



Steady-state polarization response of chloride-induced macrocell corrosion systems in steel reinforced concrete – numerical and experimental investigations



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ABSTRACT

Chloride-induced macrocell corrosion of steel is recognized as a frequent and harmful problem affecting reinforced concrete structures. For this specific corrosion mechanism, anodic and cathodic areas at steel-concrete interface are spatially separated. Thereby, at equilibrium, anodic and cathodic potentials are different due to electrical resistivity of concrete, which means there is a potential gradient and a macrocell corrosion current flowing in the concrete volume. Despite some available works on electrochemical modelling of free macrocell corrosion systems in reinforced concrete, the literature is very scarce regarding the response of such systems under polarization. Due to this lack of knowledge, the assessment of steel corrosion rate in reinforced concrete is still commonly based on the uniform corrosion assumption. In this paper, the detailed polarization response of the different components of macrocell systems is studied in steady-state condition on the basis of numerical simulations and the qualitative conclusions are validated by original experiments. Some important results are achieved, with potentially major consequences regarding the non-destructive evaluation of steel corrosion in reinforced concrete. In particular, the distribution of the polarizing current in the macrocell system is found to be dependent on the polarization nature (anodic or cathodic) and magnitude. Moreover, in case of chloride-induced macrocell systems, the apparent linear polarization range is actually much higher than usually assumed and significant differences are observed between anodic and cathodic responses.

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1. Introduction

Chloride-induced macrocell corrosion of steel is recognized as a frequent and harmful problem leading to premature ageing of reinforced concrete structures. In sound concrete, the reinforcing steel is naturally protected against corrosion by a passivation phenomenon consisting in the rapid formation of a compact oxide film on steel surface. The ingress of chloride ions in concrete may lead to local breakdown of the passive film, making the steel locally active. The detailed local effect of chlorides on steel surface and the critical chloride content leading to depassivation are not the purpose of this paper, but abundant literature may be found on this issue [1–3].

The local dissolution of the passive film generates the condition of macrocell corrosion. For this type of corrosion, anodic (active) and cathodic (passive) areas at the steel-concrete interface are spatially separated. Therefore, anodic and cathodic potentials are different due to the

electrical resistivity of concrete, causing a potential gradient and a macrocell corrosion current in the concrete volume.

Electrochemical modelling and numerical simulation of free macrocell corrosion systems in reinforced concrete is a relatively recent research field. The main theoretical concepts on corrosion modelling in propagation phase are described in [4]. Kranc and Sagués provided an important contribution by implementing a finite difference approach of the problem including nonlinear electrochemical behaviour as boundary condition [5]. Different numerical approaches may be used to simulate macrocell corrosion [6,7]. However, due to the tridimensional nature of the physical problem, boundary elements [8,9] or finite elements [10,11] methods are more suitable. A numerical benchmark allows comparing the respective performances of these methods [12]. Regarding the rate of chloride-induced corrosion in reinforced concrete, an important contribution to macrocell modelling was recently provided by Warkus and Raupach [13]. The authors demonstrate the significance of geometrical effects on local corrosion rate.

Despite these research works on modelling and simulation of free macrocell corrosion, the literature is very scarce regarding the response of such systems under polarization. This topic is of major

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importance regarding the non-destructive evaluation of the corrosion activity of steel reinforcing bars in concrete. The work of Poursaeed and Hansson [14] deals with the numerical simulation of the polarization response of a macrocell system. However, the local response of steel on anodic and cathodic areas is modelled as a pure resistive electrical behaviour and the free driving potential between anodic and cathodic areas is not considered.

Due to this lack of knowledge, the assessment of steel corrosion rate in reinforced concrete is, in many cases, based on the Stern–Geary equation, which was established for the case of uniform corrosion [15–17]. The uniform corrosion is thereby implicitly assumed in corrosion rate measurements.

Regarding half-cell potential mapping, the relevance of macrocell corrosion is yet clearly recognized and RILEM Technical Committee 154-EMC recommends paying attention to half-cell potential gradients rather than half-cell potential values [18]. Concerning measurements of linear polarization resistance, RILEM protocol [16] only proposes a semi-empirical approach for taking into account the localized nature of corrosion. Sagüés and Kranc were the first to report on the difficulty in interpreting polarization measurements in the case of macrocell corrosion [19]. Based on a numerical work, Gulikers also pointed out problems related to the application of the linear polarization resistance technique to macrocell corrosion systems [20]. Elsener discussed the implication of macrocell existence on corrosion monitoring [21] and showed that the response of a macrocell system is actually more complex than the response of uniform corrosion systems. In particular, the author established that the externally applied polarizing current is collected by anodic areas in case of macrocell corrosion. Recently, Angst and Büchler called into question the applicability of the Stern–Geary equation to extract the corrosion rate from linear polarization resistance measurements in case of macrocell systems [22]. The authors developed an original analytical approach to highlight the limits of the classical LPR technique when applied on macrocell systems. Among several results, they showed that two types of errors have compensating effects that can result, in specific cases, in an apparent efficiency. However, regarding on site corrosion rate measurements, these errors cannot be controlled and make the existing techniques inaccurate.

The short preceding discussion highlights the need to improve the knowledge on the behaviour of macrocell corrosion systems under external polarization in order to better interpret on-site measurements of steel corrosion rate. In this paper, the detailed polarization response of the different components of macrocell systems is studied in steady-state condition by means of numerical simulations and the qualitative conclusions of the numerical study are verified by original experiments. Some important results are achieved, with potentially major consequences regarding further developments in the field of steel corrosion assessment in reinforced concrete.

2. Electrochemical background

In this section, the basic theoretical concepts relative to microcell and macrocell corrosion are introduced. The corrosion of steel in concrete is an electrochemical phenomenon, resulting from two concomitant half-cell reactions. In the electrochemical process, the dissolution of steel constitutes the anodic reaction (Eq. (1)), which provides electrons consumed by the cathodic reaction (Eq. (2)) corresponding to the reduction of O_2 dissolved in the pore solution of the concrete.



According to the respective spatial locations of anodic and cathodic reactions, two main types of corrosion may be distinguished in steel reinforced concrete: microcell corrosion and macrocell corrosion.

2.1. Microcell corrosion system

Reinforcing steel bars embedded in a uniform electrolytic environment exhibit a uniform electrochemical state (active or passive) and consequently a uniform corrosion rate. In this specific case, anodic and cathodic areas appear as corrosion microcells, immediately adjacent on the steel surface. Therefore, the ohmic drop between anodic and cathodic microcells due to electrolytic resistivity is negligible and each electron produced by an anodic reaction is locally consumed by a cathodic reaction. At macroscopic scale, anodic and cathodic microcells may not be dissociated, there is no current in the bulk concrete volume and the potential field appears uniform. It has to be noticed here that passive steel may be considered as a microcell corrosion system, although the corrosion rate is very low and commonly neglected.

The polarization behaviour of such uniform systems may be modelled by the Butler–Volmer equation (Eq. (3)), either for active or passive steel, using appropriate sets of electrochemical parameters.

$$i = i_{corr} \left(\exp\left(\frac{E - E_{corr}}{\beta_a}\right) - \exp\left(-\frac{E - E_{corr}}{\beta_c}\right) \right) \quad (3)$$

where:

- E_{corr} is the free corrosion potential ($V_{/ref}$) of the uniform corrosion system at equilibrium,
- i is the net current density (A/m^2) flowing through the metal–electrolyte interface of the uniform system forced at potential E (involving a polarization with respect to the equilibrium potential E_{corr}),
- i_{corr} is the corrosion current density (A/m^2), corresponding to the exchange current density of the uniform corrosion system,
- β_a and β_c are anodic and cathodic Tafel coefficients (V) of the electrochemical system, respectively.

Fig. 1 presents the typical shapes of polarization response relative to active and passive microcell systems. Qualitatively, electrochemical features of active and passive uniform corrosion systems may be distinguished as follows ('a' and 'p' indices referring to active and passive uniform systems, respectively):

- $E_{corr,a} < E_{corr,p}$,
- $i_{corr,a} > i_{corr,p}$,
- $\beta_{a,a} < \beta_{a,p}$,
- $\beta_{c,a} \approx \beta_{c,p}$.

The Butler–Volmer equation of a microcell corrosion system actually corresponds to the algebraic sum of the current density associated with the anodic reaction (Eq. (1)) and the current density associated with the cathodic reaction (Eq. (2)). For each system, the equilibrium corrosion potential ($E_{corr,a}$ or $E_{corr,p}$) corresponds to the value at which anodic current is balanced by cathodic current. At the corrosion potential, the value of anodic current density defines the corrosion current density of the microcell system (i_{corr}). Since anodic and cathodic reactions do not involve the same chemical species, the phenomenon is characterized by a dynamic equilibrium, in which steel is continuously dissolved and oxygen is reduced. At this dynamic equilibrium, corrosion of steel occurs, but no net current flows through the metal–electrolyte interface of the electrochemical system ($i = 0$), due to the balance of anodic and cathodic current densities and to potential uniformity at the steel–concrete interface.

A perturbation of such a system from the equilibrium corrosion potential is referred to as polarization and results in a net current density flowing through the metal–electrolyte interface, since one reaction is enhanced whereas the other reaction is decreased.

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