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*Review Article*

# Modelling of type 304 stainless steel crevice corrosion propagation in chloride environments

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## Abstract

Modeling of Type 304 stainless steel crevice corrosion propagation in terms of penetration rate in freshwater, brackish and marine environments has been carried out. The crevice assembly used for this study comprised of coupon (SS-304), polytetrafluoroethylene (crevice former) and fasteners (titanium bolt, nut and washers) designed with crevice scaling factor of 8 and 40 crevice sites immersed in various chloride solution concentrations of 1.5, 3.0 and 4.5 w/w % simulating environmental conditions in full immersion test. The test coupons were subjected to the respective environments. The set up were allowed to stand for 60 days with a set withdrawn 15 day-intervals to measure the depth of attack, y(t) in the crevice. The model equation developed could be used for the estimation of SS-304 crevice propagation trend in freshwater, brackish and marine environments as it showed good correlation with the experimental data, provided all other factors are constant.

**Keywords:** 304 stainless steel, crevice corrosion propagation, chloride environments modelling

## 1. Introduction

Crevice corrosion of SS-304 is an important damaging phenomenon in the chemical, petrochemical, nuclear and diverse industrial installations. It becomes unavoidable when such assemblies and installations operate in chloride environments (Atashin, Pakshir, & Yazdani, 2010) and creviced geometries (Engelhardt, & Macdonald, 2004). Such Chloride environments could be both natural and anthropogenic sources, such as the use of inorganic fertilizers, landfill leachates, septic tank effluents, animal feeds, industrial effluents, irrigation drainage, and seawater intrusion in coastal areas (Ijsseling, 2000).

Unexpected failures of creviced components and assemblies occur in service, which have economic, safety, health and environmental consequences and are not easily detected with traditional non-destructive techniques. These damages followed an unpredictable nature of initiation and a significant speed of propagation; they are mostly detected during shutdown inspection. The causes of the damages are not clearly identified and not similar to general corrosion, their penetration rates are not easily predicted.

Electrochemical techniques are commonly used for the study of crevice corrosion growth Penetration rates, in which crevice corrosion penetration rates, such as pit growth Penetration rates, is often described by a relationship between

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current density and applied potential (Pickering, 2003). However, there are several problems associated with this approach. First, the active area during crevice corrosion is typically unknown and difficult to measure (Ijsseling, 2000). Many assumptions regarding the number and morphology of crevice corrosion sites are needed in order to translate measured current transients into localized corrosion growth rate. This is complicated by the fact that localized corrosion may take more than one form, such as pits, crevices, and intergranular (IG) attack. Finally, localized corrosion in a real structure occurs under open circuit (OC) conditions.

Crevice corrosion of SS-304 in a range of chloride containing environments like freshwater, brackish and marine is a well-documented phenomenon with significant practical implications (Heppner, Evitts, & Postlethwaite, 2002; Kennell, Evitts, & Heppner, 2008; Postlethwaite, Evitts, & Watson, 19 95). SS-304 has wide applicability in various industries and can be used as good alternatives for other types of stainless steel because of its weld-ability and cost. However, penetration rates study of the metal-environment degradation behavior in non-electrochemical approach has not been popularly approached (Mon, Gordon, & Rebak, 2005) for the purpose of estimating the growth constant and parameters in chloride containing environments.

The development of effective crevice corrosion damage prediction technologies is essential for the successful avoidance of unscheduled downtime in industrial systems and for the successful implementation of life extension strategies (Engelhardt & Macdonald, 2004). Deterministic and statistical models have been both developed for better understanding of corrosion environments (Nicolas, Philippe, & Denise, 2013). Deterministic models are based on fundamental mathematical relations of processes, in which the effects are generated by the causes. Models from mathematical theories or mechanistic approaches are associated with protracted simulation times (Zhang, Ruan, Wolfe, & Frankel, 2003); however, this work is based on the practicable factors of geometry and environment that directly affects the crevice corrosion.

In this research, the regression analysis was used to develop an exponential model equation for the crevice corrosion penetration rate as a function of time. Also, a model equation (Y:t) was developed. A correlation analysis for the observed corrosion parameters was also carried out. It is envisaged that these parameter will be of uttermost importance in both maintenance of SS-304 creviced assemblies in chloride containing environments and in crevice assembly design involving stainless steel. An attempt was also made to compare the experimental results with the data generated using the developed model equations by plotting for proper comparison.

## 2. Material and Method

### 2.1 Material

The engineered crevice assembly designed for the purpose of this study consists of: Specimen (AISI 304) (coupons), Multiple Crevice Former; polytetrafluoroethylene (PTFE), Titanium Bolt, Nuts and Washers. The SS-304 sheets (3 mm thick), teflon sheets (5mm thick), titanium bolt, nuts and washers were all obtained at Owode Onirin international market, Lagos State of Nigeria.

### 2.2 Preparation of chloride containing environments

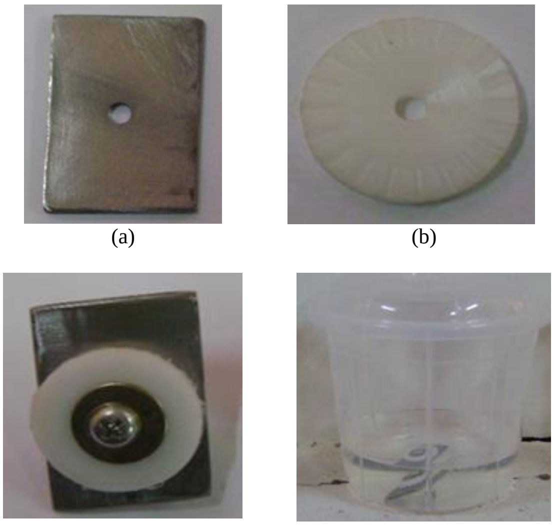
The test solution was NaCl solution prepared from analytical-reagent-grade reagent NaCl and double distilled water at: 1.5% w/w NaCl by weight to simulate the fresh water environment; 3%w/w NaCl by weight to simulate the brackish water environment and 4.5%w/w NaCl by weight to simulate the marine water environment.

### 2.3 Preparation of SS-304 crevice assembly

According to ASTM G-78, multi-crevice assemblies of crevice scaling factors of 8 and 40 crevice sites each were totally immersed in chloride containing environments to be monitored for the rate of propagation of crevice corrosion in terms of maximum depth of attack respectively at different immersion time intervals.

### 2.4 Set-up and experimentation

The test coupons (Figure 1) were divided into three groups of 4 test coupon each. The three group were exposed to the fresh-water, brackish and marine environments respectively. Each set was allowed to stand for 60 days with a set of coupon withdrawn every 15 days to measure the depth of crevice corrosion attack and morphological micrographs with reflective and inverted microscope respectively.



(a) (b)

(c) (d)

Figure 1. (a) Cut and polished corrosion test coupons of varied dimension (b) Photograph of fabricated crevice former (c) multiple crevice assembly for crevice corrosion test according to ASTM G78. (d) Total immersion of crevice former

## 3. Results and Discussion

The multiple regression analysis of rate of propagation of crevice corrosion of SS-304 in freshwater, brackish and marine environments characterized in terms of depth of creviced attack as a function of time was carried out. The crevice corrosion of SS-304 in various marine environments is confirmed to propagate without repassivation and the localized corrosion penetration relates to time and conform to equation (1) by regression:

$$y\,(t) = kt^{n} \tag{1}$$

where y is the Maximum depth of penetration, t is the immersion time, and k is a growth constant and n is the time exponent. For each of the simulated environment under study, the following equations were obtained after the pattern of Equation (1):

$$y\,(t) = 0.005t^{0.823} \tag{2}$$

$$y\,(t) = 0.021t^{0.865} \tag{3}$$

$$y\,(t) = 0.029t^{0.886} \tag{4}$$

From these equations, the k and n parameters are summarized as Table 1

Table 1. Parameters of the model

| Chloride Environments | k | n | $R^2$ |
|---|---|---|---|
| Freshwater | 0.005 | 0.823 | 0.971 |
| Brackish water | 0.021 | 0.865 | 0.917 |
| Marine | 0.029 | 0.886 | 0.941 |

The model and its parameters is similar to the Localized corrosion growth kinetics study by Alkire and Wong, (1988), Frankel (1998), Hunkeler and Böhni (1981), Mughabghab and Sullivan (1989), Szklarska-Smialowska (2005) where the propagation conform to a power equation in the same form as equation (1), where *k* and *n* are constants, typically in the range $0 < n < 1$. Using Equations (2), (3) and (4), modeled values were plotted with the experimental values to check the validity of the developed model equation. From Figure 2, it can be seen that the experimental values fitted well with the modeled values, hence giving a good insight into the SS-304 behavior in chloride containing environments like freshwater, brackish and marine environments being studied. The coefficient of correlation of each model shows high positive correlation. It is evident that 97.1%, 91.7% and 94.1% of propagation of crevice corrosion in freshwater, brackish and marine environments is caused by change in time.

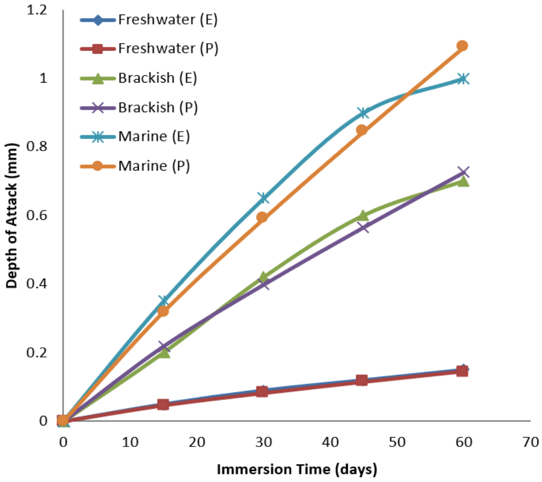


Figure 2. Crevice Corrosion penetration depth as a function of time for the experimental data (E) and observed model data (P) in freshwater, brackish and marine environments.

In all cases of environmental conditions considered, it is observed, as put in Table 1 that both k and n increase as the system behavior depart from freshwater condition to brackish and to marine conditions. This explains the fact that crevice corrosion becomes more severe as the chloride concentration increases. This further confirms the initial position due to the formation of corrosion products on the metal surface. If n is smaller than 0.5, the corrosion products show protective, passivating characteristics, otherwise n is greater than 0.5 (Engelhardt & Macdonald, 2004).

The growth constant confirms the properties of SS-304, particularly its susceptibility to anodic dissolution in the range of chloride concentrations, describing activities in freshwater, brackish and marine environments. The time exponent, n, was in the range of $0.5 \leq n < 1$. This confirms that among others processes the crevice corrosion process was diffusion-controlled (i.e., diffusion of metal ions out of the crevice) (Stewart, 1999; Matjaz, Franc, & Matja, 2012) subject to the ss-304 resistance of the chloride environments which occurs only through surface passivation phenomenon, provided all other metallurgical and environmental conditions are kept constant. Availability of reactants such as oxygen in an aerobic environmental condition and electrons as the time progresses in other words can hamper the modeled conditions and that could constitute the limits of the model. However, the proposed model overcomes inconveniences of the modeling crevice corrosion propagation and penetration rates by electrochemical methods.

The obtained function is differentiable, used to estimate the rates of crevice corrosion penetration. The rate of crevice corrosion penetration (*i.e dy/dt*) is proportional to $t^{n-1}$ meaning that the rate of penetration changes with increasing time, particularly over time scales. This position is established as follows for freshwater, brackish and marine respectively in form of Equations (5), (6) and (7):

$$\frac{dy}{dt} = 0.00412t^{-0.18} \tag{5}$$

$$\frac{dy}{dt} = 0.0182t^{-0.13} \tag{6}$$

$$\frac{dy}{dt} = 0.0257t^{-0.11} \tag{7}$$

This behavior for rate of penetration against time is illustrated in Figure 3 for freshwater, brackish and marine environments.

The Morphological Micrographs of corroding creviced SS-304 after 45 days of immersion in freshwater, brackish and marine environments are presented in Figure 4 a,b,c, this explains the extent of crevice corrosion damage based on environmental chloride strength.

## 4. Conclusions

The proposed mathematical model allows the estimation of crevice corrosion propagation in terms of penetration

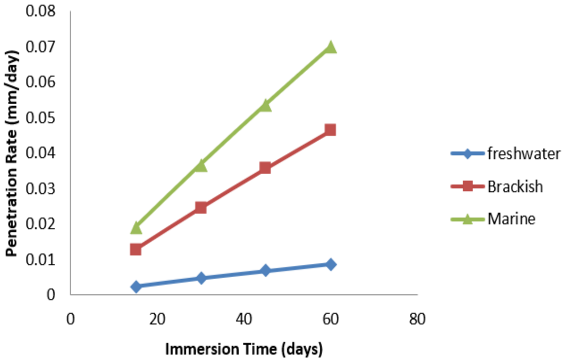
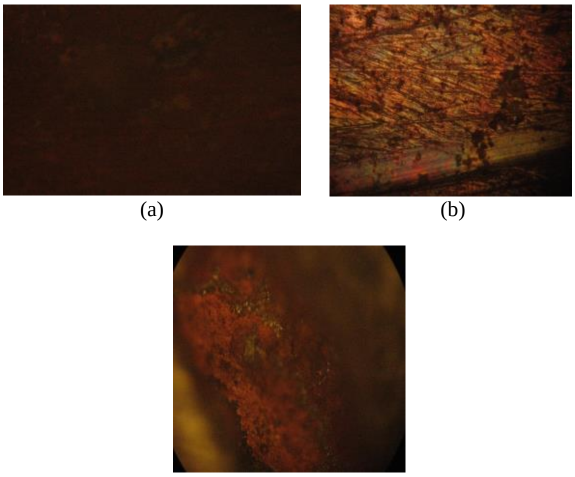


Figure 3. Effect of immersion time on rate of penetration in freshwater brackish and marine environments


(a) (b)

(c)

Figure 4. Morphological micrographs of corroding SS-304 after 45 days of immersion in (a) Freshwater (b) Brackish (c) Marine environments

depth and rates from process parameters that are typically measurable under a diffusion control regime of crevice corrosion of ss-304 in freshwater, brackish and marine environments. Comparison of modeled and measured data showed excellent quantitative agreement. No empirical adjustment to the theoretical development of the model is mandatory before using the model to explain or predict the real system. This result gives strong support to the application of models for assessing ss-304 corrosion in creviced chloride systems under the specified metallurgical and environmental conditions.

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