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Multi-phase modelling of electrochemical rehabilitation for ASR and chloride affected concrete composites

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ABSTRACT

Reinforced concrete structures experience severe durability degradation when subjected to alkali–silica reaction (ASR) and chloride attack. A special electrochemical rehabilitation treatment, containing lithium compound anolyte, has been developed to drive lithium ions into concrete composites and remove chlorides simultaneously, for mitigating both the ASR-induced cracks and the chloride-induced reinforcing steel-bar/fiber corrosion. In this study, the efficiency of the impregnation of lithium ions and the removal of chloride ions through a specific electrochemical treatment is numerically evaluated, which results into the distribution profiles of all typical ionic species. A heterogeneous numerical model, which treats concrete as a three-phase composite, is presented to examine the response of inner structures, especially the interaction between active aggregates and lithium ions that are supposed to mitigate ASR. The ionic interaction between different species, binding and the electrochemical reaction at electrodes are also considered. Through a detailed modelling of multi-phase and multi-species, a systemic parametric analysis based on a series of significant factors during electrochemical treatment (e.g., current density, treatment time, temperature, cathode position and concentration of lithium solution) reveals some important tendencies of ionic electromigration in concrete structures, which are supposed to guide the field applications.

1. Introduction

Reinforced concrete (RC) structures experience severe durability degradation when subjected to chloride-induced reinforcing steel-bar/fiber corrosion and alkali–silica reaction (ASR). The former is due to the destruction of passive film surrounding a steel-rebar when adjacent chlorides reach a threshold concentration [1,2]. The latter is a chemical reaction between reactive siliceous compounds in aggregates and alkali ions in pore solution, producing a hygroscopic gel, which absorbs water from mortar and swells inducing cracks in the concrete [3–5]. As a result, the ASR affected concrete becomes more vulnerable to other deterioration mechanisms and leads to the destruction of concrete composites.

McCoy et al. [6] discovered the benefits of adding lithium based admixtures to suppress ASR expansion in fresh concrete. Lithium salts are known to alter the ASR by producing non-expansive gel or hindering the formation of the alkali–silica gel [7–9]. However, it is difficult to adopt the scheme of lithium based admixtures for the massive existing structures. A technique for driving lithiums into hardened concrete is necessary to prevent the ASR expansion. The available treatment methods against ASR of existing structures, such as submitting samples to immersion, spraying and vacuuming impregnation, are already investigated [10–12]. Limited research is available regarding the effective penetration depth and concentration of lithiums. In the treatment of highway barriers, Folliard et al. [11,12] showed that only the first 2–4 mm depth had obtained enough lithium to avoid ASR expansion through vacuum impregnation. The application of electrochemical treatment was considered as an effective mitigation method to cope with ASR [13,14].

Electrochemical rehabilitation techniques are originally developed for treating reinforced concrete structures that were about to suffer or already suffered from chloride attack [15–26]. Besides, these techniques are nowadays also used for driving lithium ions into concrete to avoid ASR [27]. During the migration process, not only the lithium ions

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are impregnated into concrete, but also at the same time chloride ions can be removed. The lithium ions impregnation will suppress ASR while chloride removal will mitigate the chloride-induced corrosion of reinforcing steel. This combined treatment of ELM & ECR (i.e., electrochemical lithium migration and electrochemical chloride removal) involves setting an auxiliary anode surrounded by a lithium-based electrolyte solution on the surface of the concrete and applying a direct current (DC) density between the anode and the embedded rebar acting as a cathode.

To investigate the feasibility and effectiveness of the ELM method, considerable efforts have been made on the evaluation of ionic transport, especially for lithium, by using experimental or numerical methods [12–14,28–37]. For example, Sergi et al. [28] utilized various lithium based electrolytes to investigate the concrete realkalisation which show that the impregnation of lithium is effective for specimens when the current density is 2 A/m^2. The experiments done by Farias et al. [30] indicate that migration of lithium is time-dependent and more effective when higher voltage was applied. Whitmore et al. [14] reported that the application of the electrochemical treatment to two bridge decks achieve better penetration and higher concentration of lithium in a relatively short time as compared to spraying and vacuuming impregnation. To assess the valid conditions of electrochemical treatment for achieving sufficient concentration and depth of lithium, the influence of temperature, treatment time, current density as well as the concentration of lithium solution are also considered. Liu et al. [38] observed that the time required for lithium ions to pass through the sample decreases with the increase of the average current density in a two-chamber set-up. Ueda et al. [39] evaluated the efficiency of the electrochemical treatment in two different concentrations of anolyte solution (LiOH), which suggest that the treatment with the higher concentration of anolyte solution is more effective than the lower. Recently, Ueda et al. [40,41] further reported the behavior of the lithium ions along with other ionic species in the concrete at different temperatures. Results showed that both the penetration of lithium ions and the removal of chloride ions are more effective when higher temperature is applied.

Notwithstanding the aforementioned experimental studies have made notable progress to investigate lithium migration processes in concrete composites, existing literature shows limited information in specifically describing the mechanism of ionic transport. Besides, only one or two factors (e.g., temperature, treatment time, current density, concentration of lithium solution, etc.) are evaluated in a single experiment due to cost, time and system power consumption considerations. In contrast, the numerical methods have greater advantages in investigating combined effects and providing more visual information. The existing numerical models developed for simulating chloride migration during ECR treatment [17,20,42–50], to some extent, are also incapable of assisting in providing more accurate predictions on the lithium migration, or the binary actions of ELM & ECR. In the earlier ECR models, the concrete was generally assumed as a homogeneous medium. A series of one-dimension transport models had been established to simulate the penetration process of chlorides [51,52] and lithiums [35]. Wang et al. [53] developed a two-dimension numerical model to simulate ionic migration which concluded that the amount of removal chlorides increase with the increase of current density, while Wang's model still has only one phase. Regarding the aggregate effects on ionic transport, Shi et al. [15] and Šavija et al. [54] developed two-phase finite element analysis (FEA) models to investigate the ionic transport during ECR and Rapid Chloride Migration (RCM) test, respectively, which suggested that the aggregates cause some effects on chlorides migration. Particularly, since the lithiums can prevent ASR by reacting with aggregates (or ASR-gel around aggregates), the significance of considering aggregate phase becomes even greater when study ELM. In order to mitigate the binary deterioration caused by chloride attack and ASR, a multi-species transport model is appreciated to reflect the performances of both lithium and chloride ions during the electrochemical rehabilitation treatment. Recently, some numerical researches [20,21,47,55,56] have utilized Poisson-Nernst-Planck equations to investigate the electrochemical coupling phenomena between multi-species. Note that the hydroxide ions produced at electrodes, which would also influence the distribution of lithium and chloride ions, are difficult to be represented in above multi-species models involving boundary conditions of electrical potential. Apart from aforementioned two key factors, multi-phase and multi-species, there are still some secondary factors influencing ionic transport. For example, the binding effect will slow down the ionic transport in concretes, not only for chlorides [17,21,57], but also for lithiums [35]. The modeling work done by Samson et al. [48] showed that the temperature plays an important role in the diffusion coefficient of all ionic species. Xia et al. [20] reported that the boundary and initial conditions also have some effects on the ionic distribution profiles.

The abovementioned literature review shows that, there have been a set number of numerical studies on ionic transport in concrete composites, however, very limited work has been paid on the interaction of different factors on the transport of each individual ionic species. The accurate electro-chemical-physical mechanism of ELM & ECR is not fully understood. In this study, the efficiency of the impregnation of lithium ions and simultaneously the removal of chloride ions through a specific electrochemical treatment are numerically evaluated, which results into the distribution profiles of all typical ionic species (e.g., potassium, sodium, chloride, hydroxide and lithium). A heterogeneous numerical model, which treats concrete as a three-phase composite, is presented to examine the response of inner structures, especially on the interaction between active aggregates and lithium ions that are supposed to mitigate ASR. The ionic interaction between different species, binding and the electrochemical reaction at electrodes are also considered. Based on the proposed multi-phase and multi-species modelling, a series of significant factors during the electrochemical treatment, i.e., the current density, treatment time, temperature and concentration of lithium-based electrolyte, are taken into account for their effects on the migration of lithium ions and the extraction of chlorides. In addition, a further study regarding the efficiency of different cathode positions is discussed. Such a systemic numerical study will reduce the amount of experimental work while providing valuable insights for transport mechanism of different ions during ECR & ELM.

2. Basic equations

The pore solution in concrete composites not only involves ionic species like hydroxide, sodium, potassium and calcium ions but also harmful ions like chloride and sulphate. The transport of these ionic species is dominated by two major driving forces, i.e. diffusion and migration. Thus the flux of an ionic species can be expressed using Nernst-Planck equation as follows:

$$J_k = -D_k \nabla C_k - D_k C_k \frac{z_k F}{RT} \nabla \Phi, \; k = 1, 2, \ldots, n \tag{1}$$

where J_k, C_k, D_k, Z_k represent the flux, concentration, diffusion coefficient and charge number of the k-th ionic species in mortar matrix, respectively. $F = 9.648 \times 10^4 \text{C mol}^{-1}$ is the Faraday constant, $R = 8.314\,\text{J mol}^{-1}\,\text{K}^{-1}$ is the ideal gas constant, $T = 298.15\,\text{K}$ is the absolute temperature, Φ is the electrostatic potential, and n represents the total number of the species involved in the pore solution.

Assuming that the concrete composites is a saturated pore medium, the pore solution is an ideal dilute solution and there are no chemical reactions between ionic species, the mass balance of species k can be expressed as:

$$\frac{\partial C_k}{\partial t} = -\nabla \mathrm{J}_k, \; k = 1, 2, \ldots, n \tag{2}$$

Substituting Eq. (1) into Eq. (2) yields,

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