



## Three-phase modelling of electrochemical chloride removal from corroded steel-reinforced concrete



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### HIGHLIGHTS

- This paper proposed a three-phase heterogeneous model to investigate ECR treatment.
- This paper highlighted a discussion of different configurations of multiple cathodes.
- The present model reveals some important features of aggregate effects during ECR.

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### ABSTRACT

Electrochemical chloride removal (ECR) is an efficient and curative way for treating reinforced concrete structures about to suffer or already suffering from chloride attack. In order to provide a deep understanding of the mechanisms of ECR, this paper presents a mesoscale numerical model, which considers the multi-species coupling in pore solution, electrochemical reactions taking place at electrodes, multi-species binding between solid and liquid phases, as well as the effect of applied current density and treatment time on the efficiency of chloride removal. The concrete presented in this model is treated as a heterogeneous composite material with three constituent parts, including not only mortar but also volume-controllable aggregates and interfacial transition zones; each as one phase. Through a further study on the different configurations of multiple reinforcing bars as cathodes, a quantitative discussion regarding the economic issue of different cathode configurations is highlighted. In addition, the present three-phase model reveals some important features of aggregate effects which could not be discovered from previous one-phase models.

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## 1. Introduction

It is well known that the penetration of ions, especially chloride ions, is the main reason causing the corrosion of reinforcing steel in concrete structures. Considering both the cost and efficiency of the rehabilitation methods, electrochemical chloride removal (ECR) or electrochemical chloride extraction (ECE) is a conventional and curative way for treating reinforced concrete structures about to suffer or already suffering from chloride attack. The repair method of ECR was first reported in the 1970s [1]. The idea involves mounting an external anode surrounded by a suitable liquid electrolyte

on the surface of the concrete and applying a high direct current density between the anode and the embedded reinforcing bar acting as a cathode. During a conveniently short time (usually a few weeks), most of the chlorides in the concrete are driven away from the reinforcement cathode into the external electrolyte near the anode due to the action of the applied current. As a result, the chloride-contaminated concrete can be eventually rehabilitated and its service life thus extended.

Since the invention of ECR in the 1970s, a large number of laboratory tests have been carried out to investigate the effectiveness and efficiency of the method as well as the possible side effect of the electrochemical treatment on the mechanical properties of concrete and the bond strength between reinforcement and concrete [2–25]. Apart from the experimental studies, significant progress has also been made in developing numerical models

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[28–40] to simulate the process of the treatment. Concrete is a heterogeneous material with complicated microstructure organisation. Experiments can provide important and valuable data but individual effects of various factors are difficult to determine because of the interactions between the different factors [26,27]. In contrast, numerical models have the great advantage in providing detailed information and investigating parametric effects. With the advance of computer science and the progress of computational mathematics, numerical modelling becomes more and more popular, particularly for problems of complicated geometries and those involving many physical parameters. The existing numerical models of ECR often focus on obtaining the chloride distribution profiles versus time and/or space under the influence of various factors (i.e. treatment time, current density, binding effect, additives, temperature etc.) by assuming the concrete as a one- [28–38] or two-dimensional single phase material [39,40]. In most of these studies, a single cathode was used, indicating that the reinforcing bars were isolated from each other in the structure, which may not be true in reality [18]. It is obvious that if the reinforcing bars are connected together, the situation would be quite different for the ECR treatment as short circuits are produced between steel electrodes, which will obviously have an impact on the chloride removal. This kind of impact has not been properly addressed in literature. Moreover, most of the existing models use the single phase model, i.e. assuming the concrete as a homogeneous porous material. However, recent studies showed that aggregates and the interfacial transition zone (ITZ), which is the area around the aggregate where there is a porosity gradient as the cement paste close to the aggregate is less porous than the rest of the cement paste, can have significant effects on the transport of ions [41], indicating that the concrete should be treated as a heterogeneous material of multiple phases. More recently, Liu and Shi [40] presented a numerical model of ECR, which demonstrated the importance of inclusion of aggregates in the simulation of ECR and reported the results of chloride removal when the cathode is selected at different positions. However, their work did not discuss or show how the current distribution affects the chloride removal.

In this paper, a two-dimensional mesoscale model is presented, which considers the multi-species coupling in pore solution, electrochemical reactions taking place at electrodes, multi-species binding between solid and liquid phases, as well as the effect of applied current density and treatment time on the efficiency of chloride removal. The concrete presented in this model is treated as a heterogeneous composite material with three constituent parts, namely mortar matrix, aggregates and ITZs, each as one phase. Through a further study on the different configurations of multiple reinforcing bars as cathodes, a quantitative discussion regarding the economic issue of different cathode configurations is highlighted. In addition, the present three-phase model reveals some important features of aggregate effects which could not be discovered from previous single phase models.

## 2. Theoretical background

The ECR treatment in the present study is applied to a two-dimensional piece of concrete, which is treated as a heterogeneous composite material of three constituent parts, namely mortar matrix, aggregates and ITZs, each as one phase. Since the aggregates are much denser and have much higher resistance to ionic penetration than the other two parts, they are assumed to be impermeable. Correspondingly, the other two parts, mortar and ITZs are assumed to be two independent porous materials, in which ionic transport is taking place.

Assume that the porous material in ITZs and mortar phases is saturated, the pore solution in these two phases is the ideal and

dilute solution, and there are no chemical reactions involved between ionic species. In this case the transport of ionic species can be expressed in terms of the mass conservation and current conservation as follows,

$$\frac{\partial C_k}{\partial t} = -\nabla J_k \quad k = 1, \dots, n \quad (1)$$

$$I = F \sum_{k=1}^n z_k J_k \quad (2)$$

where  $C_k$  is the concentration,  $z_k$  is the charge number,  $J_k$  is the flux,  $t$  is the time,  $I$  is the current density,  $F = 9.648 \times 10^{-4} \text{ C mol}^{-1}$  is the Faraday constant, subscript  $k$  represents the  $k$ -th species, and  $n$  is the total number of the species contained in the mortar.

Diffusion and migration are considered as the dominated parts of ionic transport in this study; therefore, the flux of an ionic species can be described using Nernst-Planck equation as follows,

$$J_k = -D_k \nabla C_k - D_k C_k \frac{z_k F}{RT} \nabla \Phi \quad k = 1, \dots, n \quad (3)$$

where  $D_k$  is the diffusion coefficient,  $R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$  is the ideal gas constant,  $T = 298 \text{ K}$  is the absolute temperature, and  $\Phi$  is the electrostatic potential. Substituting Eq. (3) into (1) and (2), yields,

$$\frac{\partial C_k}{\partial t} = D_k \nabla^2 C_k + \nabla \left[ z_k D_k \left( \frac{F}{RT} \nabla \Phi \right) \right] C_k \quad k = 1, \dots, n \quad (4)$$

$$\frac{F}{RT} \nabla \Phi = - \frac{(I/F) + \sum_{k=1}^n z_k D_k \nabla C_k}{\sum_{k=1}^n z_k^2 D_k C_k} \quad (5)$$

Since the present simulation employs a 2D numerical concrete model, the current density term in Eq. (5) need be expressed in terms of its two components in a rectangular coordinate system when solving for the electrostatic potential, i.e.,

$$\frac{F}{RT} \frac{\partial \Phi}{\partial x} = - \frac{(I_x/F) + \sum_{k=1}^n z_k D_k \frac{\partial C_k}{\partial x}}{\sum_{k=1}^n z_k^2 D_k C_k} \quad (6)$$

$$\frac{F}{RT} \frac{\partial \Phi}{\partial y} = - \frac{(I_y/F) + \sum_{k=1}^n z_k D_k \frac{\partial C_k}{\partial y}}{\sum_{k=1}^n z_k^2 D_k C_k} \quad (7)$$

where  $I_x$  and  $I_y$  are the components of the current density in  $x$ - and  $y$ -directions, respectively. Since the current density satisfies  $\nabla \cdot I = 0$ ,  $I_x$  and  $I_y$  can be calculated by solving the following Laplace equation,

$$\nabla^2 \psi = \frac{\partial^2 \psi}{\partial x^2} + \frac{\partial^2 \psi}{\partial y^2} = 0 \quad (8)$$

where  $I_x = \frac{\partial \psi}{\partial x}$  and  $I_y = \frac{\partial \psi}{\partial y}$ . The boundary conditions of  $\psi$  is that, at the cathode its normal derivative equals to the externally applied current density, that is  $\frac{\partial \psi}{\partial n} = I_\Omega$ , where  $\Omega$  represents the cathode boundary, while at the anode its function value equals to zero, which enforces the conservation of current between the inward at anode and the outward at cathode.

Note that, for concrete ionic transport takes place in the pore solution and during the course of the transport of ions, some of them may be bounded to the pore surface both physically and chemically. To take into account the ionic binding, Eqs. (1) and (4) need to be modified as follows,

$$\frac{\partial C_k}{\partial t} + \frac{\partial S_k}{\partial t} = -\nabla J_k \quad k = 1, \dots, n \quad (9)$$

$$\frac{\partial C_k}{\partial t} + \frac{\partial S_k}{\partial t} = D_k \nabla^2 C_k + \nabla \left[ z_k D_k \left( \frac{F}{RT} \nabla \Phi \right) C_k \right] \quad k = 1, \dots, n \quad (10)$$

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