A NEW INDEX FOR THE CREVICE CORROSION RESISTANCE OF MATERIALS

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A New Index for the Crevice Corrosion Resistance of Materials

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Recent studies have revealed the crucial role played by the macro corrosion cell (potential coupling between the inside and outside of a cavity) in crevice and pitting corrosion. It was found that acidification and the existence of chloride ions in the local cell are not the sole and necessary conditions for localized corrosion to occur, and that their accelerating effects on crevice corrosion and pit growth can be explained within the frame work of the macro cell (the IR drop mechanism). Upon analysis of the results of the experiments, quantitative modeling, and the literature, a new characteristic parameter - the critical distance into the crevice, $d_c$ - has been suggested for indexing the crevice corrosion resistance of a material under specified conditions. The advantages of using $d_c$ as the index of the crevice corrosion resistance are: (1) it may be obtained through experiment and may also be estimated through computational approaches; (2) it has a distinct and straightforward physical meaning; (3) it may be employed in engineering design and (4) it is a single parameter which can reflect the integrated influence of several factors known to affect the crevice corrosion resistance of a material from past practical experience and research work. Preliminary work has shown good agreement between the measured and the computed values of $d_c$. The experimental technique and the principle of the mathematical approach to obtain $d_c$ are described.
Yuan Xu\textsuperscript{1} and Howard W. Pickering\textsuperscript{2}

\textbf{A NEW INDEX FOR THE CREVICE CORROSION RESISTANCE OF MATERIALS}


\textbf{ABSTRACT:} Recent studies have revealed the crucial role played by the macro corrosion cell (potential coupling between the inside and outside of a cavity) in crevice and pitting corrosion. It was found that acidification and the existence of chloride ions in the local cell are not the sole and necessary conditions for localized corrosion to occur, and that their accelerating effects on crevice corrosion and pit growth can be explained within the framework of the macro cell (the IR drop mechanism). Upon analysis of the results of the experiments, quantitative modeling, and the literature, a new characteristic parameter - the critical distance into the crevice, $d_c$ - has been suggested for indexing the crevice corrosion resistance of a material under specified conditions. The advantages of using $d_c$ as the index of the crevice corrosion resistance are: (1) it may be obtained through experiment and may also be estimated through computational approaches; (2) it has a distinct and straightforward physical meaning; (3) it may be employed in engineering design and (4) it is a single parameter which can reflect the integrated influence of several factors known to affect the crevice corrosion resistance of a material from past practical experience and research work. Preliminary work has shown good agreement between the measured and the computed values of $d_c$. The experimental technique and the principle of the mathematical approach to obtain $d_c$ are described.

\textbf{Keywords:} quantitative test, predictive method, accelerated test, corrosion testing, crevice corrosion susceptibility

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INTRODUCTION

Numerous crevice corrosion testing methods have been proposed and used with varying success. ASTM Specification G78 provides guidance in the conduct of crevice corrosion tests for stainless steels and related nickel-base alloys in sea-water and other chloride-containing environments [1], although it does not provide any particular test technique. The large amount of testing methods may be divided into two major groups. The first includes those methods which involve the use of artificial crevices. Samples with artificial crevices are immersed into the testing solution for a period of time. The crevice corrosion resistance is evaluated by the number of crevices which are found to have been corroded during the test. The spool specimen test racks [2], Ferric chloride tests [3], the Materials Technology Institute Tests (MTI-1 to MTI-5) [4] and Multiple-Crevise Assembly Testing [5] fall in this group. Among these, the Multiple-Crevise Assembly Testing [5] is the most often used. The other group employs electrochemical techniques. It includes two ASTM standard methods, ASTM G61 for iron-, nickel- and cobalt-based alloys [6] and ASTM F746 for metallic surgical implant materials [7], as well as other methods, e.g., potentiostatic test [8]; potentiodynamic test [9] and remote crevice assemblies test [10]. Although each of the techniques has its own merits and has been used with varying success, the currently used testing methods have several common problems. For instance, the data obtained through one of these tests can only serve the purposes of comparison and screening but can not be used directly for quantitative engineering design. Another problem is that data obtained by the different methods are not convertible to each other. Therefore, no method can be claimed to be the best. The reason for this may lie in the lack of a generally recognised theory on crevice corrosion so that it is difficult to link the method itself and the obtained data with the crevice corrosion mechanism.

In the present paper, a brief introduction on the progress of the mechanistic study of crevice corrosion is given first. Based on the new progress, a characteristic parameter, $d_c$ - the critical distance into the crevice - is proposed to index the crevice corrosion resistance of a metal. The advantages for using $d_c$ are discussed. The suggested experimental technique and the mathematical approach for obtaining $d_c$ are described.

PROGRESS ON THE UNDERSTANDING OF CREVICE CORROSION

Two major mechanisms on crevice corrosion have been proposed. The one based on the solution composition change within crevices [e.g. 11] suggests that the hydrolysis of dissolved metal ions increases the acidity (lowers the pH) and the resulting autocatalytic effect increases appreciably the metal dissolution rate (causing crevice corrosion). However, how the so-called autocatalytic effect can increase the corrosion rate within a crevice has never been explained. As a matter of fact, the actual metal dissolution rate measured in an acidified solution (equivalent to the hydrolysed crevice electrolyte solution) is far less than the observed crevice corrosion current density. Furthermore, crevice corrosion sometimes occurs in the absence of a pH decrease. Another mechanism, the IR-drop mechanism [12-13] focuses on the macro corrosion cell between the active crevice wall and the passivated sample's outer surface where the (cathodic) reduction of oxygen occurs. Or, in a potentiostatic test, the cathodic reduction occurs at the
counter electrode. The crevice corrosion process may be explained by the potential distribution along the crevice wall (Fig. 1). Due to the electric
shield effect, the electrode potential at some distance into a sufficiently deep crevice always remains at the mixed or equilibrium potential in the crevice electrolyte, referred to as the limiting potential (12,13), irrespective of the more oxidizing potential at the outer surface. The latter is established either by a potentiostat or by the reduction of dissolved oxygen in the bulk solution at the passivated outer surface. Thus, along the crevice wall, the local electrode potential decreases gradually from the value at the crevice opening (the more oxidizing potential) to a less oxidizing potential inside the crevice. At a certain distance, $d_c$, where the electrode potential equals the passivation potential of the anodic polarization curve of the crevice electrolyte, the crevice wall changes its state from passive to active. Thus, active dissolution occurs beyond $d_c$ to the distance where the potential decreases to the limiting potential. The crevice wall dissolution current is highest at distances slightly greater than $d_c$, corresponding to the peak current in the anodic polarization curve. This can be seen in cross sections of the crevice wall as shown in Fig. 1c where the penetration of the crevice corrosion is deepest just to the right of $E_{pass}$. Since this potential distribution is controlled by the IR drop of the dissolution current flowing out of the crevice (i.e., the IR drop within the crevice must exceed the difference between the sample's outer surface potential and the passivation potential, $E_{pass}$, of the crevice electrolyte polarization curve, IR > $s_f$ (Fig. 1), in order that crevice corrosion occurs), this process is referred to as IR induced crevice corrosion. The IR drop mechanism suggests: (1) Crevice corrosion is due to the macro corrosion cell involving well separated anodic and cathodic reactions occurring at very different electrode potentials. This is consistent with the generally accepted current understanding of crevice corrosion. (2) The highest metal dissolution rate within a crevice corresponds to the peak current of the crevicing electrolyte polarization curve. That is, the crevice corrosion current is the anodic dissolution rate at high anodic overpotentials within the active region, which is hundreds or even a thousand times larger than the corrosion rate at the limiting potential due to local micro corrosion cells deeper in the crevice or at the corrosion potential (or applied potential) existing at the outer surface in the passive region. The IR mechanism overcomes the difficulty of the traditional hydrolysis mechanism which is solely based on the composition changes of the solution, not considering the distribution of anodic overpotential on the crevice wall.

The IR-drop mechanism can satisfactorily explain the accelerating effect on crevice corrosion of the pH decrease (due to hydrolysis) and of the chloride ion build up in the crevice. The pH decrease of the crevice electrolyte and the chloride ion build up may always result in a higher peak current and a more noble passivation potential in the anodic polarization curve, which in turn increases the maximum anodic dissolution rate on the crevice wall (IR increases) and decreases the $D_f$ value. Consequently, the total crevice corrosion current increases. The increase in IR and decrease in $D_f$ both lead to a decrease in the $d_c$ value.

**CRITICAL DISTANCE INTO THE CREVICE, $d_c$**

The critical distance into the crevice, $d_c$, at which the crevice wall changes its state from passive into active, has a specific meaning for crevice corrosion. When the depth of a crevice is less than the maximum critical distance under the specified conditions, the crevice wall will be
fully passivated and no crevice corrosion occurs. On the other hand, when the depth is larger than the maximum critical distance, the crevice will be active at distances into the crevice greater than the \( d_c \) distance. It has been found, in both experiments and computations [15-19], that the critical distance into the crevice, \( d_c \), is affected by several parameters as shown in Figs. 2 and 3. The crevice gap dimension has a remarkable influence on \( d_c \). The larger the crevice gap, the larger the \( d_c \) (Fig. 2a). On the other hand, the crevice depth, \( d_0 \), has a weak influence and \( d_c \) increases slightly as \( d_0 \) decreases to the \( d_c \) value, i.e., the maximum \( d_c \) occurs for \( d_c = d_0 \) [14]. The magnitude of the passive current density affects \( d_c \) to a less extent than the gap dimension, with a larger passive current giving rise to a smaller \( d_c \) (Fig. 2b). A high solution conductivity decreases the resistance and so increases \( d_c \) (Fig. 2c). Also the higher (more oxidizing) the electrode potential (produced by an oxidant or power supply) at the sample's outer surface, the larger the \( d_c \). An almost linear relationship exists between \( d_c \) and the outer surface electrode potential (Fig. 3). In addition the anodic behaviour of the metal, especially the peak current in the anodic polarization curve, has been found to significantly influence \( d_c \). To have the same critical distance for a larger crevice gap requires a higher peak current density for otherwise identical conditions [18]. All these relations are fully compatible with the IR drop mechanism.

The above factors affecting \( d_c \) are surprisingly consistent with those factors which are known, by experience and past research work, to influence the crevice corrosion resistance of materials. In the work by Fitzgerald and his predecessors [20-23], the crevice corrosion resistance has been found to increase with increasing crevice gap, passive potential range and solution conductivity but with decreasing peak current density of the anodic polarization curve and the active potential range. These are exactly the same factors affecting \( d_c \) described above. Therefore, it may be suggested to use the critical distance into the crevice, \( d_c \), as the index of the crevice corrosion resistance of a material, i.e., the larger the \( d_c \) in a specified situation, the more resistant the material is to crevice corrosion.

The advantages of using \( d_c \) are apparent. First, it has a distinct and straightforward meaning. That is, \( d_c \) is the distance into a crevice beyond which crevice corrosion occurs. Secondly, \( d_c \) may be used in corrosion prevention design. As stated before, when the depth of an existing crevice \( d_0 \), is less than \( d_c \) for the specified conditions, the crevice will be in the passive state and so it can be tolerated. But when \( d_0 \) is larger than \( d_c \), the crevice wall beyond \( d_c \) will be in the active state and crevice corrosion will occur down to the of the limiting potential. A Nomograph or table of the relationship between \( d_c \) and the crevice geometry in a specified environment/metal combination may be prepared by computations (as explained later) and experiments. By referring to the graph or table, one may find out whether a crevice is safe (no crevice corrosion) or not. Thirdly, \( d_c \) is a single parameter to represent the crevice corrosion resistance. It combines the comprehensive effects of the several factors affecting the crevice corrosion resistance (the peak current density, the solution conductivity, the passive potential, etc.).

**Experiment For Measuring \( d_c \)**

The experimental set up for measuring \( d_c \) is given in Fig. 4a. It consists of a three electrode corrosion cell, a potentiostat and a current recording device. The working electrode is made of the material to be tested. An artificial crevice is made on it. Two kinds of artificial crevice may be used: the straight crevice (Fig. 4b) and the cylindrical crevice...
Fig. 2 The critical distance into the crevice, $d_c$, as a function of the (a) crevice gap dimension, (b) passive current density, and (c) solution conductivity.
Fig. 3 Effect of the applied potential at the sample's outer surface on the critical distance into the crevice, $d_c$. (a) High purity iron in a 0.5 M acetic acid + 0.5 M sodium acetate solution. Computation results are in good agreement with the experiments. (b) High purity iron in 1 M ammonia hydroxide + 1 M ammonia nitrate.
The former has been used for a long time in the Corrosion Laboratory of the Department of Materials Science and Engineering, The Pennsylvania State University. The straight crevice is made of a flat pyrex sheet on which a groove, 0.5 cm x 1.0 (or longer, say 2.0) cm x g, was cut, where g is the crevice gap dimension which can be made as small as 25 mm. The grooved face of the pyrex sheet is pressed against the flat surface of the sample. Thus, an artificial crevice of dimensions 0.5 cm x 1.0 cm (or longer) x g cm is formed. One advantage of using the straight crevice is that a camera may be installed in front of the pyrex sheet so that the crevice corrosion process may be observed and recorded in situ [17]. The cylindrical crevice is made of a hole drilled into a bulk component and a core component which is to be inserted into the hole. The core component may be positioned in the center of the hole by an orifice. The crevice is formed between the walls of the hole and the core. The bulk component is made of the testing metal or alloy and the core is made of an insulating high polymer, or the opposite. Alternatively, both the bulk and the core components may be made of the material to be tested. This device is similar to that used by France and Greene [23]. By adjusting the diameters of the hole and the core, the crevice gap may be adjusted.

The Luggin probe is placed close to the crevice opening to minimize the IR drop between the sample's outer surface and the opening of the capillary. This is particularly important when the solution conductivity is low. The area of the counter electrode should be large enough to make the potential distribution at the sample's outer surface more uniform so that the electrochemical test may best simulate the actual crevice corrosion situation in the natural environment. The test solution can be the one in which the metal is immersed during its service or a specially prepared corrosive solution for accelerating the test. The applied potential at the sample's outer surface must be within the passive region of the anodic polarization curve of the tested material. For comparison of different materials, the applied potential and the crevice gap dimension should be the same. The total current of the circuit is monitored from the beginning of the test. It takes a few hours for the current (which is mainly the current flowing out of the crevice) to reach a relatively stable value. After a period of time (one or two days if the peak current is large, or one or two weeks if the peak current is low), the experiment is terminated and the crevice wall or cross section is checked in an optical microscope to measure the critical distance. Sometimes the current declines to a very low value after an initial high reading, indicating that the crevice has become passivated. For some material/environment combinations, e.g., 304 stainless steel in sea water, the anodic polarization curve does not exhibit an active-passive transition and so the initial current reading is low. It takes longer time for the acidification and chloride on buildup to occur, and then the crevice corrosion current increases with time.

In the steady state, the potential distribution in the system observes the Laplace equation. For a straight crevice, in Cartesian coordinates, it is written as

$$\frac{\partial^2 \phi}{\partial x^2} + \frac{\partial^2 \phi}{\partial y^2} = 0$$
Fig. 4  (a) Experimental set up for measuring the critical distance into the crevice, $d_c$: 1. working electrode (the sample); 2. crevice; 3. Luggin capillary; 4. counter electrode; 5. gas purger; 6. reference electrode; 7. precision resistor for current measurement; 8. potentiostat; 9. pen recorder. (b) straight crevice with transparent (Plexiglas) wall. (c) cylindrical crevice.

For a cylindrical crevice, in cylindrical coordinates, the Laplace equation has the following form:

$$\frac{\partial^2 \phi}{\partial r^2} + \frac{1}{r} \frac{\partial \phi}{\partial r} + \frac{\partial^2 \phi}{\partial z^2} = 0$$

**ESTIMATION OF $d_c$ BY COMPUTATION**

Another advantage of using $d_c$ is that it can be estimated by computation. The coordinate system for the computation is shown in Fig.5.
Fig. 5. (a) Boundary conditions for the computation of $d_c$ (Cartesian coordinates, straight crevice). (b) Boundary conditions for the computation of $d_c$ (cylindrical coordinates, cylindrical crevice).

The boundary condition at the sample's outer surface ($S$ in Fig. 5) is:

$$\varphi_S = -E_1$$

where $E_1$ is the applied potential at the sample's outer surface. Along boundaries encircling the system ($B$ in Fig. 5), the boundary condition is:
In the upper part of the crevice wall (passivated part, C₁ in Fig. 5), the boundary condition is:

\[
\left( \frac{\partial \phi}{\partial n} \right)_{C_1} = 0
\]

where \( i_{\text{pass}} \) is the passive current and \( \sigma \) is the solution conductivity. In the bottom part of the crevice (active dissolution part, C₂ in Fig. 5), the boundary condition becomes:

\[
\phi_{C_2} = -E_{C_2} = f(i_{C_2}) = f\left(-\frac{\sqrt{\sigma}}{\gamma} \frac{\partial \phi_{C_2}}{\partial n}\right)
\]

where \( E_{C_2} = f(i_{C_2}) \) is the active loop portion of the anodic polarization curve. This boundary condition is nonlinear with respect to the potential and its gradient.

The above Laplace equation contains a nonlinear boundary condition and so cannot be solved by conventional numerical methods. A boundary variation and trial and error technique may be used to solve the problem. The full details of the computation method have been given elsewhere [14–16]. The solution of the Laplace equation is the potential distribution in the system, including on the crevice wall. Thus the critical distance into the crevice, \( d_c \), where the potential is decreased to the passivation potential, \( E_{\text{pass}} \), may be obtained. From the potential distribution and the polarization curve, the current distribution along the crevice may be determined. Then by integration, the total crevice corrosion current, \( I \), may also be calculated. Preliminary computation has shown good agreement between the computed and the experimental values. Tables 1 and 2 and Fig. 2a are comparisons of the computation results with experiments for high purity iron in an acetic acid buffer solution [16].

TABLE 1. Comparison between the computational [15,16] and experimental data [16,18] of the critical crevice distance into the crevice, \( d_c \), and the total crevicing current, \( I \), for pure iron in 0.5 M acetic acid + 0.5 M sodium acetate solution. Crevice gap: \( g = 0.05 \text{ cm} \); applied potential at the sample's outer surface: \( E_1 = 115 \text{ mV (SCE)} \); crevice length: \( l = 0.5 \text{ cm} \); crevice depth: \( d_0 = 1.0 \text{ cm} \).

<table>
<thead>
<tr>
<th>Critical distance, ( d_c ) (cm)</th>
<th>Total crevicing current, ( I ) (mA)</th>
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<tr>
<td><strong>Computational</strong></td>
<td><strong>Experimental</strong></td>
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<tr>
<td>0.14</td>
<td>0.12–0.20</td>
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It is noted here that the above computation can only give an estimation of the $d_c$ because of two reasons. (1) the polarization curve of the crevice electrolyte should be used in the computation. If the polarization data for the bulk solution is used, the obtained $d_c$ is accurate only at the beginning of the crevice corrosion process when the composition of the crevice solution has not changed appreciably. With the progress of crevice corrosion, the composition and pH of the crevice electrolyte change with time and so does $d_c$. (2) The shape of the crevice changes with time due to corrosion so that the boundary condition on the crevice wall should also be changed with time. Thus, using the original boundary condition results in additional errors in $d_c$. Therefore, although the computation can sometimes give a good estimation of $d_c$, it cannot replace the experiment.

TABLE 2. Comparison between the computational and experimental data of the critical distance into the crevice, $d_c$, for high purity iron on 0.5 M acetic acid + 0.5 M sodium acetate solution at different applied potentials, $E_1$, at the sample's outer surface. Crevice gap: $g=0.05$ cm; crevice depth: $d_o = 1.0$ cm.

<table>
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<th>Applied potential, $E_1$ (mV SCE)</th>
<th>Critical distance, $d_c$ (cm)</th>
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<tr>
<td>115</td>
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<td>485</td>
<td>0.37</td>
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SUMMARY

A new index for the corrosion resistance of materials - the critical distance into the crevice, $d_c$ - has been proposed. At $d_c$, the local potential of the crevice wall is the passivation potential in the anodic polarization curve of the material in the crevice electrolyte. At greater distances, the crevice undergoes active anodic dissolution (crevice corrosion) down to the distance of the limiting potential. The larger the $d_c$, the better the crevice corrosion resistance of the material. Two artificial crevice designs have been introduced which can be used to measure the critical distance into the crevice. The $d_c$ may also be estimated through computation. The advantages in using $d_c$ as the index of crevice corrosion resistance have been discussed.
ACKNOWLEDGEMENT

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