Practical Modeling of Uniform Steel Corrosion in Concrete

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Abstract:

Steel corrosion in concrete can be modeled by solving the Laplace's equation for potentials with anodic and cathodic polarization boundary conditions. Although the solution is non-linear and computationally expensive due to imposed boundary conditions, it is still possible using specialized numerical procedures. The main difficulty lies in the prediction of the locations of anodic and cathodic sites along the steel surface. In the case of uniform corrosion that takes place due to pH-altering processes, such as carbonation, the determination of the anode-to-cathode ratio is the main challenge. In this study, a model is developed to determine the optimum (i.e. equilibrium) anode-to-cathode ratio that corresponds to the maximum corrosion rate in uniformly depassivated steel in concrete. The model is a function of resistivity and oxygen concentration around the reinforcement. The result of the model is verified with available experimental data.

1. Introduction

Quantification of active corrosion rate of steel in concrete through nondestructive and rapid methods is a crucial task for scheduling maintenance/repair operations and for achieving accurate service life predictions. Significant progress has been made on the nondestructive measurement of steel corrosion in concrete, and RILEM has already made recommendations for best practices for quantifying corrosion rates [1]. One of the most widely-used methods of measuring corrosion rate in concrete structures is by means of measuring the polarization resistance, R_{p} , of the corroding system and using the Stern-Geary equation to calculate the corrosion current density of active steel. Since calculations that are based on measured polarization resistance data provides instantaneous corrosion rates that are influenced greatly by changes in temperature and humidity, realistic interpretations of the measurements can only be made after integrating polarization resistance data over time. Therefore, although the polarization resistance methods are quite practical for instantaneous monitoring of structures, they can become expensive and impractical when monitoring is to be done continuously over long periods.

The main objective of this paper is to develop a practical numerical model for predicting corrosion rate of an actively corroding steel reinforcement in concrete. It should be noted that this paper discusses only the uniform corrosion of steel in concrete (e.g. uniform corrosion after the carbonation of concrete cover), in which active steel surface is assumed to be covered with uniformly distributed anodes and cathodes. The development of a practical model for pitting corrosion in concrete has its challenges, and research in this area is still being carried out by the authors; therefore the scope of this paper is limited to uniform corrosion. The main assumption used in this model is built on Stern's earlier work [2] that an optimum anode-to-cathode ratio exists for which the corrosion current on the metal surface reaches a maximum value. In the first part of the paper, the background to this approach will be explained to provide a proof of concept. After the proof of concept, a more realistic arrangement of uniform corrosion of steel will be investigated numerically using the finite element method to obtain a practical numerical model for corrosion rate. Finally, the developed model will be validated using experimental data obtained from the literature.

2. Background

Stern studied the effect of anode-to-cathode ratio on the corrosion rate of a metal in a system where both anodic and cathodic polarization follow Tafel behaviour (i.e. governed by activation polarization only) and *IR* drop is negligible [2]. Based on this study, it was analytically demonstrated that maximum corrosion current, I_{corr} , occurs when:

(1)
$$\frac{A_a}{A_c} = \frac{\beta_a}{\beta_c}$$

where A_c [m²] is the cathode area, A_a [m²] is the anode area, β_a [volt/dec] is the Tafel slope of the anodic reaction, and β_c [volt/dec] is the Tafel slope of the cathodic reaction. This means that for a metal with similar anodic and cathodic Tafel slopes (i.e. $\beta_a \approx \beta_c$), the maximum corrosion rate occurs at an anode-to-cathode ratio of unity. At any other anode-to-cathode ratio, the corrosion rate would be smaller, reaching zero at a ratio equal to zero or infinity [2]. It is well established that the corrosion of steel in concrete is rather complicated, and using Stern's assumptions may oversimplify the problem [3]. In order to clarify this point, a brief review of the corrosion process in concrete is presented here.

The corrosion of steel in concrete is a result of iron dissolution in pore water which can be represented by the following half-cell reaction:

(2) Fe
$$\rightarrow$$
 Fe²⁺ + 2e⁻

The electrons that are produced in this anodic reaction must be consumed at the cathodic sites on the steel surface to preserve electrical neutrality. The cathodic reaction in which these free electrons are consumed is the oxygen reduction given by:

(3)
$$\frac{1}{4}O_2 + \frac{1}{2}H_2O + e^- \to OH^-$$

The corrosion rate at any point on the surface of steel in concrete is related to the current density, which can be predicted if the electrical potential (abbreviated henceforth as "potential") distribution around that point is known. Once the potential distribution along the reinforcement is known, the current density at any point on the steel surface can be calculated by:

(4)
$$i = -\frac{1}{\rho} \frac{\partial \phi}{\partial n}$$

where *i* $[A/m^2]$ is the current density, ϕ [volts] is the potential, ρ [Ω -m (ohm-m)] is the electrical resistivity of concrete and *n* is the direction normal to the equipotential lines. As it can be observed from Eq.4, the calculation of current densities requires the knowledge of potential distribution around the reinforcement. The equation governing the potential distribution can be derived from first principles [4]. Assuming electrical charge conservation and isotropic conductivity, the potential distribution can be represented by the Laplace's equation

$$(5) \qquad \nabla^2 \phi = 0$$

where ∇^2 is the harmonic operator. Calculation of the potential distribution around the surface of the steel involves the solution of Eq.5 subjected to prescribed boundary conditions that comprise the relationship between potential and current density for the anodic and cathodic regions as well as prescribed current densities across domain boundaries. For the anodic and cathodic regions of the steel surface, the boundary condition can be defined as:

(6) $\phi = \phi_a$

$$(7) \qquad \phi = \phi_c$$

where ϕ_a and ϕ_c [volts] are the polarized surface potentials of the anodic and cathodic surfaces, respectively. In reinforced concrete, polarization of anodic surfaces can be considered to be activation limited and can be written as [5]:

(8)
$$\phi_a = \phi^o{}_{Fe} + \beta_a \log \frac{i_a}{i_{oa}}$$

where ϕ^{o}_{Fe} is the standard half-cell potential of iron, i_{a} is the anodic current density, and i_{oa} is the anodic exchange current density. Although the anodic reaction can be assumed to be controlled by activation polarization, the effect of concentration polarization on the cathodic reaction cannot be ignored since the oxygen concentration around the cathodic sites on the steel surface may be low, resulting in further polarization. The polarization of the cathodes can be presented by [2, 5]:

(9)
$$\phi_c = \phi_{O_2}^o + \beta_c \log \frac{i_c}{i_{oc}} - \frac{2.303RT}{z_c F} \log \frac{i_L}{i_L - i_c}$$

where $\phi^{\circ} o_2$ [volts] is the standard half-cell potential of oxygen, i_c [A/m²] is the cathodic current density, R is the universal gas constant (\approx 8.314 [J/mol.K]), T [K] is the temperature, F is the Faraday's constant (\approx 96500 [C/mole]), z_c is number of electrons that are involved in cathodic reaction, i_{oc} [A/m²] is the exchange current density of the cathodic reaction, and i_L [A/m²] is the limiting current density given by [2]:

(10)
$$i_L = 6.25 z_n C_{o_2}$$

where z_n is the number of electrons transferred (for oxygen reduction, $z_n=4$) and C_{O_2} [kg/m³] is the oxygen concentration in concrete pore solution near the rebar surface around the cathodic regions. In addition, the *IR* drop can become a factor in the corrosion of steel concrete as the concrete resistivity can vary significantly in reinforced concrete structures due to environmental factors.

Using an analytical approach similar to Stern's, one can obtain the maximum corrosion current, I_{corr} , of a one-dimensional problem shown in Fig.1, which illustrates one anode and one cathode separated by a concrete layer. It should be noted that although this model can represent a number of scenarios for corrosion of steel in concrete structures, it is not a typical problem that is encountered in real world. However, for the purpose of proving the concept before the detailed finite element investigation of a more realistic system, the arrangement in Fig.1 will be simulated using the following closed-form solution [6]:

(11)
$$(i_a A_a) R_{conc} = \phi^o{}_c - \phi^o{}_a - \beta_c \log i_{oc} + \beta_c \log r + \beta_a \log i_{oa} + (\beta_c - \beta_a) \log i_a$$
$$- \frac{2.303 RT}{z_c F} \log(\frac{i_L}{i_L - i_a \cdot r})$$

Eq.11 is the closed-form solution of the Laplace's equation for anodic current density. Using a nonlinear solution algorithm and the parameters in Table 1, the corrosion current, $I_{corr} = i_a A_a$, for the one-dimensional problem defined in Fig.1 can easily be calculated. In Fig.2, the corrosion current, I_{corr}, vs. anodic-to-cathode ratio, r, for various oxygen concentrations and concrete resistivities are plotted by solving Eq.11 numerically. It is clear from this figure that there is an optimum value of anode-to-cathode ratio for which the corrosion current reaches its maximum value. Based on the basic thermodynamic principles, this maximum can be interpreted as corresponding to the state at which the total energy is minimized by achieving an optimum (or equilibrium) anodeto-cathode ratio along the reinforcement. In this paper, this anode-tocathode ratio that lead to the maximum corrosion current will be defined as the "equilibrium state." It should be noted that there are claims against this argument as a number of researchers state that anodes and cathodes on the steel surface can form at the same location at the microscopic level; hence it is impossible to define an anode-to-cathode ratio [2]. Although this claim has its merits, within the scope of uniform corrosion of steel rebars in concrete, a macro-level anode-to-cathode ratio can still be defined, and this approach will be adopted here in this paper as well.

Parameter	Value / Range
Standard cathode potential (Eoc)	0.160 V (SCE)
Standard anode potential (Eoa)	- 0.780 V (SCE)
Cathodic Tafel slope (β_c)	- 0.160 V/dec
Anodic Tefel slope (β_a)	0.060 V/dec
Cathodic exchange current density (ioc)	10 x 10 ⁻⁶ A/m ²
Anodic exchange current density (ioa)	300 x10 ⁻⁶ A/m ²
No. of electrons in the cathodic reaction (z_c)	1
Limiting current density (i_L)	calculated
Resistivity (p)	50 to 1000 Ω.m
Oxygen concentration (C_{O2})	2.5x10 ⁻⁴ to 1.25 x10 ⁻⁴ kg/m ³

Table 1: Input parameters used in the analyses presented in the paper

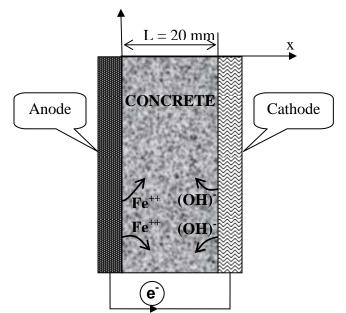


Figure 1: One-dimensional macrocell corrosion in concrete with parallel anodic and cathodic surfaces

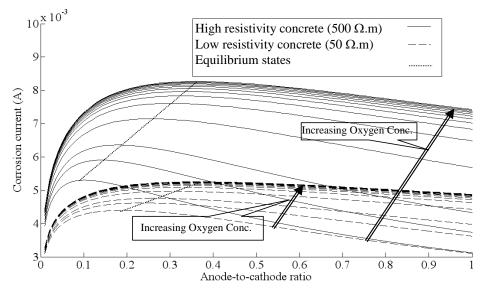


Figure 2: The effect of anode-to-cathode ratio on corrosion current for 1-D simulation

3. Practical Modeling of 2-D Uniform Steel Corrosion in Concrete

Although the analytical model for a one-dimensional system can represent a number of scenarios for corrosion of steel in concrete structures, it is not a typical problem that is encountered in real world. A more realistic, albeit still simplified, model of steel corrosion in concrete is presented in Fig. 3. This presentation can be used to simulate uniform steel corrosion in

concrete, as observed in structures experiencing extensive carbonation. Analytical solution of this problem is not possible due to the twodimensional geometry and the nonlinear boundary conditions imposed by the polarized steel surface. Therefore, using finite element technique, an extensive numerical investigation was carried out to investigate the effect of anode-to-cathode ratio, concrete resistivity and oxygen concentration on the corrosion of steel in concrete. The analysis was carried out using data presented in Table 1 and assuming a domain size of 100 mm x 300 mm. The domain size was selected after a comprehensive sensitivity analysis, but this discussion will not be presented here. More information on the selection of parameters can be obtained from reference [6]. In order to quantify the corrosion current, a comprehensive regression analysis has been carried out on the results of the numerical experiments. Based on the regression analysis ($R^2 = 0.97$, Maximum Error = 2.83x10⁻⁶ A), the following equation for corrosion current has been obtained for the corrosion of uniformly depassivated steel rebars in reinforced concrete members:

(12)
$$I_{\text{max}} = -4.00 \times 10^{-6} + \frac{0.009}{\rho} - 1.15 \times 10^{-6} \ln(C_{O_2}) + 0.001 \frac{\ln(C_{O_2})}{\rho}$$

where I_{max} is the maximum corrosion current [A]. Normalizing I_{max} with the surface area of steel, A_s , over which the analysis was carried out, we obtain:

(13)
$$i_{corr} = \frac{I_{max}}{A_s} = -1.33 \times 10^{-3} + \frac{3.00}{\rho} - 3.83 \times 10^{-4} \ln(C_{O_2}) + 0.333 \frac{\ln(C_{O_2})}{\rho}$$

where i_{corr} is the corrosion current density [A/m²]. Equation 13 can be used to predict the corrosion rate of uniformly depassivated steel in concrete structures and to verify or to extrapolate nondestructive corrosion rate measurements such as the ones that are based on polarization resistance techniques.

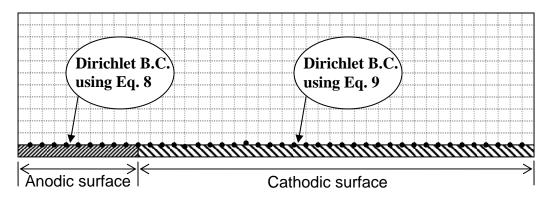


Figure 3: Finite element mesh used to develop the corrosion model

4. Experimental verification

In a recent study by Luping [7], three types of instrument for measuring corrosion rate were tested under controlled conditions. Two of these instruments were using galvanostatic pulse technique; the last one was based on linear polarization resistance technique. In this paper, these instruments are referred to as GSP1, GSP2 and LPT, where the acronyms GSP and LPT refer to galvanostatic pulse technique and linear polarization resistance technique, respectively. A part of this experimental study is used to verify the proposed equations presented in this paper. A brief summary of the experimental setup and procedure is presented here, but further details can be obtained from [7].

In the experimental study, plain cool-drawn carbon steel bars of 10 mm diameter were used as reinforcement. The steel bars were cleaned with degreasing agent and acetone. The ends of each bar were coated with cement grout and epoxy to avoid unexpected crevice corrosion. Concrete batches with a water-cement ratio of 0.5 were pre-contaminated with chloride ions at concentrations of 1.5, 3 and 6% by mass of cement. Concrete slabs of size 250 × 250 × 70 mm were cast in wood moulds. Two steel bars in parallel were embedded in the centre portion of each slab at the mid-height (about 35 mm), with a space of 100 mm between each other. The slabs were kept under a relative humidity of about 85%. The resistivity data used in the model for each specimen were obtained from [7]. The resistivity of concrete specimens having 1.5, 3 and 6% chloride contamination were indicated in the figures as cases B, C and D, respectively. Case A corresponds to concrete specimens without chloride contamination.

Three comparative measurements using three types of instrument were carried out. The first comparative measurement was carried out at the age of 4 weeks. The second and the third comparative measurements were conducted at the age of 3 and 8 months, respectively. At the end of the electrochemical measurements, the steel bars were removed from each concrete slab to determine the mass loss in accordance with ASTM G1 [8].

By substituting the electrical resistivity and oxygen concentration in Eq. 13 the corrosion current density are determined. The results of these experimental studies and the predictions obtained by the proposed model (Eq.13) are shown in Figs. 4 to 6. The measurements shown for each instrument are the average of three readings. As it can be observed, and also as concluded by the researchers of the experimental study [7], there is a wide range of scatter in the measurements from different instruments. The large difference between two instruments that are based on the galvanostatic pulse technique (i.e. GSP1 and GSP2) is noteworthy.

Despite the scatter in the experimental measurements, the proposed model captures, approximately, an average value of the corrosion rate for each specimen. Especially, the measurements by GSP1 and the predictions by the proposed model compare exceptionally well, considering the fact that there are significant number of uncertainties involved in the corrosion process. But in passive state, case A, where the corrosion is not initiated, the result of the model is much greater than the measurement results. It's due to the fact that the major assumption in this corrosion modeling is a complete depassivation of steel in concrete.

Figure 7 presents the values of corrosion rate obtained from destructive method (mass loss) and proposed equation (Eq.13) as well as instrumental measurements. It should be noted that the resistivity that was considered in proposed equation is the average value along the period of exposure. As it is shown in this figure, the rates obtained by the proposed equation are in good agreement with the mass loss measurements.

5. Conclusions

A practical numerical model for predicting corrosion rate of uniformly depassivated steel in concrete is developed. The model is built on Stern's earlier work that an optimum anode-to-cathode ratio exists for which the corrosion current on the metal surface reaches a maximum value. Based on the basic thermodynamic principles, this maximum can be interpreted as corresponding to the state at which the total energy is minimized by achieving an optimum (or equilibrium) anode-to-cathode ratio along the reinforcement. The developed model, which represents the corrosion rate as a function of concrete resistivity and oxygen concentration, captures expected corrosion behavior of steel in concrete and is validated using experimental data obtained from the literature. Authors acknowledge that further experimental validation and sensitivity analyses are still necessary to increase the confidence in the model. This work is currently in progress.

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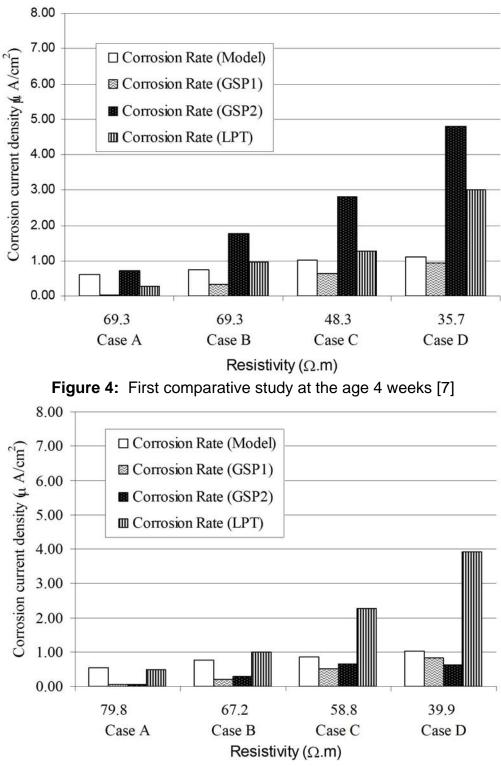


Figure 5: Second comparative study at the age 3 months [7]

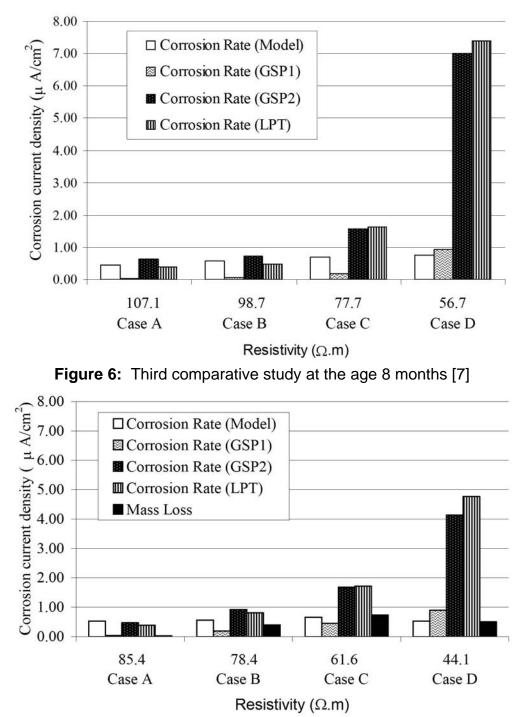


Figure 7: Comparing the mass loss of corroded rebar and the results predicted by model

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