Non-equilibrium Statistical Theory of Pitting Corrosion in Metallic Materials

Authors: You Yanhua, Wang Binrong, Hu Haiyun, Hu Haiyun

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Abstract

Abstract Metal materials often suffer from corrosion due to environmental influences during service, which greatly reduces metal Abstract Metal materials often suffer from corrosion due to environmental influences during service, which greatly reduces their service life; therefore, studying the corrosion behavior of metal materials has strong practical significance. Based on existing metal corrosion theories, this paper introduces a corrosion current decay constant, derives the relationship between metal corrosion current density and corrosion time using the transient process of impedance circuits, analyzes the effect of temperature on pit growth, and then obtains the growth rate of metal pits and the relationship between corrosion depth and time through Faraday's formula. Based on non-equilibrium statistical theory, the probability density distribution function of pitting is derived by establishing and solving the Fokker-Planck equation, and the perforation probability and reliability are calculated using the weakest link model. Finally, taking aircraft aluminum alloy LD2 structural material as an example, the pitting depth, rate, and probability density distribution function are calculated, and the most probable pitting depth value, perforation probability, and reliability of the material at different corrosion years are obtained, with the expectation of applying them to pitting life prediction and reliability analysis of metal materials.

Full Text

Non-Equilibrium Statistical Theory of Pitting Corrosion in Metal Materials

You Yanhua, Wang Binrong, Hu Haiyun
School of Physics, Beijing Institute of Technology, Beijing 100081, China; huhy@bit.edu.cn

Abstract

Metal materials are widely used in various fields. Due to environmental influences, materials are prone to corrosion during service, which significantly reduces their service life and reliability. Therefore, studying the corrosion behavior of metal materials holds substantial practical significance. Building upon existing metal corrosion theories, this paper introduces a corrosion current attenuation constant and derives the relationship between metal corrosion current density and corrosion time using the transient process of an LR circuit. The influence of temperature on pitting growth is analyzed, and the time-dependent relationships for pitting growth rate and corrosion depth are obtained through Faraday' s law. Based on non-equilibrium statistical theory, the probability density distribution function of pitting is derived by establishing and solving the Fokker-Planck equation, and the perforation probability and reliability are calculated using the weakest link model. Finally, taking aircraft aluminum alloy LD2 structural material as an example, we calculate its pitting depth, growth rate, and probability density distribution function, obtaining the most probable pitting depth values, perforation probability, and reliability at different corrosion ages, with the aim of applying these results to pitting life prediction and reliability analysis of metal materials.

Keywords: Pitting corrosion; Non-equilibrium statistical theory; Probability density function
CLC number: TG171

Introduction

Metal materials are extensively utilized in industrial and domestic applications due to their high mechanical strength, excellent heat resistance, and dimensional stability. However, metals are susceptible to corrosion when exposed to natural or industrial environments. Pitting corrosion is a particularly destructive form of localized corrosion that can cause catastrophic failure in materials, making it significantly more hazardous than uniform corrosion [1-2]. Current research on pitting corrosion in metal materials primarily focuses on experimental and computational studies of pitting depth and growth rate [3-5], as well as the effects of temperature and chloride ion concentration on pitting behavior [6-7]. Caleyo et al. [8-10] established a stochastic pitting model using Monte Carlo simulations, proposing that pit nucleation follows a non-homogeneous Poisson process with nucleation times obeying a Weibull distribution, while pit growth follows a non-homogeneous Markov process. Based on this theory, they derived a maximum pit depth model and calculated the temporal probability distributions of pitting depth and rate in underground pipelines. Elahe et al. [11] employed a non-linear Markov process to develop a maximum pit depth model and evaluated failure probability and remaining life by calculating the pressure-bearing capacity of corroded materials. Pan et al. [12] compared the pitting characteristics of nanocrystalline 304 stainless steel with conventional 304 stainless steel using electrochemical measurements and atomic force microscopy, finding that pit initiation sites in nanocrystalline materials shifted to grain boundaries, facilitating metastable pitting while reducing the probability of stable pit formation, thereby demonstrating enhanced corrosion resistance. Liu et al. [13-14] developed an accelerated corrosion test environmental spectrum based on equivalent relationships, conducted accelerated corrosion experiments on aircraft aluminum alloys, and used SEM to observe corrosion damage, obtaining test results for pitting depth and density at different exposure times while proposing three predictive methods: data fitting, neural networks, and time series analysis. Mameng et al. [15] performed chloride immersion and electrochemical tests on austenitic stainless steel, showing that increasing chloride concentration and temperature progressively decrease pitting potential and increase current density, thereby accelerating pit propagation.

This paper builds upon existing metal corrosion theories and employs non-equilibrium statistical methods to derive the probability density distribution function for pitting, along with perforation probability and reliability, providing a theoretical basis for metal component usage and preventing catastrophic losses from corrosion failure. Additionally, we present algorithms for calculating corrosion depth, corrosion rate, and pitting probability density distribution functions for aircraft aluminum alloy LD2 structural material, compute perforation probabilities at different corrosion ages, and analyze material reliability. This approach organically combines microscopic pitting mechanisms with macroscopic material properties, demonstrating the rationality of this theory for analyzing metal corrosion behavior.

1 Pitting Growth Model for Metal Materials

The presence of condensed water, even in trace amounts, causes metal corrosion to proceed via electrochemical processes accompanied by corrosion currents. The metal' s environment can be treated as a large impedance, allowing the corrosion process to be analogized to the transient response of an LR circuit to derive the relationship between corrosion current and time. For the discharge process in an LR circuit, the instantaneous current i satisfies [16]

$$i = i_0 e^{-\frac{Rt}{L}}$$

where E is the electromotive force, R is the resistance, L is the inductance, i_0 is the initial maximum current, and $\tau = L/R$ is the LR circuit time constant. A larger L results in a larger time constant and slower current attenuation. For the metal corrosion process, which is an extremely slow electrochemical process, we introduce an attenuation constant λ, where λ is an environment-dependent constant. When the time constant τ is large, the corresponding attenuation constant λ is small, indicating slow decay of the corrosion current.

The pitting process in metal materials can be divided into two stages: pit nucleation (metastable pitting) and pit growth (stable pitting) [17-19]. Let the incubation period from pit initiation to nucleation be dt, which can last for months or even years and shortens with increasing ambient temperature [20]. After the incubation period, pit growth begins, with time t corresponding to the time variable in the LR circuit. The maximum corrosion current density i_a corresponds to the initial maximum current in the LR circuit, yielding the time-dependent relationship for metal corrosion current density:

$$i(t) = i_a e^{-\lambda t}$$

The pitting process is a stochastic process [21]. Brownian motion theory for atomic diffusion in crystals shares similarities with the microscopic mechanism of pit growth, representing a combination of diffusion and hindrance processes where the system' s resistance to charge movement increases with time. Therefore, we define the activation energy for atomic diffusion as

$$\varepsilon(t) = \varepsilon_0 + \alpha t$$

where ε_0 is the potential barrier between interstitial sites, α is a constant related to the pitting environment, and t is the atomic diffusion time. The jump rate of atoms between lattice interstitials via thermal fluctuations is [22]

$$\nu = \nu_0 e^{-\frac{\varepsilon}{k_B T}}$$

where ν_0 is the atomic vibration frequency, k_B is the Boltzmann constant, and T is temperature, with ν representing the number of jumps per second for interstitial atoms.

Assuming a lattice spacing d, charge carrier density n, and charge q per carrier, the material corrosion current density can be derived as

$$i = nq\nu d$$

Comparing equations (2) and (3) reveals

$$i_a = nqd\nu_0 e^{-\frac{\varepsilon}{k_B T}}$$

Equation (4) shows that the maximum corrosion current density i_a is proportional to the charge carrier concentration n, while the current attenuation constant λ depends on both the environmental hindrance to charge movement and temperature T. Elevated temperatures reduce λ and increase corrosion current density because higher temperatures destabilize the passive film on metal surfaces, decreasing material impedance.

From Faraday' s law, the metal pitting growth rate formula is derived [23-25] as

$$v = \frac{M}{\rho ZF} i$$

where a is pitting depth, M is the atomic weight of the corroding metal, Z is the metal ion valence, ρ is metal density, and F is Faraday' s constant. Integrating equation (5) yields the time evolution of pitting depth:

$$a(t) = \frac{Mi_a}{\rho ZF\lambda} \left(1 - e^{-\lambda t}\right)$$

As $t \to \infty$, substituting into equation (6) gives the formula for maximum pitting depth a_m:

$$a_m = \frac{Mi_a}{\rho ZF\lambda}$$

2 Non-Equilibrium Statistical Theory of Metal Pitting Process

Under environmental action, pits on metal surfaces continue to grow. Due to inhomogeneities in alloy composition, surface condition, heat treatment temperature, ambient temperature, and pH value [26], the corrosion process can be viewed as uniform corrosion superimposed with random fluctuations. While uniform corrosion is deterministic, these inhomogeneity fluctuations are stochastic, allowing the pitting growth process to be treated as a non-equilibrium statistical process. The pitting growth rate should follow the generalized Langevin equation [27]

$$\frac{da}{dt} = K(a,t) + \beta(a)\xi(t)$$

where $K(a,t)$ is the drift growth rate determined by material type and environment, $\xi(t)$ is the fluctuation function related to environmental and material inhomogeneity fluctuations, $\beta(a)$ is the fluctuation amplification function.

Since pitting growth can be approximated as a Markov process [28] and the fluctuation function satisfies a Gaussian distribution

$$\langle \xi(t) \rangle = 0, \quad \langle \xi(t)\xi(t') \rangle = Q\delta(t - t')$$

where Q is the fluctuation coefficient and δ is the Dirac delta function.

According to stochastic theory, the equivalent Fokker-Planck equation for equations (8) and (9) is [27]

$$\frac{\partial P(a,t)}{\partial t} = -\frac{\partial}{\partial a}[K(a,t)P(a,t)] + Q\frac{\partial^2 P(a,t)}{\partial a^2}$$

This differential equation describes the stochastic pitting growth process, where $P(a,t)da$ represents the probability of pit depth being between a and $a+da$ at time t. The initial and boundary conditions are clearly

$$P(a,0) = \delta(a-a_0), \quad P(\infty,t) = 0$$

The growth rate given by equation (5) after the incubation period actually represents the drift growth rate $K(a,t)$. Combining equations (6) and (11) yields

$$K(a,t) = \lambda(a_m - a)$$

From references [27, 29], for a pit with initial depth a_0 at time t, where τ is the pitting life of the metal material and η is the sum of relative deviations of the four physical quantities M, i_a, Z, and ρ, i.e.,

$$\eta = \frac{\Delta M}{M} + \frac{\Delta i_a}{i_a} + \frac{\Delta Z}{Z} + \frac{\Delta \rho}{\rho}$$

Substituting equations (12)-(14) into the Fokker-Planck equation yields the pitting probability density distribution function

$$P(a,t) = \frac{1}{\sqrt{4\pi Qt}} \exp\left[-\frac{(a-a(t))^2}{4Qt}\right]$$

From equation (15), the probability density distribution function shows that $P(a,t)da$ represents the probability of a pit with depth a existing in the interval $[a, a+da]$ at time t. Clearly, the initial and boundary conditions are $P(a,0) = \delta(a-a_0)$ and $P(a,\tau) = 0$, respectively, with the physical meaning that any pit of depth a cannot exist for infinite time, i.e., the pitting life of metal materials is always finite.

3 Perforation Probability and Reliability

Typically, materials contain numerous pits rather than a single one. Let S be the material area and N the pit density per unit area, then $M = SN$ represents the total number of pits in the material. From equation (6), pitting depth a increases with time t, and when it reaches the material thickness, perforation failure occurs. According to the weakest link model for pitting perforation, the probability of material perforation within time $0-t$ is

$$P_f(t) = 1 - \exp[-MP(a_m, t)]$$

where a_m is the maximum pitting depth, taken as the material thickness. The perforation probability $P_f(t)$ satisfies $P_f(0) = 0$ and $\lim_{t\to\infty} P_f(t) = 1$, meaning that at $t = 0$ perforation is impossible, while at infinite time perforation is inevitable, demonstrating that all materials have a finite pitting life.

Using equation (16) to solve the approximation of equation (17) yields [30]

$$P(a_m, t) \approx \theta t$$

where θ is a constant. Substituting equation (18) into equation (17) gives the approximate expression for metal pitting perforation probability $P_f(t)$:

$$P_f(t) \approx 1 - \exp(-M\theta t)$$

From equation (19), the probability that the material continues to operate safely without pitting perforation under environmental action within time $0 - t$, i.e., the material reliability, is

$$R(t) = \exp(-M\theta t)$$

Clearly, equation (20) satisfies $R(0) = 1$ and $R(\tau) \to 0$.

4.1 Calculation of Pitting Depth and Rate

For aircraft aluminum alloy LD2 material, the following parameter values are selected [31]: $M = 27.18 \times 10^{-3}$ kg/mol, $Z = 3$, $\rho = 2.7 \times 10^3$ kg/m³, and $F = 96500$ C/mol. Additionally, based on experimental results from reference [13], the incubation period is assumed to be $\tau = 3.1536 \times 10^7$ s (2 yr). Using equations (4)-(6), the pitting rate and depth are simulated with a corrosion current density $i_a = 0.009$ A/m² and corrosion current attenuation constant $\lambda = 0.18$/yr at temperature $T = 40°$C. The resulting time evolution curves for pitting depth and rate are shown in Figures 1 [Figure 1: see original paper] and 2 [Figure 2: see original paper], respectively.

Figure 1 Time evolution curves of pitting depth: (a) observed and estimated pitting depth at 40°C, (b) pitting depth at different temperatures.

Figure 1 shows that pitting depth a increases with corrosion time t. When $t = 5/\lambda$, the pitting depth $a \approx 0.99a_m$, meaning that after $5/\lambda$ time, the pitting depth essentially approaches the maximum pitting depth a_m. Perforation occurs when material thickness is less than pitting depth. At a given corrosion time, pitting depth a increases with ambient temperature T because elevated temperatures lower pitting potential, increase corrosion current density I, and reduce passive film impedance.

Figure 2 [Figure 2: see original paper] Time evolution curves of pitting rate: (a) observed and estimated pitting rate at 40°C, (b) pitting rate at different temperatures.

Figure 2 demonstrates that pitting rate v decreases with corrosion time t, primarily because increasing pitting depth a enhances resistance to corrosive medium penetration at the aluminum alloy surface, while dissolved corrosion products accumulate at pit openings, continuously reducing the corrosion rate. Since corrosion current density I increases with temperature T, Figure 2(b) shows that pitting rate v also increases with temperature. Due to fluctuations caused by temperature, pH, and metal surface conditions, experimental data exhibit some scatter, but theoretical curves fit the experimental data reasonably well, providing a basis for subsequent probability density distribution calculations.

4.2 Calculation of Pitting Probability Density Distribution Function

Based on the previous simulations of pitting depth and rate, when $t = 20$ yr, the metal material' s pitting depth becomes essentially constant and the pitting rate approaches zero. Therefore, the material' s pitting life is assumed to be $\tau = 20$ yr. Using equations (15) and (16) with $\eta = 0.2$, the pitting probability density distribution function curves are plotted in Figures 3 [Figure 3: see original paper] and 4 [Figure 4: see original paper].

Figure 3 shows that the spatial probability density distribution function exhibits a bell-shaped distribution with a large middle region and small tails. At a given time t, the maximum probability density value corresponds to the most probable pitting depth, allowing prediction of the most likely pitting depth range at different times. For example, at $t = 4$ yr, pitting depth a most likely grows to the 45-55 m range. As pitting time increases, the curve peak shifts rightward, indicating continuous pit growth over time, while the curve becomes narrower, showing that most pits are more concentrated near the most probable value.

Figure 3 Probability density function of pitting depth distribution for several exposure times.

Figure 4 illustrates the temporal probability density distribution function for different pitting depths a, where the maximum value indicates the most probable pitting time, enabling prediction of the most likely time range for different depths. For instance, pits with depth $a = 50$ m most likely appear between 5-6 yr. As pitting depth increases, the temporal probability density distribution curve shifts rightward, indicating that deeper pits require longer times. It can be predicted that growing pitting depth a to 70 m requires approximately 11-12 yr.

Figure 4 [Figure 4: see original paper] Probability density function of exposure time distribution at different pitting depths.

4.3 Calculation of Perforation Probability and Reliability

From equation (7) and the previous pitting depth simulations, the maximum pitting depth a_m is obtained. Substituting the material' s pitting area and density [14] into equations (17) and (20) yields the alloy's perforation probability P_f and reliability R, as shown in Table 1 .

Table 1 Failure probability and reliability at different pitting times

t /yr	S / mm²	N / mm ²	Pf	R
Note: t —exposure time, S —corroded area, N —pitting density, Pf —failure probability, R —reliability				

Table 1 shows that as pitting time increases, adjacent pits gradually coalesce, increasing individual pit area and causing a decreasing trend in pitting density. At small pitting times, material perforation probability is nearly zero and the material is essentially safe and reliable. As pitting time continues to increase, perforation probability gradually rises while reliability declines.

Assuming pitting density remains constant over time, equations (19) and (20) can be used to plot the perforation probability and reliability curves as functions of pitting time, as shown in Figure 5 [Figure 5: see original paper] (with $N = 67$ mm ² and $\theta = 0.051$).

Figure 5 Time evolution curves of failure probability and reliability: (a) failure probability and reliability at pitting density of 67 mm ², (b) reliability at different pitting densities.

Figure 5 shows that when pitting time t is less than 12 yr, the material remains essentially safe and reliable, with perforation probability P_f nearly zero. However, when pitting time approaches the limiting value, perforation probability reaches 100% while material reliability R drops to zero. At the same pitting time, higher pitting density leads to lower reliability and greater susceptibility to perforation. Therefore, material reliability can be improved and pitting resistance enhanced by reducing pitting density through methods such as material nanocrystallization.

Conclusions

(1) Based on existing metal corrosion theories, a pitting growth model for metals was established, deriving the time-dependent relationships for pitting depth and rate, and obtaining the formula for maximum pitting depth. This approach organically combines microscopic pitting mechanisms with macroscopic material properties, demonstrating that metal pitting resistance decreases with increasing temperature T.

(2) Using non-equilibrium statistical methods, the time- and depth-dependent probability density distribution function for metal pitting was derived by establishing and solving the Fokker-Planck equation. This enables calculation of the most probable pitting depth at different service times and prediction of the most likely time range for different pitting depths to occur.

(3) Based on the weakest link model for pitting perforation, the perforation probability and reliability of metal materials were calculated. Material reliability gradually decreases while perforation probability continuously increases with pitting time, allowing inference of the safe service period for metal materials. These results are expected to be applied to pitting life prediction and reliability analysis of metal materials.

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Note: Figure translations are in progress. See original paper for figures.