResNet 50, AlexNet, and custom CNN's which gave the best performance so far.
    - Determined that overfitting (excessive response when applied to test data) and recall (a measure of the rate of false negative predictions) need to be improved in CNN algorithm development.
    - Found that deeper neural networks don't necessarily result in better performance.
    - Performed hyperparameter optimization, including genetic algorithm methods.
    - Due to number of hyperparameters internal gradients (a custom method) will be used for optimization.
    - The fine scale detail associated with cracks (in terms of grayscale and color intensity, along with height variation), complicates the process of distinguishing them from scratches, tooling marks, and sequences of pits protrusions on the surface.
- Developed Convolutional Autoencoder (CAE) to obtain transfer learning for CNN's used for image analysis.
    - The CAE supports transfer learning for accelerating the training of the CNN's, for the case of a relatively small amount of training data.
- Developed software to enable rapid labeling of corrosion training data (Figures 1 and 2).
    - Labeled data is necessary for training the CNN's. A means for rapid labeling is necessary due to the volume of training data required.
    - Completed software for mouse click labeling of corrosion training data for supervised learning (Figure 7).
    - Labeled pixels are retained during geometric transformation for data augmentation (Figure 8).
- Developed baseline software for data augmentation via simulated geometric variations (Figures 8-9).
- Developed software that enables reading of WAMS binary data files.
    - Will provide software to the LANL statistics group for their use.
    - Will compare surface contour data from WAMS with that from the LCM files.
        - Tests include proportionality between data sets for areas and heights of pits and protrusions.
- Collaborating with the Los Alamos National Laboratory (LANL) statistics directorate.

- USC/SRNL will provide LANL with a method for extracting binary data from WAMS images (a newly applied technology for producing images used to screen for surface defects).

- Generated baseline Fe-O atomic force data for the FF training set.
  - Calculated Fe/H2O interfaces using ReaxFF to illustrate the effectiveness of a weighted nearest-neighbor subsampling algorithm.
  - Created reference data set of iron oxide environments from quantum mechanical calculations for >4600 chemical environments.
  - Fingerprinted the atomic environment to enable mapping of atomic force components

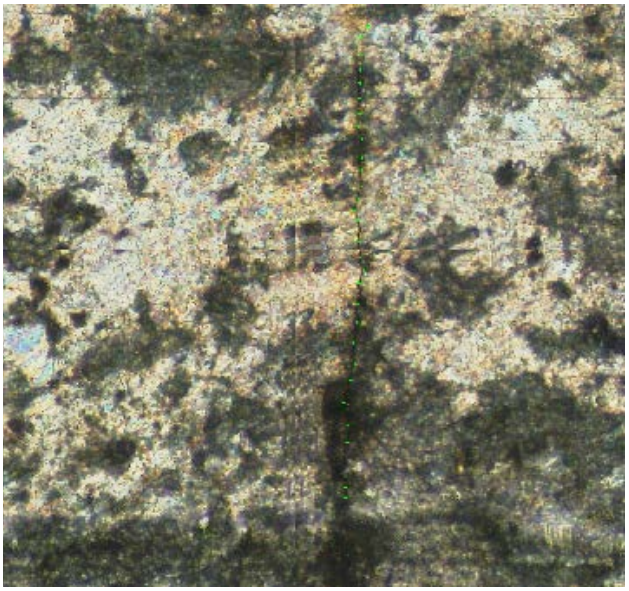

Figure 7. Pixels selected by mouse click are highlighted by green dots. The pixels are used to label the crack in rotational transformations that augment available data.

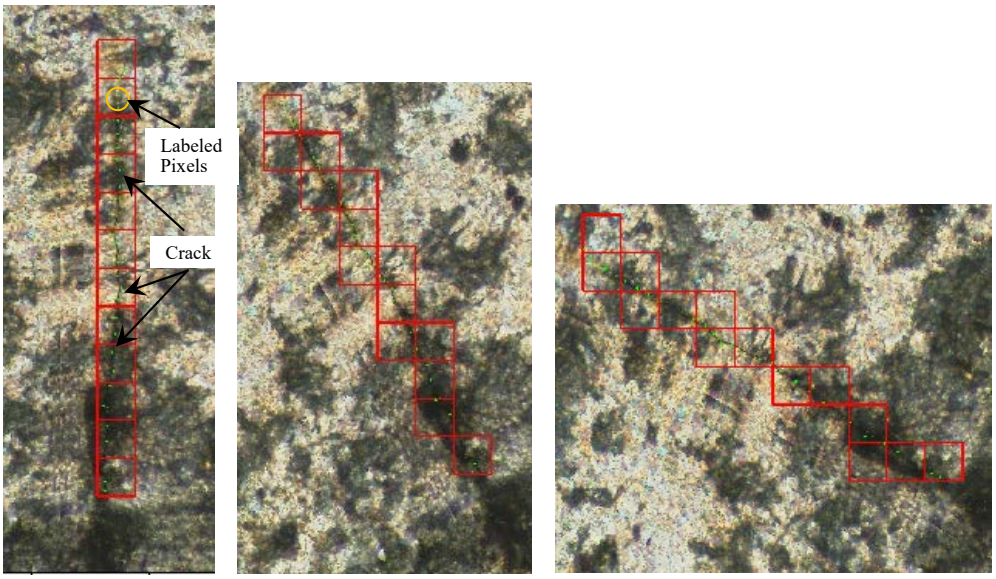

Figure 8. Data augmentation by rotation of crack image. The images are cropped show the highlighted region at a larger scale. Green dots (barely visible) along crack are pixels manually selected by the analyst by mouse click. The baseline orientation is shown in (A). When the image is divided into tiles, those containing a crack pixel are highlighted with a red boundary and labeled as a crack containing tile. Tiles that do not contain a crack pixel are defined and labeled as not containing a crack, but not highlighted. In (B) the image and crack pixels are rotated 30 degrees in the counterclockwise direction and the image is divided into tiles. Again, those tiles containing a crack pixel are highlighted with a red boundary and labeled as a crack containing tile. Tiles that do not contain crack pixels are defined and labeled as such, but unlabeled. Similarly, (C) shows a rotation 60 degrees in the counterclockwise direction.

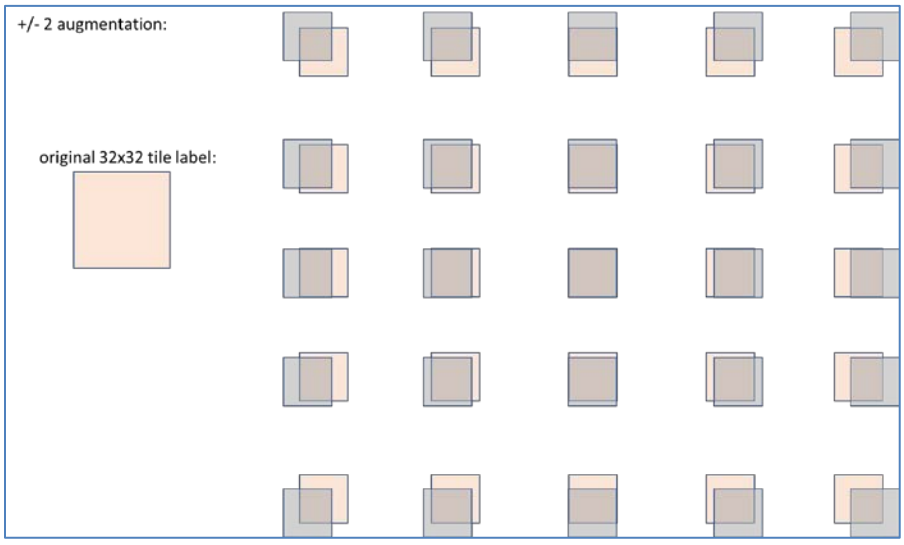

Figure 9. Data augmentation by translation of image.

Future Directions

- L-basin corrosion analysis.
- Extension of image analysis to inclusions in articles produced by additive manufacturing.
- Application of AI methods to data analysis, particularly for corrosion and material degradation. This would also include analysis of analytical data (XPS, XRD, SEM, etc.).
- Apply ML to material synthesis based on empirical data with imposed physical constraints.
- Applications to advanced process control, invoking reachability theory and fault tolerance. This aspect of AI, which takes advantage of the volume of data yielded by advanced sensor capability, would be particularly suitable for isotope separation, pit productions and waste processing.
- Development of surrogate molecular models having reduced complexity but retaining a high degree of accuracy. This is related to material design at a fundamental level and is a compliment to data analysis that is used to identify material degradation.
    - Submit proposals for more complete development of ML applications for FF derivation from DFT calculations, including experimental validation.
    - Test FF algorithm with validation set
    - Incorporate iron/iron oxide defects with different water phases
    - Replace water with halides for corrosion and reactivity

FY 2020 Peer-reviewed/Non-peer reviewed Publications

None.

Presentations

None.

References

1 Ian Goodfellow, Yoshua Bengio, and Aaron Courville. Deep Learning. MIT Press, Cambridge MA (2016)

2 Aurélien Géron. Hands-On Machine Learning with Scikit-Learn &Tensorflow. O'Reilly, Sebastapol, CA (10/12/2018).

3 Gunasegaram, D. R.; *et al.*, *Int. Mater. Rev.* **2014**, *59*, 84.

4 Ghosh, S.; Suryanarayana, P. *Comput. Phys. Commun.* **2017**, *216*, 109.

5 Roy, L. E.; Jakubikova, E.; Guthrie, M. G.; Batista, E. R. *J. Phys. Chem. A* **2009**, *113*, 6745.

6 Joyce, J. J.; *et al.*, *Mater. Res. Soc. Symp. Proc.* **2010**, *1264*, Z09-04.

7 Li, W.; Ando, Y. *Physical Chemistry Chemical Physics* **2018**, *20*, 30006.

Acronyms

AI	Artificial Intelligence
AIMD	Ab initio molecular dynamics
CAE	Convolutional autoencoder
CNN	Convolutional Neural Network
CS	Computer Science
DNN	Deep Neural Network
DOE	Department of Energy
DFT	Density Functional Theory

EAM	Embedded Atom Model – an interatomic potential that represents the energy between atoms
FF	Molecular mechanics Force-Field
GT	Georgia Institute of Technology, Atlanta, GA
GUI	Graphical User Interface
ICCWR	Inner Can Closure Weld Region
LCM	Laser Confocal Microscope
MD	Molecular Dynamics
MIS	Material Identification and Surveillance program
ML	Machine Learning
MM	Molecular Models
NN	Neural Network
TIP3P	Transferrable Intermolecular Potential with 3 Points – a 3 site rigid water model
USC	University of South Carolina, Columbia, SC
WAMS	Wide Angle Microscope System

Intellectual Property

None.

Total Number of Post-Doctoral Researchers

1 Post-doctoral researcher, performed work at GT.

Total Number of Student Researchers

2 Undergraduate students, performed work at USC
1 Graduate student, performed work at GT
1 MSIPP student, performed work at SRNL

External Collaborators (Universities, etc.)

The University of South Carolina, Columbia, South Carolina.
The Georgia Institute of Technology, Atlanta, Georgia.