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Crack Initiation Life of Materials Under Combined Pitting Corrosion and Cyclic Loading

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Pitting corrosion triggered damage is responsible for the degradation of many metallic materials affecting structural integrity. As pitting and crack initiation processes govern the overall life of such structures and components, particularly at nominal cyclic stresses, there is a need to develop simple models to estimate crack initiation life of materials. This paper presents a simple deterministic model that considers the effect of cyclic stressing under pitting corrosion conditions. The developed model is validated on an aluminum alloy 2024-T3, and 12% Cr stainless steel used in aircraft and steam turbines, respectively. The predicted critical pit depth values are in fair agreement with the limited experimental data available in the literature. The model indicates that at high stresses, the crack initiation can occur very rapidly even from relatively small pits. The crack initiation life predictions when compared with the available experimental data, suggest a probable stress-level dependency with regard to the form and extent of the influence of cyclic stresses on pit growth and subsequent crack formation.

Keywords aluminum, cyclic stresses, model, pitting corrosion, steel

1. Introduction

The combination of pitting corrosion and fatigue in most metallic materials can be potentially disastrous, which when left unchecked, can lead to catastrophic failures (Ref 1-10). Be it in aircraft structures (Ref 1-8) or in steam turbine blades (Ref 9, 10), for instance, pitting corrosion assisted by fatiguing conditions is recognized to be one of the most significant degradation mechanisms. Materials such as high-strength aluminum alloys and stainless steel that otherwise have good general corrosion resistance exhibit strong propensity to localized pitting corrosion (Ref 11-13). The degradation in these materials on exposure to chloride-containing environment leads to the formation and growth of pits and the eventual nucleation and propagation of cracks from these sites. Although pits can initiate from both physical and chemical heterogeneities on the surface, the role of impurities and constituent particles in inducing pitting corrosion has been more commonly observed and studied. In aluminum alloys, usually the intermetallics including the precipitate particles have been observed to induce corrosion (Ref 14, 15), whereas in stainless steels it is usually the sulfides of Mn (or of Mn and Cr) that are responsible for the same (Ref 16). Recent work has however shown that although pitting corrosion may initiate from sulfides, the aluminum oxide inclusions are the ones which actually promote the formation of pits (Ref 16). Since these materials contain numerous such particles, electrochemical reactions are triggered between them and the surrounding matrix (Ref 14-16). The pit growth is seen to be controlled by the limiting cathodic current density supported by the exposed constituent/impurity particles within a growing pit (Ref 4, 15). Such particle-induced pitting corrosion described above has indeed been observed by scanning and transmission electron microscopic techniques (Ref 4, 5, 9). Although the pits are seen to take different shapes (Ref 17), corrosion-fatigue cracks are more usually found to nucleate from the deepest grown pits (Ref 6).

Recent experimental investigations (Ref 18, 19) have shown fatigue stressing in a corrosive environment influencing the pit growth process in Al-alloys. Likewise, other investigators (Ref 9, 16) have also observed that the severity of pitting in stainless steel including the number and the size of corrosion pits is strongly influenced by cyclic loading. Apparently, the pitting potential decreases, and the current densities for pitting corrosion increase with the application of cyclic stress (Ref 20). The reason for fatigue loading affecting pitting is attributed to not only its ability to facilitate the breakdown of the passive oxide film that is present on such material surfaces but in also preventing repassivation of the pits (Ref 9, 16).

With pitting corrosion degradation being recognized as a potential cause for structural failures, the need for predictive methodologies and models cannot be overstated. As most of the component's life, particularly at nominally stressed environments, could be attributed to corrosion pit growth and crack initiation (Ref 9), it becomes extremely important to be able to predict this stage of the damage process. While existing models from the literature are briefly discussed in the following section, the paper presents a simple integrated deterministic model for crack initiation life prediction in pitting corrosion-prone materials subject to fatigue loading. The model has been validated on two alloys viz. precipitation-hardenable high-strength aluminum alloy 2024-T3 commonly used in aircraft fuselage splices, and tempered 12% Cr martensitic stainless steel used as steam turbine blade material.

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2. Review of Existing Damage Models in High-Strength Al-Alloys and Stainless Steel

This section briefly reviews existing models on aluminum alloys and stainless steel pertaining to crack initiation under the influence of pitting corrosion and cyclic stressing.

In order to quantify pitting corrosion-fatigue damage in Al-alloys, Hoeppner et al. (Ref 21) came up with a mechanistic model to predict the critical pit size, from which a corrosion-fatigue crack nucleates and grows. Their model along with other mechanistic models of Lindley et al. and Kawai and Kasai referenced by them (Ref 21) mostly try to address the pit growth stage and the ensuing crack initiation in Al-alloys. However, the effect of the electrochemical reactions has not been clearly brought out in their formulations nor any influence of fatigue stressing considered during pitting. Wei and Harlow's group proposed probabilistic models for the growth of corrosion pits induced by constituent particles in Al-alloys in order to predict the time required for pit growth and potential fatigue crack nucleation (Ref 7, 22). Rokhlin et al. (Ref 3) developed a mechanistic-deterministic model for fatigue crack initiation and propagation from single artificial and actual pits in Al-alloys, using a fracture mechanics approach. In this case, the pit nucleation and growth was not considered. Engelhardt and Macdonald (Ref 23) tried to model pitting events and pit growth in stainless steel on the basis of a deterministic-statistical approach but did not estimate the corrosion-fatigue crack initiation life.

Although recent experimental observations have shown pitting to be affected by cyclic loading (Ref 9, 16, 18-20), with fatigue stressing in a corrosive environment even influencing the pit growth process (Ref 18, 19), models pertaining to Al-alloys is situations involving simultaneous corrosion and fatigue have however not considered any synergy between them. In the models on Al-alloys developed by Wei and Harlow's group (Ref 7, 24), again based on mechanistic-probabilistic approach, the pit nucleation stage is ignored while the pit growth taken to be solely dictated by corrosion. The probabilistic model of Al-alloys proposed by Shi and Mahadevan (Ref 8) considers pitting to be entirely controlled by corrosion, with fatigue merely facilitating the nucleation of cracks from the pits once they are formed. Rajasankar and Iyer (Ref 12), even while acknowledging the influence of fatigue on pit growth, consider cyclic stresses to merely alter the shape of pits, in their probabilistic model pertaining to Al-alloys. The pit nucleation is also taken to be independent of the applied cyclic load, both in magnitude and frequency. Ishihara et al. (Ref 19) used a probabilistic-deterministic approach to explain their experimental observations in Al-alloys and came up with the form for the stress amplitude to describe its effect on pit growth/crack initiation, but failed to include any corrosion parameters in their pit growth formulation. This form for the stress amplitude (Ref 19), however, appeared to depend on the operating stress level itself (Ref 25).

Based on the above discussion, there seems to be no single model (deterministic or probabilistic) that addresses the pit growth and crack initiation life predictions that go into the complex pitting corrosion-fatigue process, under conditions of their coexistence. Therefore, as mentioned earlier, a simple deterministic model that attempts to address this issue based on an integrated approach is presented here.

3. Model Development

The proposed model is based on a simple deterministic approach that considers the scenario of simultaneous presence of a corrosive environmental condition and cyclic stressing. Based on the influence of fatigue stress on the pitting process itself, as observed by some investigators (Ref 18, 19), the model incorporates a stress function into the formulation originally proposed by Kondo (Ref 2) and Wei (Ref 7). The corrosion pit growth and the ensuing crack initiation are therefore dependent on both the anodic current that drives the electrochemical reactions as well as the cyclic stress amplitude. It is important to emphasize here that our work does not attempt to simulate any actual environmental or loading condition of real components or structures but only predicts the crack initiation lives of the associated materials under pitting corrosion and cyclic loading.

The premise of the model and the assumptions made are as follows:

1. The alloy materials are exposed to a chloride ion containing aqueous environment and cyclic stressing involving complete load reversal.
2. The constituent/impurity particles responsible for pitting corrosion in the materials are clustered.
3. The crack initiation process takes place under the combined influence of corrosion and fatigue loading and is preceded by pit nucleation and growth, attainment of critical pit depth, and the latter's transitioning into a crack.
4. Conditions favoring pitting corrosion and fatigue damage are present right from the start and cyclic loading influences the process from the pit nucleation phase itself.
5. Pits that are nucleated at such particles are stabilized almost instantaneously and their growth is controlled both by the pitting current and the stress amplitude.
6. The pits are of hemispherical shape right from nucleation and during the entire period of their growth.
7. The model is based on the "single dominant flaw approach" in that although several pits may nucleate and grow (individually or in combination), there is only one pit that attains critical depth, exceeding the threshold level for crack formation, and thereby initiating a crack.
8. The crack that is responsible for material damage is "instantly" formed only at the pit that has reached critical depth, and the crack initiation time is, in effect, the time for the pit to grow to this stage.

Now, the depth a_p of a corrosion pit in a material depends not only on the corrosive environment but also on its microstructure (Ref 7). Based on Faraday's law, a_p, at any given time t is given as (Ref 2, 7):

$$a_p = \left(\frac{3MI_p}{2\pi nF\rho}\right)^{1/3} t^{1/3} \quad \text{(Eq 1)}$$

where M is the atomic mass of the corroding material, ρ its density, n the number of electrons released during its corrosion, F the Faraday's constant, and I_p the pitting current, which in turn is given by (Ref 7):

$$I_p = I_{p0} \exp\left(\frac{-\Delta H}{RT}\right) \quad \text{(Eq 1a)}$$

where I_{p0} is the pitting current coefficient (a random variable following a normal distribution (Ref 26)), ΔH the activation energy, R the universal gas constant, and T the absolute temperature.

Now that pit initiation and growth are also known to be governed by the fatigue stress (Ref 18, 19), Eq 1 needs to be suitably modified. Based on Ishihara et al.'s (Ref 19) finding that the influence of stress takes the form C^{σ_a}, a stress-dependent function of this form is incorporated into Eq 1, which can now be written as:

$$a_p = \left(\frac{3M}{2\pi nF\rho}\right)^{1/3}\left(I_p^{1/3}\right)\left(C^{\sigma_a}\right)\left(t^{1/3}\right) \quad \text{(Eq 2)}$$

In other words,

$$t = \frac{2\pi nF\rho}{3M}\left(a_p^3\right)\left(\frac{1}{I_p}\right)\left(\frac{1}{C^{\sigma_a}}\right)^3 \quad \text{(Eq 3)}$$

Substituting for t as $t = N/f$, in Eq 3 where N is the number of stress cycles and f the frequency, and rewriting it, we get:

$$N = \frac{2\pi nF\rho}{3M}(f)\left(a_p^3\right)\left(\frac{1}{I_p}\right)\left(\frac{1}{C^{\sigma_a}}\right)^3 \quad \text{(Eq 4)}$$

The above equation gives the number of cycles N that are required for a pit on the material surface to reach a particular depth a_p under conditions of simultaneous corrosion and fatigue.

Now, considering a component surface containing hemispherical pits to be similar to that of an infinite plate consisting of semicircular surface flaws in 2-D, the expression for the stress intensity factor range ΔK for a pitted surface can be written as (Ref 14):

$$\Delta K = \left(\frac{2.2}{\pi}\right)K_t \Delta\sigma\sqrt{\pi a_p} \quad \text{(Eq 5)}$$

where $\Delta\sigma(= 2\sigma_a)$ is the stress range and K_t the stress concentration factor resulting from a circular rivet hole (Ref 24). Since the pit shape is assumed to be hemispherical throughout its growth, the stress concentration factor is essentially a constant.

The pit is considered to have reached a critical depth a_{pc} and ready to "transition" into a small crack the moment it satisfies the threshold requirement for crack initiation, according to linear elastic fracture mechanics (Ref 8, 26). What this essentially means is that the kinetics of small crack extension exceeds the pit growth rate at this juncture, thus resulting in the crack taking over the damage mechanism. In other words, the pit is said to have attained the critical depth when ΔK for the pit equals ΔK_{th} the threshold stress intensity factor range for crack initiation (Ref 8, 26).

Thus, a_{pc} can be written as (Ref 14):

$$a_{pc} = \pi\left(\frac{\Delta K_{th}}{4.4K_t\sigma_a}\right)^2 \quad \text{(Eq 6)}$$

Replacing a_p with a_{pc} in Eq 4, and substituting Eq 6 into Eq 4, the number of cycles to crack initiation N_i is thus given by:

$$N_i = \frac{2\pi nF\rho}{3M}(f)\left[\pi\left(\frac{\Delta K_{th}}{4.4K_t\sigma_a}\right)^2\right]^3\left(\frac{1}{I_p}\right)\left(\frac{1}{C^{\sigma_a}}\right)^3 \quad \text{(Eq 7)}$$

From the above equation, it is easily seen that the crack initiation cycle is not only inversely proportional to the stress amplitude but also to the pitting current. The pitting current represents the galvanic current flowing between the anodic aluminum/iron matrix and the cathodic particle/particle clusters as a result of the electrochemical reactions. For the sake of convenience, the last term in Eq 7 will henceforth be referred to as the "stress factor", and C the stress factor parameter.

4. Results and Discussion

In order to make predictions using the developed integrated model, the individual systems of aluminum alloy 2024-T3 and tempered 12% Cr stainless steel, both exposed to 3.5% NaCl solution and cyclic stressing under complete load reversal, have been considered. The input parameters for the corresponding (or similar) materials were taken from the literature (Table 1 and 2). Substituting for the parameter values into Eq 6 and 7, the critical pit depth estimates and the predicted number of cycles to crack initiation for chosen values of the "stress factor" parameter are determined for each of the materials. The value of C had to be clearly greater than 1.0 to have a positive effect on pit growth and eventual crack initiation.

Table 1 Input data for corrosion-fatigue crack initiation life predictions in aluminum alloy 2024-T3

Parameter	Value
Atomic mass, M	27×10^{-3} kg/mol
Valence, n	3
Faraday's constant, F	96485 C/mol
Density, ρ	2700 kg/m^3
Threshold stress intensity range, ΔK_{th} (Ref 5)	2.32 MPa√m
Stress concentration factor, K_t (Ref 26)	2.8
Pitting current coefficient, I_{p0} (Ref 26)	3.52×10^{-2} C/s
Enthalpy, ΔH (Ref 26)	40×10^3 J/mol
Universal gas constant, R	8.314 J/mol K
Temperature, T	293 K
Frequency, f	10 Hz

Table 2 Input data for corrosion-fatigue crack initiation life predictions in tempered 12% Cr stainless steel

Parameter	Value
Atomic mass, M	56×10^{-3} kg/mol
Valence, n	2
Faraday's constant, F	96485 C/mol
Density, ρ	7780 kg/m^3
Threshold stress intensity range, ΔK_{th} (Ref 10)	8.14 MPa√m
Stress concentration factor, K_t (Ref 26)	2.8
Pitting current coefficient, I_{p0} (Ref 30)	1×10^{-7} C/s
Enthalpy, ΔH (Ref 31)	15.5×10^3 J/mol
Universal gas constant, R	8.314 J/mol K
Temperature, T	293 K
Frequency, f	60 Hz

4.1 Aluminum Alloy 2024-T3

Figure 1 shows the variation of the predicted critical pit depth values in 2024-T3 as a function of stress amplitude. Being inversely proportional to the stress amplitude, the critical pit depth decreases with increasing stress levels and vice versa. The results point to how crack initiation from pit sites can be extremely fast at high stress levels and can occur even from relatively small pits. What this implies is that even when the stress levels are high, pitting does take place but the pit grows only to a relatively small depth before initiating a crack. This is not surprising, given the fact that pits like cracks generally tend to nucleate from defects and second phase particles/impurities on material surfaces. For comparison, two data points available from Ishihara et al.'s experimental work (Ref 19) on the alloy under simultaneous exposure to 3.5% NaCl and cyclic loading are also included in Fig. 2. It is seen that these data points fall in line with our predictions.

The predictions made from our model with respect to the crack initiation lives in 2024-T3 for different stress amplitudes are shown in Fig. 2. The data points available from Ishihara et al. (Ref 19) are also shown for comparison. Assuming that the data points are truly representative, it is interesting to note that no single prediction curve seems to fully fit into the trend of the experimental results. The data points actually fall within the curves corresponding to $C = 1.001$ to $C = 1.02$. This reinstates the idea that the influence of the cyclic stress (as reflected in the form and magnitude of the stress factor) on pitting and crack initiation need not necessarily be defined the same way over all the stress regimes. Looking at the profiles of the prediction curves, this is quite obvious in that the stress factor becomes increasingly sensitive with increasing stress levels.

As pointed out in Eq 7, the crack initiation is a function of the pitting current and is inversely related to it. In order to determine to what extent the latter influences crack initiation life, a plot between the predicted number of cycles to crack initiation and the stress amplitude is made for two arbitrary values of the pitting current coefficient, as shown in Fig. 3. As expected, an increase in the pitting current pushes the curve to the left viz. lower crack initiation lives for a given stress amplitude. A thousand fold increase in the pitting current is seen to reduce the crack initiation time by nearly as much.

4.2 Tempered 12% Cr Stainless Steel

Figure 4 shows the predicted critical pit depth variation in tempered 12% Cr stainless steel as a function of stress amplitude. It is not surprising that the profile is similar to what was noticed for 2024-T3 (Fig. 1). The prediction seems to

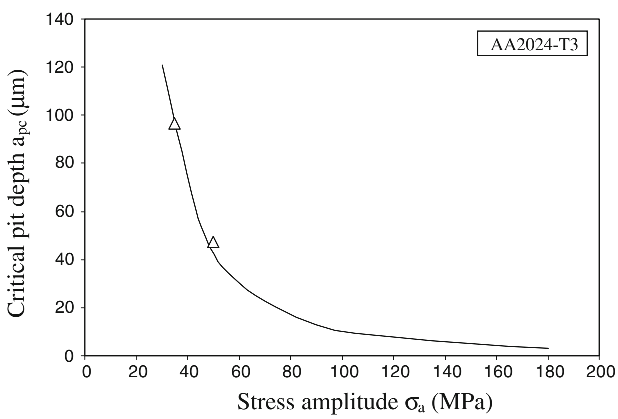

Fig. 1 The variation in predicted critical pit depth as a function of stress amplitude under simultaneous corrosion and cyclic loading conditions in Al-alloy 2024-T3. The open triangles correspond to the values of Ishihara et al. (Ref 19)

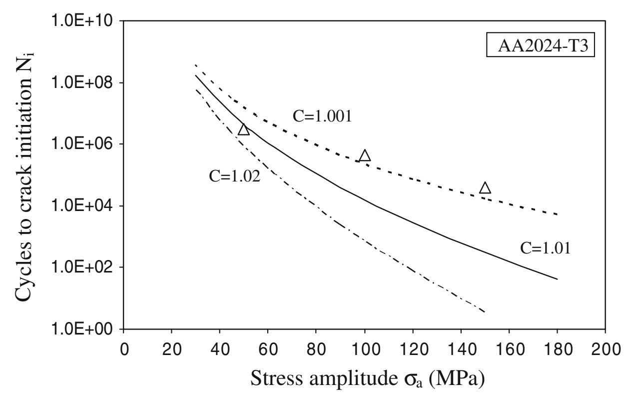

Fig. 2 Predicted number of cycles to pitting corrosion crack initiation as a function of stress amplitude in Al-alloy 2024-T3 for three different values of the "stress factor". The open rectangles correspond to the values of Ishihara et al. (Ref 19)

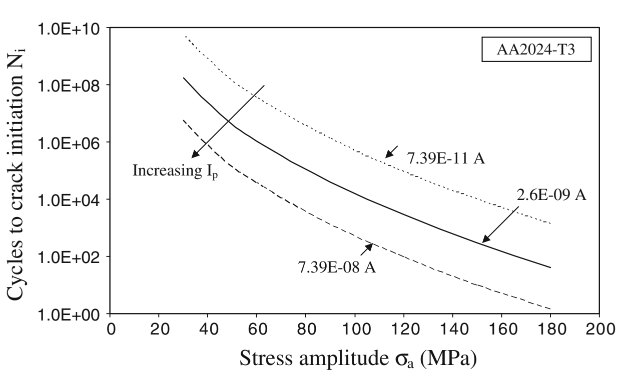

Fig. 3 Plot showing the effect of pitting current on the stress amplitude vs. predicted number of cycles to pitting corrosion crack initiation behavior in Al-alloy 2024-T3 (for $C = 1.01$)

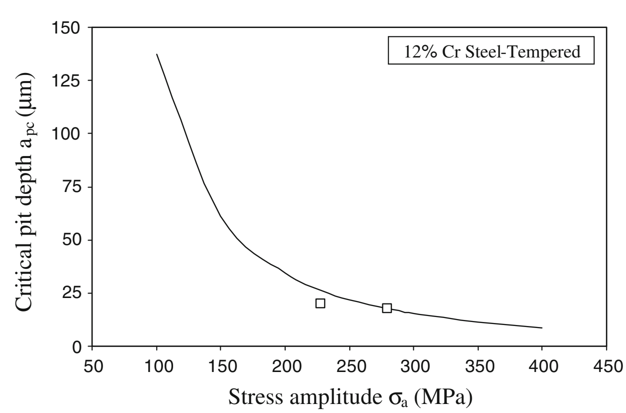

Fig. 4 The variation in predicted critical pit depth as a function of stress amplitude under simultaneous corrosion and cyclic loading conditions in tempered 12% Cr stainless steel. The open rectangles correspond to the experimental values from Ebara (Ref 9)

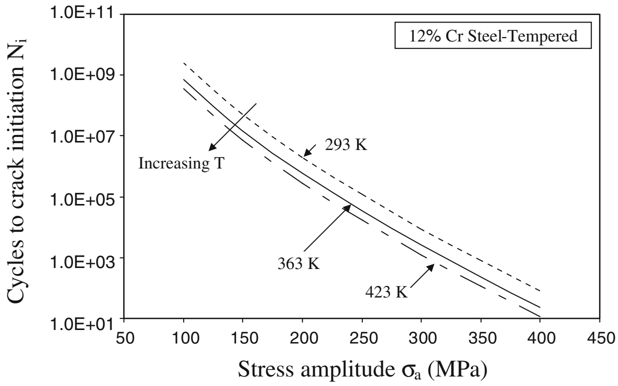

Fig. 5 Plot showing the effect of temperature on the stress amplitude vs. predicted number of cycles to pitting corrosion crack initiation behavior in tempered 12% Cr stainless steel (for $C = 1.01$)

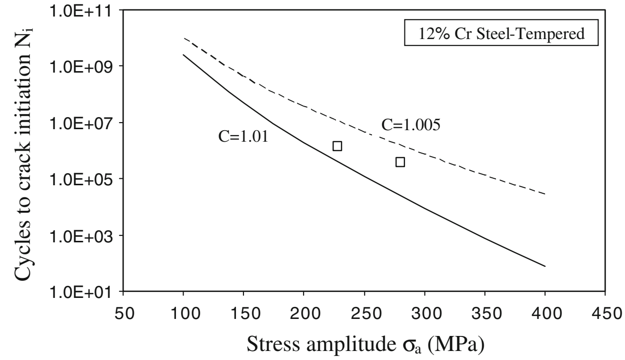

Fig. 6 Predicted number of cycles to pitting corrosion crack initiation as a function of stress amplitude in tempered 12% Cr stainless steel for two different values of the "stress factor". The open rectangles correspond to the experimental data points from Ebara (Ref 9)

agree well with the two data points of Ebara (Ref 9) that were the only available experimental result in the literature. The results are for the same alloy tested under simultaneous cyclic loading and exposure to 3.5% NaCl. It must be mentioned though that these points are said to actually correspond to a test temperature of 318 K. However, with the effect seeming quite nominal for this temperature difference (Fig. 5), the data can be taken as valid for comparison. Nevertheless, it needs to be mentioned that increasing temperatures accelerate the corrosion damage (according to Eq 1a) as has been experimentally observed by Cho et al. (Ref 27) as well.

The model predictions of the crack initiation lives with respect to tempered 12% Cr stainless steel for different stress amplitudes can be seen in Fig. 6. Again, with very little experimental results on tempered 12% Cr stainless steel available in the literature, the predictions are compared with the values deduced from Ebara's work (Ref 9). The data seem to fall within the prediction curves corresponding to $C = 1.005$ and $C = 1.01$ (Fig. 6). With the trends being similar to 2024-T3 (increasing sensitivity of the stress factor with stress amplitude), the same argument that there are probably different stress factor parameters at different regimes can be construed to hold good here as well.

Thus, the model is seen to make a fair prediction of the crack initiation cycles when compared with the values from literature however limited, in the presence of corrosion and cyclic stressing conditions. The stress factor not only shows a stress-level dependency but also has varying influence on the two alloy systems. While the stress factor parameter can probably take values between 1.001 and 1.02 to make a good prediction of the crack initiation phenomenon in 2024-T3 at different fatigue stress regimes, the range for C in the case of 12% Cr steel seems to be between 1.005 and 1.01. It is quite apparent therefore that the formulations for different alloy systems may have to address this issue depending on the operating stress. Given the fact that cyclic stressing in a corrosive environment has been observed to influence pitting (Ref 18, 19) and induce early micro cracking (Ref 28), a better knowledge of its precise role and influence can help determine the appropriate form and magnitude of the stress factor.

Furthermore, it is recognized that there are some limitations to the model. The assumption of "instant" pit initiation and stabilization in our formulation may not be truly valid and this could also have its influence on the predicted crack initiation life. Again, a hemispherical pit may not be quite representative of reality (Ref 28, 29). Moreover, according to the present formulation, the number of cycles to crack initiation is directly proportional to the frequency suggesting that any frequency increase or decrease necessarily amounts to a change in the crack initiation life however small or large. It would nevertheless be interesting to see the effect of frequency as pitting corrosion is believed to be influenced by cyclic loading frequency (Ref 12, 16). Finally, being a deterministic approach, our model suffers from other limitations by virtue of the fact that pitting corrosion (and even fatigue) is stochastic in nature.

5. Conclusions

A simple deterministic model for predicting crack initiation lives in metallic materials has been developed and tested on alloys such as aluminum alloy 2024-T3 and tempered 12% Cr stainless steel used in aircraft and turbine applications, respectively. The model considers the coexistence of corrosive environment and fatigue loading conditions and takes into account the influence of cyclic stresses in the pitting corrosion process. The predictions of the critical pit depth are found to be in fair agreement with limited experimental data available in the literature. The results also indicate how crack initiation from pit sites can be extremely fast at high stress levels and can occur even from relatively small pits. As for the crack initiation predictions, there seems to be, again based on very limited experimental data, a stress-level dependency with regard to the form and magnitude of the effect of cyclic stressing on pit growth and crack initiation. This may call for some modifications in the formulations based on more experimental observations and deeper understanding of the phenomenon. Due to the random process of corrosion pit initiation, a stochastic-deterministic approach may be called for. Future work will attempt to address some of these issues.

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