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Polarization behavior of activated reinforcing steel bars in concrete under chloride environments

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HIGHLIGHTS

- Electrochemical parameters related to corroded rebars in concrete were examined.
- Degrees of activation polarization and concentration polarization were quantified.
- Cathodic exchange current density has the most significant effects on corrosion rate.
- A practical model for the corrosion rate of rebars in concrete is proposed.

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ABSTRACT

Polarization behavior of activated reinforcing steel bars (rebars) in concrete was investigated by use of measured cathodic polarization curves. Five factors thought to contribute to the polarization behavior of rebars in concrete were examined, including relative humidity, Cl^- content, rebar diameter, water-cement ratio and corrosion duration. The Levenberg-Marquardt curve-fitting algorithm made it possible to determine the corrosion rate and electrochemical parameters of underlying anodic and cathodic sub-process simultaneously. Degrees of activation polarization and concentration polarization related to cathodic reaction were quantified. A prediction model for the average value of corrosion rate in regard to the whole surface of a corroded rebar was proposed based on the electrochemical corrosion theory. Finally, the accuracy of this model was testified by comparing the predictions of this model with published corrosion test results.

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1. Introduction

For a reinforced concrete (RC) structure, the deterioration process with respect to the reinforcing steel bars (rebars) corrosion consists of two periods [1]: (1) An initiation period defined as the time for the aggressive substance to reach the rebars and induce depassivation; (2) A propagation period during which the rebar is actively corroding and the rate of structural deterioration increases greatly.

Up to now, extensive research has been conducted to predict corrosion initiation time; however, little focus has been placed on the assessment and prediction of corrosion rate. Accurate modeling of rebar corrosion in concrete is of great importance for the assessment of time-dependent load bearing capacity as well as the prediction of residual service life of corroded RC structures.

The corrosion of rebars in concrete is an electrochemical process. Factors which influence the corrosion propagation may be classified into two categories: direct and indirect ones. The former refers to electrochemical parameters of underlying anodic and cathodic sub-processes and the concrete resistance between the anode and cathode; the latter is related to environmental conditions, material properties and loading of the structure, etc. In view of the electrochemical nature of corrosion and the numerous influencing factors involved, the prediction of the corrosion rate is very complex.

The existing prediction models for corrosion rate may be grouped as analytical, numerical or empirical depending on the criterion used in their development. The analytical model was deduced from theoretical analysis taking all relative factors into account [2]. The numerical model presented herein involves the solution of Laplace's equation for electrical potentials with required boundary conditions and calculates the current density on the steel surface using these potentials [3–7]. The application of both the analytical and numerical models required the predeter-

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mination of all values of anodic and cathodic electrochemical parameters (i.e., direct influencing factors). The empirical model was based on assumed direct relationships between corrosion rate and some easily quantifiable indirect influencing factors [8–11]. However, due to different opinions on the control process of corrosion rate and the uncertain range of values for electrochemical parameters related to active corrosion of rebars in concrete, it is difficult to apply an analytical model in practical projects. Meanwhile, the numerical model only focused on estimating the effects of electrochemical parameters on the corrosion rate. The empirical model is only suitable for conditions under which it was developed. Consequently, it is important to understand both the control process and the value ranges of electrochemical parameters for the modeling of corrosion rate of rebars in concrete.

In general, there are two views on the control process of corrosion rate of an activated rebar in concrete: cathodic reaction control or concrete resistance control. Furthermore, cathodic reaction can be limited by two polarization factors, i.e. activation polarization and concentration polarization, which may act individually or in combination. Activation polarization is the resistance to electrochemical reaction at the metal-electrolyte interface caused by an energy barrier, which must be overcome to convert species involved in the corrosion reaction. Concentration polarization occurs as a result of a deficiency of necessary reactants (e.g., oxygen) at the metal surface. While, concrete resistance control results from dry concrete providing a significant ohmic resistance between anode and cathode. Gonzalez [12,13] found a great increase in the corrosion rate of an embedded rebar during the reduction of degree of pore saturation in mortars, PS, from 100% to 70%, and an inversely proportional relationship between logarithmic corrosion rate and mortar resistivity on the condition of 45% < PS < 70%. Hence, he speculated that the corrosion rate was under the cathodic concentration control in the case of PS > 70% and under the resistance control at 45% < PS < 70%. Raupach [14] and Hussain [15] observed the variations of the cathodic current of corroded rebars with time at large negative external potentials, and they suggested that the corrosion rate was under cathodic activation control even at a relative humidity of 95%. As the understanding of the rate-limiting step of activated rebars in concrete was mainly drawn by experimental findings qualitatively, further quantitative analysis based on electrochemical corrosion theory needs to be done.

Cathodic and anodic electrochemical parameters are usually analyzed based on measured polarization curves implemented by a 3-electrode configuration. In order to ensure a good electrical connection and minimize the effect of the resistance between the reference electrode and the rebar, either bare steel bar samples [16,17] or rebars embedded in concrete [18,19] submerged in salt solution are generally used in experimental investigations. Nevertheless, these findings may have been limited to the immersed condition. Although specific electrode configurations have been designed for rebars in concrete under atmospheric environment, they may not be suitable for extensive experimental research in view of the complicated implementation process [20–22]. Two common methods used to analyze polarization curves are the Tafel extrapolation method [20–24] and the curve fitting method [17,25]. The Tafel extrapolation method extrapolates the Tafel region of a polarization curve back to the corrosion potential so as to determine the corrosion rate. This is valid provided that the branch of the polarization curve is under activation control and a well-defined Tafel region exists, that is, the logarithm of the current density varies linearly with the electrode overvoltage over at least one decade of current. As the requirements are difficult to be met for activated rebars in concrete, the error of this method is inevitable. In contrast, a curve fitting method is more acceptable for providing an accurate evaluation of corrosion rates and electrochemical parameters. However, the existing research results [17,18,25] were all obtained on the assumption that the cathodic reaction are under pure activation control.

This paper investigates the polarization behavior of activated rebars in concrete using measured cathodic polarization curves. Fifty-four RC small beams were exposed to aggressive chloride environments in a large marine environmental chamber to induce corrosion of rebars. Five factors deemed to contribute to the polarization behavior of rebars in concrete were examined, including relative humidity, Cl^- content, rebar diameter, water-cement ratio and corrosion duration. Polarization curves were measured by a three-electrode configuration and analyzed by the Levenberg-Marquardt curve-fitting algorithm, which enabled determining the corrosion rate and electrochemical parameters of underlying anodic and cathodic sub-processes. The indicator θ, which is the ratio of the cathodic corrosion current density to the limiting current density for oxygen reduction, was introduced to quantify the degrees of activation polarization and concentration polarization related to cathodic reaction. A parametric study was carried out to identify the electrochemical parameter that has the most significant influence on corrosion rate. Taking this key electrochemical parameter as the intermediate variable, a practical model for the corrosion rate of rebar in concrete was developed based on the electrochemical corrosion theory. It should be pointed out that pitting corrosion is common for rebars in concrete structures exposed to aggressive chloride environments. Pitting corrosion is non-homogeneous longitudinally and concentrated over small areas of a rebar, which may result from macrocell corrosion. However, the effect of macrocell corrosion could be taken into account indirectly using the probability distribution model for cross-sectional area of a corroded rebar in concrete as proposed by Zhang et al. [26]. So, this paper focuses on the average value of corrosion rate in regard to the whole surface of a corroded rebar, aimed at predicting the average weight loss ratio of rebars in concrete accurately.

2. Theoretical basis of polarization behavior of rebars

Based on previous research results [12–15], it can be inferred that the control process of corrosion rate of an activated rebar in concrete (i.e. cathodic reaction control or concrete resistance control) can be determined using a critical value of concrete resistance [R_{con}]. When the resistance of concrete electrolyte between the anode and the cathode of a corroded rebar, R_{con}, is smaller than the critical value [R_{con}], the effect of R_{con} on ionic current flow of the corroded rebar can be described as follows: with the increase of R_{con}, the distance between the anode and the cathode will shortens, meanwhile, the cathode area and the effective cross sectional area for the ionic current flow will decrease accordingly. However, the corrosion current, I_{corr}, is almost constant and independent on R_{con}. Therefore, the corrosion rate can be recognized to be under the cathodic reaction control. Nevertheless, once R_{con} is more than [R_{con}], the value of corrosion current, I_{corr}, is influenced significantly by R_{con} and then the corrosion rate is under concrete resistance control. For concrete structures subjected to marine atmosphere, internal relative humidity of concrete is always within the range of 80% ~ 100% [27–29]. Considering that the corrosion rate of an activated rebar in concrete is under cathodic reaction control at such a high relative humidity [12,13,18], the effect of R_{con} on polarization behavior of rebars is ignored in this paper.

Based on the mixed potential theory, the polarization diagrams of a corroding rebar is illustrated in Fig. 1. It is considered that the anodic reaction of iron oxidation is under pure activation control, and the cathodic reaction of oxygen reduction is well described in terms of mixed activation and concentration control [30]. The

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