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Two-dimensional Corrosion Pit Initiation and Growth Simulation Model

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Abstract: A two-dimensional corrosion initiation and growth model for aircraft aluminum materials is developed. The model takes into account the electro-chemical parameters as well as specific rules governing corrosion mechanisms. The simulation program is implemented in a cellular automata framework. The corrosion initiation and growth patterns obtained from simulations are compared qualitatively and quantitatively to the experimental data obtained from the Center for Materials Diagnostics at the University of Dayton Research Institute, Dayton. The results indicate that the present model effectively captures the corrosion damage process including initiation and growth. The effects of various electro-chemical parameters on the damage growth obtained from the simulation are presented and discussed. The results presented illustrate a new approach to modeling corrosion damage in aircraft aluminum materials. The simulation program is developed in a JAVA environment for ease of portability and usability.

keyword: Pitting, Initiation, Growth, Simulation Model, Aluminum

1 Introduction

Pitting corrosion is known to be one of the major damage mechanisms affecting the integrity of many aerospace structures, associated with the dissolution of metal caused by the breakdown of the passive film on the metal surface. Corrosion pits generally initiate due to some chemical or physical heterogeneity at the surface, such as inclusions, second phase particles, flaws, mechanical damage, or dislocations. The aluminum alloys contain numerous constituent particles, which play an important role in corrosion pit formation [Wallace and Hoppner (1985)]. To better understand particle-induced pitting corrosion in 2024/T3 and 7075/T6 aluminum alloys, optical microscopy, scanning electron microscopy (SEM) and Transmission Electron Microscopy (TEM) techniques have been used [Wei, Liao and Gao (1998)]. Due to an aircraft's special service environments (e.g. salt water), electrochemical reactions are possible and corrosion pits are readily formed between the constituent particles and the surrounding matrix in these alloys. It is well known that corrosion pitting has a strong effect on the fatigue life of aluminum alloys used in aircraft structures [Wei, Liao and Gao (1998); Pao, Feng, and Gill (1999); Hoppner (1979)]. Fatigue cracks usually initiate from the corrosion pit sites. Under the interaction of cyclic load and the corrosive environment, cyclic loading facilitates the pitting process, and corrosion pits, acting as geometrical discontinuities, lead to crack initiation and propagation and then final failure. Corrosion can lead to accelerated failure of structural components under fatigue loading conditions. Prediction of corrosion damage growth is very important for the structural integrity of aircraft materials and structures.

The corrosion mechanisms depend on the material composition, electrolyte and other environmental conditions [Marcus and Oudar (1995)]. Recently, Burstein et al. (2004) presented a good discussion on the origins of pitting corrosion, and demonstrated that pit nucleation is at the microscopic level, and some metals show preferential sites of pit nucleation. Most of the previous work on corrosion has been focused on chemical processes and electric currents and potentials, and limited growth models [Shreie, Jarman and Burstein (1994); Strehblow (1995); Simon, et al. (2000); Frantiziskonis, et al. (2000); Aziz and Godard (1952)]. Several pitting corrosion models exist in the literature, [Godard (1960); Johnson (1971); Rowe (1976); Hoeppner (1979); Turnbull (1993); Kawai and Kasai (1985); Dallek and Foley (1978)] which are mostly empirical or phenomenological in nature. Over

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the years, Macdonald and his research group have developed deterministic models and algorithms for predicting the accumulation of damage due to localized corrosion pitting, crevice corrosion, and stress corrosion. [Engelhardt and Macdonald (1989, 1998); Macdonald and Urquidi-Macdonald (1990, 1992); Macdonald (1996); Engelhardt,Urquidi-Macdonald and Macdonald (1997); Engelhardt, Macdonald and Urquidi-Macdonald (1999); Macdonald and Engelhardt (2003)] Their approach is called the "Damage Function Analysis (DFA). It has a physico-chemical basis and incorporates environmental and mechanical parameters.[Macdonald and Engelhardt (2003)] The DFA approach to pitting corrosion includes the nucleation, propagation, and repassivation stages explicitly defined in the prediction and constrained by natural laws (conservation of mass, conservation of charge, and Faraday's law). Even though the DFA approach has been applied to many industrial situations and the predictions are reasonable, the approach is very analytical and deterministic.

Artificial Neural Networks (ANN) has also been applied to pitting corrosion [Lu and Urquidi-Macdonald (1994); Wei and Harlow (1993)]. Even though ANN doesn't contain any empirical or deterministic models or explain the physics of the localized corrosion, it is still being used to predict future behavior with various parameters. Probabilistic approaches have also been used to model localized corrosion [Harlow and Wei (1995, 1998)]. Pitting corrosion is a very complex process and may involve many mechanisms. In general, the corrosion damage function should involve not only physico-chemical and environmental factors, but also various parameters, random in nature. Therefore, a more realistic corrosion damage function should integrate various parameters from solid mechanics, surface- and electro-chemistry, materials science, probability and statistics, and fracture mechanics [Farmer, Bedrossian, and Mccright (1998), Aoki, Amaya, Urago, and Nakayama (2004)]. In addition to various deterministic and empirical methods in the literature, there might be a need to develop computational models that can predict the pit growth morphology as well as the mechanical parameters for structural integrity analysis of corrosion damage tolerance technology.

In order to complement empirical, phenomenological, ANN and DFA approaches in the literature, there should be alternate simulation approaches, which account for the morphology and material loss in the prediction of pitting corrosion. Recently, [Pidaparti, Palakal and Fang (2004)] developed a cellular automation approach to model aircraft corrosion pit damage growth behavior. Prediction of pitting corrosion morphology and its growth is very important for structural integrity analysis, and when coupled with periodic inspections, it might be a valuable approach for avoiding corrosion related failures in general. The main objective of this work is to develop a corrosion damage initiation and growth model using specific rules governing the electro-chemical reactions, and simulate using cellular automata environment. The results of corrosion damage growth obtained from simulation are compared qualitatively as well as quantitatively to the experimental data obtained from the Center for Materials Diagnostics at the University of Dayton Research Institute, Dayton. The effect of various electro-chemical parameters on the corrosion damage growth obtained from the simulation are presented and discussed.

2 Pit initiation and growth model

Corrosion begins with small imperfections in aircraft materials referred to as "pits". Usually, once a pit is initiated in a certain near-neutral aqueous solution, the pit will continue to propagate due to the fact that the solution within the pit becomes acid, and the alumina is no longer able to form a protective film close to the metal. Further, pitting of metal results when the aluminum ions migrate away from the areas of low pH, and alumina precipitates as a membrane by isolating and intensifying local acidity [Marcus and Oudar (1995); Burstein et al. (2004); Shreie, Jarman and Burstein (1994)]. The first step of the pit initiation process is the formation of embryos. The nucleus of the pit is referred to as an embryo. Once the pit has nucleated, the embryo requires certain factors, both physical and environmental, to build up into a pit. The concept of embryos was introduced by Farmer, et al. (1998), among others, who used the term embryo to describe the first stage of pit initiation. Embryos denote the stage before the initiation step where they may survive or not depending on several conditions. The birth of embryos depends on the birth probability, λ_1 given by the equation:

$$\lambda_1 = \lambda_0 [Cl] \exp\left\langle \frac{\alpha\lambda F}{RT} (E - E_{crit}) \right\rangle \tag{1}$$

where

λ_1 is the birth probability

λ_0 is the intrinsic rate constant for birth of embryos

Cl is the Chloride concentration of solution

α_λ is constant

F is faraday's constant

R is universal gas constant

T is the absolute temperature

E is the electrochemical potential applied to the surface

E_{crit} is the critical potential over which pitting takes place

Since the embryos have a tendency to repassivate if they are not provided with the right environmental conditions, a death probability occurs. In case the embryo satisfies the death probability, it repassivates. The death probability, μ_1, is given by the equation:

$$\mu_1 = \mu_0 \left[OH^-\right] \exp\left\langle -\frac{\alpha_\mu F}{RT}\left(E - E_{pass}\right)\right\rangle \tag{2}$$

where

μ_1 is the death probability of the embryo

μ_0 is the intrinsic constant for death of embryo

OH^- is the concentration of hydroxyl anion

α_μ is constant

E_{pass} is the potential over under which the embryo repassivates

In the simulation model, the birth and death probabilities can be more than 1 depending on the environmental and chemical parameters, an assumption is made that if the probability exceeds 1, it is adjusted to 1, which means that in the case of birth probability, each cell will corrode, while in the case of death probability, each cell will re-passivate. It may be noticed that there is competitive adsorption of Cl^- and OH^-. The embryos are the first stage of the pitting process as they initiate the growth of pits. Once the embryo is formed, the next stage is the transition from embryo to Pit. This transition rate, γ_1, is given by the relation

$$\gamma_1 = \gamma_0 \exp\left(-\frac{A\gamma}{RT}\right) \tag{3}$$

where

$\gamma_1$is the transition rate from embryo to stable pit

γ_o is the intrinsic rate constant

A_γ is the apparent activation energy

The transition stage is actually the meta-stable stage in which the pits are still in the initiation stage. They are still susceptible to repassivity and haven't entered the growth stage. In a review of the temperature dependence of pit initiation, Szklarska-Smialowska (1986) found considerable variability. Arrhenius behavior was not always observed, and the tendency for pitting was found both to increase and decrease with increasing temperature. In our model, in the case of pit initiation, an increase in temperature increases λ and μ while decreasing γ_1. The increase in λ and the decrease in γ_1 increase the tendency for pit initiation as the temperature rises, while the increase in μ decreases the tendency for pitting.

Another variable that has been considered is the induction time. Induction time is the age that an embryo must reach before it becomes a stable pit. This quantity obeys an Ahrenius-like expression

$$\tau_1 = \tau_0 \exp\left(-\frac{A\tau}{RT}\right) \tag{4}$$

where

$\tau_1$is the Induction time

$\tau_0$is the intrinsic induction time

A is the apparent activation energy

T is the absolute temperature

In the simulation model, the transition stage continues from 1 till 32. After the pit reaches a value of 32, the pit enters the growth stage and cannot repassivate. It will continue to grow until the whole area gets completely corroded. The rate at which the growth will take place depends upon the local rules of the cellular automata as described in the section below, and the growth rate formula is given by

$$\ln\left(\frac{\Delta r}{\Delta t}\right) = 13.409 - \left(\frac{5558.7}{T+273}\right) - 0.087\,(pH) + 0.56965\,(Conc) \tag{5}$$

where

$\Delta r/\Delta t$ is the growth rate of the pit

T is the absolute temperature

pH is the pH of the solution

$Conc$ is the concentration of the solution

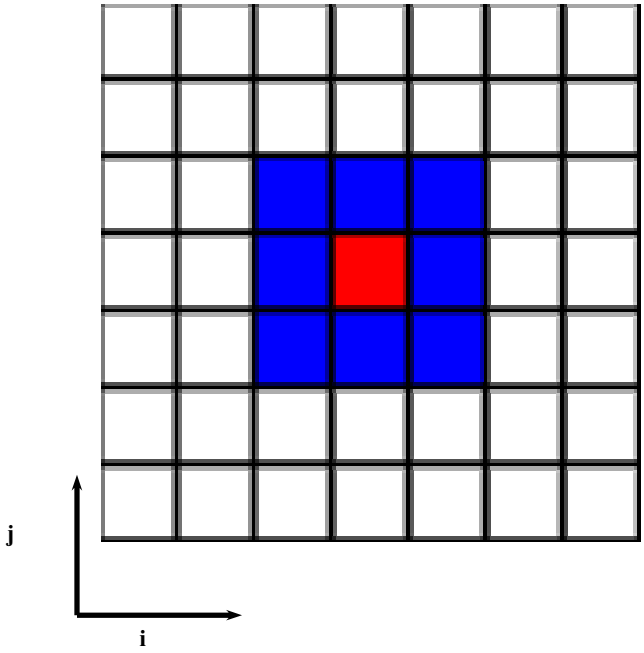


Figure 1 : The Moore neighborhood window for the simulation model.

3 Simulation algorithm and implementation

The above corrosion pit initiation and growth model was implemented as a discrete dynamic system model using the cellular automata approach. Cellular automata (CA) based modeling techniques are powerful methods to describe, simulate, and understand the behavior of complex physical systems [Chopard and Droz (1998)]. The original CA model proposed by Von Neumann [Chopard and Droz (1998)] is a two-dimensional square lattice in which each square is called a *cell*. Each of these cells can be in a different state at any given time. The evolution of each cell and the updating of the internal states of each cell occur synchronously and are governed by a set of *rules*. The cellular space thus created is a complete discrete dynamical system. Earlier work by Wolfram (1986, 1994) showed that the CA as discrete dynamical system exhibits many of the properties of a continuous dynamical system, yet CA provide a simpler framework. In the present simulation, a Moore neighborhood as shown in Fig. 1 is considered for implementing the cellular automata approach. The Moore neighborhood is a square-shaped neighborhood which limits interactions of an individual to its eight neighbors which share a vertex or edge with the center cell.

The cellular automata rules governing pit initiation and growth are as follows:

1. $S(u,t)$ is the initiation state of an un-corroded cell u at time t.

2. $E(u,t)$ is the initial state of a corroded cell u at time t.

3. The birth probability and death probability rates are set or varied according to the simulation.

4. For each simulated cycle, each un-corroded cell gets corroded according to the birth probability. In case the cell doesn't satisfy the birth probability condition, for each un-corroded cell, $S(u,t)$, the Moore neighborhood is considered. If more than four neighbors are embryos ($E(u,t)$ state), a corrosion pit is initiated at cell u and its corrosion state $S(u,t)$ is set to 1.

5. For each corroded cell, $E(u,t)$, check the death probability. If the cell satisfies the death probability condition, the cell repassivates. Also, check

the Moore neighborhood. A corroded cell needs at least one of its neighbors to be in the corroded state $E(u,t)$. If none of the neighbors is corroded, the cell repassivates and its state is set to $S(u,t)$.

6. For each corroded cell, $E(u,t)$, if there exists more than one neighbor with a corroded state, apply the transition rate, $E\ (u,\ t+1) = E\ (u,\ t) + r$, where r is the transition rate according to the transition rate formula. The transition rate takes place on cells with states between 4 and 32.

7. For the next time step, repeat step (4) on the uncorroded cells, and steps (5) and (6) on the corroded cells. According to the simulation, the pits may die till they reach the state 4. After $E(u,t)$ becomes more than 4, the pits cannot die, but they enter the meta-stable state. If the pits reach state 32, they finally initiate. At this stage the pit growth algorithm is applied on the corresponding cells.

In the initiation stage, the pits are in metastable state, i.e., in cases where they are not provided with the ideal conditions necessary for corrosion, they may die or repassivate. The metastable pits do not grow according to the formulae governing the growth rate, but are dependent on the transition rate. Once $E(u,t)$ reaches the value of 32, the pits become stable and follow the growth rate equation.

The governing rules for corrosion growth are as follows:

The cell in which pit has initiated acquires state, $C(u,t)$. $C(u,t)$ extends from 32 to 255 at which stage the pit corrodes completely.

At each time step, a partial corroded cell becomes further corroded to a degree given by pitting effect of the corroded cell on itself, and the corroded neighbors in the Moore neighborhood.

During the corrosion growth process, a fully corroded cell becomes an empty cell, i.e., all the material is gone and it has minimum effect on the corrosion of the neighboring cell.

It is important to note that we have not considered the film properties and the crystal orientation of metal as our main focus is on the effect of environmental factors on the rate of growth of corrosion.

The cellular automata rules and electro-chemical formulae described in the previous sections were implemented by developing a computer program. The simulation model is implemented in object-oriented programming with JAVA environment by taking each data set as an individual object. This way the future extension and modification of the simulation model functionality will be handled conveniently. Corrosion simulation data was obtained for this program using similar chemical parameters used in the experimental data by the Center for Materials Diagnostics at the University of Dayton Research Institute, Dayton. The various parameters were changed and modified to compare and observe the initiation and growth of corrosion in different environments.

The process of pit initiation requires special care because it has been observed that if the pits/embryos do not find the right resources or environment, they die. In other words, the nucleated pits get repassivated. The electrochemical explanation to this process is that in some duration of time, the oxide layer at the pit region redevelops so that the effect of the pit is negated. Modeling of stable pit growth began under the assumption that, once a stable pit forms, its depth increases continuously with time. This occurs when a pit moves from the transition stage to the growth stage. The growth increases until the metal is completely corroded, or in other words, the metal gets empty.

Each iteration of the simulated program is called a simulation cycle. Using experimental data, a rough estimate of the relation between the number of simulated cycles and the actual time has been calculated. This has been done by comparing the features of the experimental data and the simulated data at particular intervals and matching the same level of corrosion damage.

4 Results and discussion

To illustrate the corrosion pit initiation and growth process, a simulation is run for 200 cycles with initial environmental parameters as pH=3.5, Potential=0.6V, Concentration=290 (moles dm^{-3}), and Temperature=290 ^{0}F. Figure 2 shows the results of simulation at 8 intervals (cycles, n=60, 100, 125, 150, 165, 175, 190, and 200). It can be seen from Fig. 2 that at simulation cycle of 60, the pits are getting initiated and at cycle of 100, there are 4 distinct areas where pits are initiated. With increasing time (cycles), the initiated pits are growing in size, and new pits are also being initiated. As the cycles are increased, more pits initiate and grow, and finally at cycle 200, about 70-80% of the area is corroded. If we let

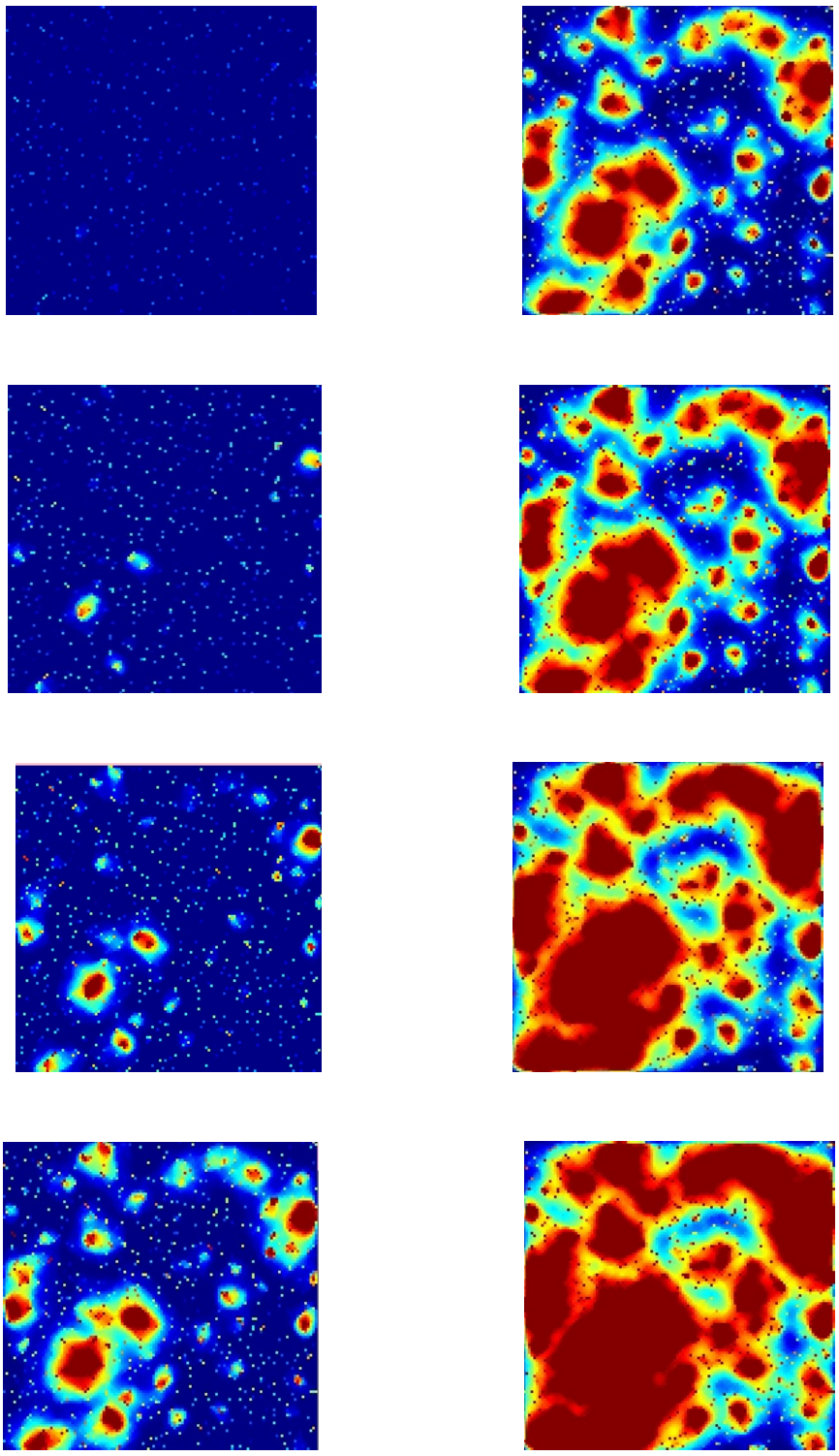


Figure 2 : Simulated images of corrosion initiation and growth (blue: non-corroded; and red: fully corroded) at various time cycles (n = 60, 100, 125, 150, 165, 175, 190, and 200)

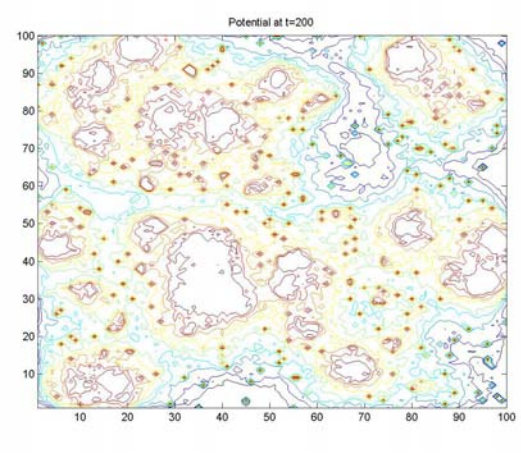
the simulation continue for some more cycles, the whole area will be corroded. The distribution of potential, pH, and temperature over the surface at cycles n = 200 is presented in Fig. 3. It can be seen from Fig. 3 that each of the parameters behaves very differently at the surface. It is useful to know that corrosion initiation and growth is different with various parameters affecting differently. It is very difficult to validate this result experimentally since it requires special instrumentation. Usually, corrosion experiments are done in a cell, by controlling the area to be corroded and monitoring the potential. Due to the nature of the electro-chemical process, the pH and temperature change during the corrosion process were not monitored. In such experimental cases, our simulation results should be helpful to further understand the effect of various parameters spatially on the metal surface.

To validate our two-dimensional corrosion pit initiation and growth simulation model with the experimental data, we extracted certain features from both the corrosion simulation images and experimental images, and then performed a feature analysis to quantitatively compare the results. This type of comparison is similar to image processing techniques [Umbaugh (1998)] for comparing two images. We utilized histogram features for this analysis. The histogram features that we extracted from an image are statistically based, where the histogram of the image is considered as the probability distribution of the pixel values. These features provide us the characteristic information of the pixel value distribution for the corrosion damage process. Five histogram features (mean, standard deviation, skew, energy and entropy) are obtained through the image feature analysis process [Umbaugh (1998)]. Based on our experience it was found that energy and entropy features correlate well with experimental data as compared to other features. The energy and entropy features are obtained using the following equations.

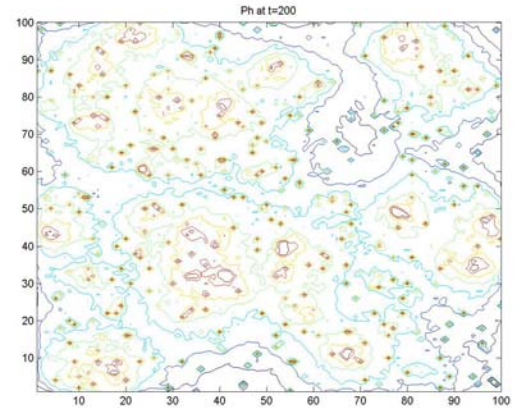
$$ENERGY = \sum_{g=0}^{L-1} [P(g)]^2 \tag{6}$$

$$ENTROPY = -\sum_{g=0}^{L-1} P(g) Log_2[P(g)] \tag{7}$$

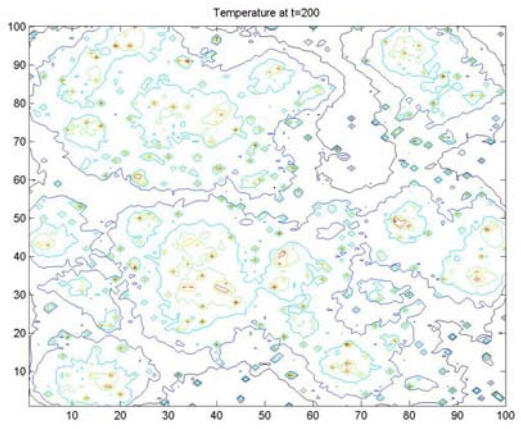
where P(g) is the histogram probability of pixel value, g, and L is the range of pixel values depicting the amount



(a)



(b)



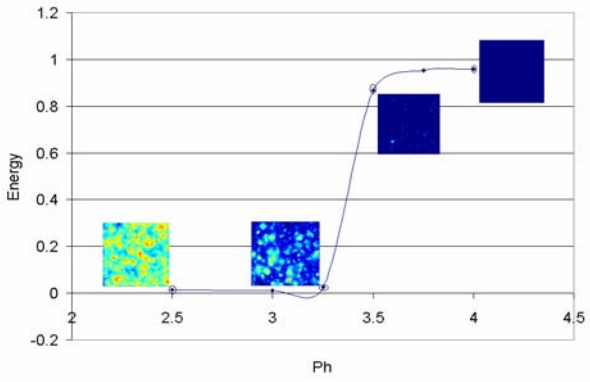
(c)

Figure 3 : Contours of (a) potential, (b) pH, and (c) temperature at simulation cycle (n=200).

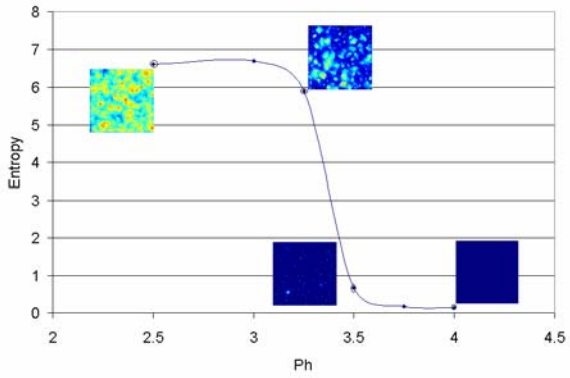
of corrosion (0-255 in the present case). The energy feature (degree of corrosion) describes how the pixel values are distributed in the region under consideration. In the corrosion damage image, energy feature indicates the degree of corrosion at the pit level. Entropy is an important measure of digital information as it indicates the number of bits we need to code the image data.

Since the model is dependent on several parameters, it is important to observe their effects on the corrosion process by changing these parameters. For each analysis, only one parameter was varied while all other parameters were kept constant. During the simulations, different sets of experiments were performed, changing the sensitive parameters such as concentration, potential, pH and temperature; and the values of energy, entropy, volume (area/unit thickness), and skew were measured. These values were compared with the experimental values. For example, the variation of energy, entropy and volume change with pH is shown in Fig. 4. It can be seen from Fig. 4(a) that energy is almost constant until a pH value of 3.2 or so, and then increases after that and reaches a steady state around pH value of 4. On the other hand, entropy follows an opposite trend, in that the entropy decreases between 3 and 3.5 and reaches a steady state at pH value of 4. It can be seen from Fig. 4(c) that the volume of corroded material decreases as pH value increases from 2.5 to 4. Similar results are obtained when other parameters like potential, concentration, and temperature are varied. Figure 5 shows the variation of energy, entropy, and volume features with simulation cycles. It is interesting to note that energy decreases in a non-linear fashion up to 160 simulations and then increases again up to 250, and stays constant. The entropy feature follows the opposite trend, i.e., it increases up to 160 simulations and then decreases after that. Interestingly, there is no corrosion up to 60 cycles. This result indicates that the volume feature is almost zero, and then increases non-linearly up to 250 cycles, by which time the whole surface is completely corroded. The results presented in Figs. 4 and 5 summarize the effects of parameters and features on the corrosion growth process.

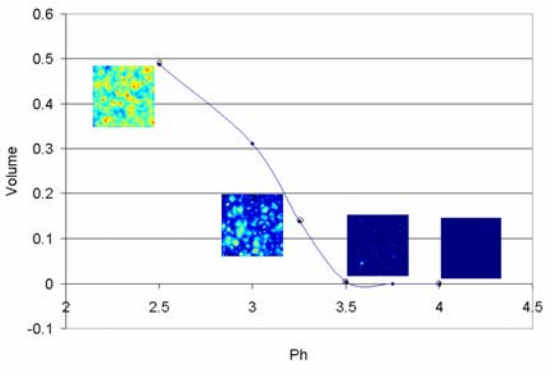
In order to validate our model quantitatively, we simulated corrosion corresponding to the experimental data obtained by the Center for Material Diagnostics (Mr. Chris Kacmar), Dayton, Ohio. Of the various features considered, we found that energy and entropy showed significance with various parameters in the corrosion



(a) Energy

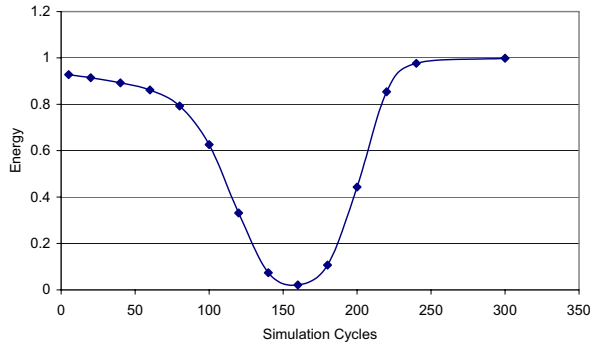


(b) Entropy

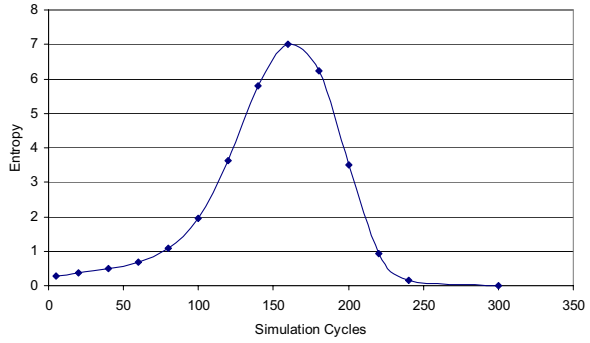


(c) Volume

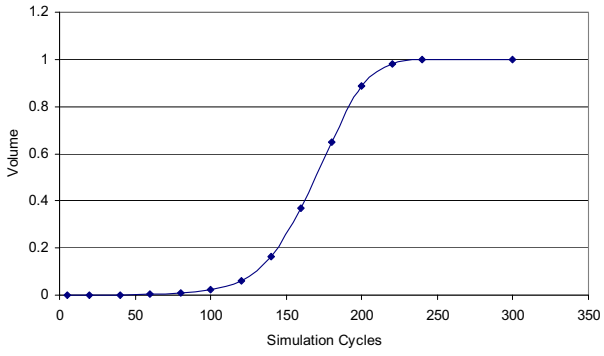
Figure 4 : Variation in Energy, Entropy, and Volume with change in pH value in the simulation model.



(a) Energy



(b) Entropy



(c) Volume

Figure 5 : Variation in Energy, Entropy, and Volume with number of simulation cycles in the present model.

damage process. The result of the features comparison is shown in Fig. 6. It can be seen from Fig. 6 that the histogram features like entropy and energy from the simulation results are in good comparison with the experimental data. The comparison of simulated results with experimental data is presented in Fig. 6. It can be also seen from Figs. 6 that the scatter in the experimental data is very large, and this is due to various parameters affecting the corrosion damage process and not knowing all the values of parameters in experiments. From the comparison in Fig. 6, we can see the areas bounded by the changing chemical parameters are in high accordance with the Dayton experimental data. This shows that given the proper chemical parameters as inputs, our simulation model can reasonably approximate the corrosion process. Also, the features extracted from the simulation are very close to the features from experiments, and therefore can be used as indications of the corrosion material loss if combined with a prediction model. Overall, the simulation results presented in Figs. 4-6 validate the corrosion damage process, giving the right trends.

5 Concluding remarks

A two-dimensional corrosion initiation and growth model for aircraft aluminum materials is developed. The model takes into account the electro-chemical parameters as well as specific rules governing the corrosion mechanisms. The simulation program is implemented in a cellular automata framework and JAVA environment for ease of portability and usability. The corrosion initiation and growth patterns obtained from simulations are compared qualitatively as well as quantitatively to the experimental data obtained from the Center for Materials Diagnostics at the University of Dayton Research Institute, Dayton. The results of the comparison indicate that the present model can capture the corrosion damage process including initiation and growth. The effect of various electro-chemical parameters on the damage growth obtained from the simulation are presented and discussed. The proposed simulation model is a general solution to structural corrosion in materials so that it can be scaled up to other structural materials like steel alloys, etc. Our future plans are to seek a more fundamental understanding of the macro- and micro-level corrosion growth and to model this based on local rules that can provide valuable information and tools for designing corrosion resistant materials for a variety of applications.

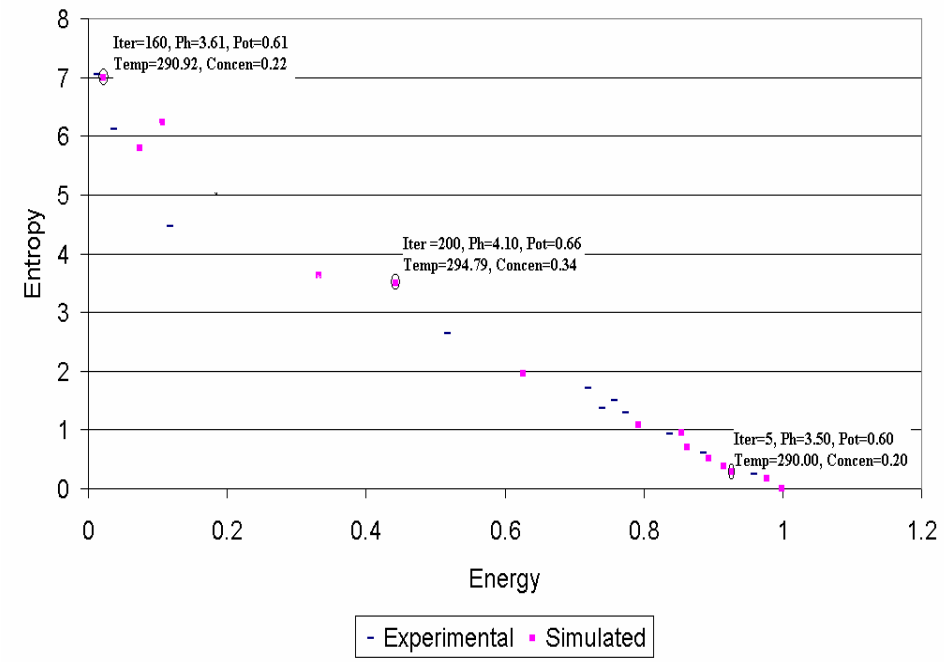


Figure 6 : Validation of simulation model by comparison of extracted features (Energy & Entropy).

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