A Numerical study for Chloride Migration in Concrete Under Electrochemical Repair Process

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Abstract: Electrochemical repair is one of the most effective methods of removing chloride from offshore engineering structures to improve their durability. The diffusion of chloride and electrochemical repair are key stages in this process. In this paper, a new numerical chloride migration-diffusion model is established by using Nernst-Planck equation. The distributions of free chloride ions in concrete in the stage of free chloride diffusion and the stage of electrochemical repair are simulated, and the effects of external potential and cathode position on electrochemical repair efficiency are analyzed. The numerical results show that electrochemical repair can effectively remove chloride ions from concrete. After 12 weeks of the electrochemical repair, the electrochemical repair efficiency is more than 66%. With the increase of repair time, the increase amplitude of electrochemical repair efficiency decreases gradually. In the initial stage of electrochemical repair, the chloride concentration on reinforcing bar surface decreases rapidly. After 4 weeks of electrochemical repair, the chloride concentration tends to be stable. As external potential increases, the electrochemical repair efficiency is also improved, and the chloride concentration on reinforcing bar surface decreases rapidly. When the cathodes are set on both sides of the concrete, the electrochemical repair time should be extended, and external potential should be increased to achieve better repair effect.

Key words: Electrochemical repair; Numerical simulation; Chloride Migration; Concrete Durability

1 Introduction

In offshore engineering structures, chloride is one of the main factors causing reinforcing bar corrosion (Arya, Vassie and Bioubakhsh, 2014; Kumar, Kumar and Kujur, 2019; Wang, Gong and Wu, 2019). Electrochemical repair is one of effective methods to remove chloride in concrete and improve the durability of offshore engineering structures (Xia *et al.*, 2019). Electrochemical chloride removal (ECR) technique refers to the formation of potential difference between reinforcing bar (*i.e.*, cathode) and external electrolyte (*i.e.*, anode). Under the action of potential difference, chloride in concrete moves from low potential to high potential, so as to remove chloride in concrete (Shi *et al.*, 2012; Xia *et al.*, 2019; Xu and Li, 2019a).

ECR technique has been favored by many research scholars since the experimental investigation work was carried out in the 1970s (Garcés, Sánchez De Rojas and Climent, 2006; Miranda *et al.*, 2007; Sánchez and Alonso, 2011). Mao *et al.*(Mao *et al.*, 2015) studied the effect of external electric field on chloride removal in sea sand concrete. And the research indicated that external electric field could effectively remove chloride in sea sand concrete, and the durability of sea sand concrete could be greatly improved by migrating corrosion inhibitor into reinforcing bar surface. The study of Zhou *et al.* (Zhou *et al.*, 2020) manifested that external electric field could effectively remove chloride in concrete, but it also led to the migration and aggregation of a large number of K^+ and Na^+ ions to the arear near the reinforcing bar. Saraswathy *et al.* (Saraswathy, H.-S. Lee, *et al.*, 2018) investigated the effect of different anode types on chloride removal efficiency. Xia, J. *et al.* (Xia, Q. Liu, *et al.*, 2018) discussed the effect of ambient temperature on ECR efficiency. The study displayed that the total efficiency was doubled as the temperature increased from 0℃ to 20℃, and it was improved by 13% as the temperature reached 50℃, indicating that temperature rise was an effective method of improving ECR efficiency. However, concrete is a heterogenous material with complex microstructure. Due to the interaction between different factors, it's hard to determine the individual effects of various factors through experiments. Comparatively speaking, numerical models have great advantages in providing detailed information and studying research parameters.

With the progress of computer science and technology and the development of finite element numerical method, especially for complicated multi-field coupling problems, numerical simulation technique has been extensively applied. In the existing ECR numerical models (Li and Page, 1998; Toumi, François and Alvarado, 2007; Xia, Q. feng Liu, *et al.*, 2018), it's assumed that concrete was isotropic material, and Nernst-Planck equation was used to simulate the chloride removal process of reinforced concrete under the action of potential. The effects of treatment time, current density, binding effect, additive, and temperature on ECR efficiency were studied. Li, *et al.* (Li and Page, 2000) established a numerical electrochemical chloride removal model by using Nernst-Planck

equation, and solved the Nernst-Planck equation by employed finite element method. And the effects of external current density, treatment time, ion coefficient, ion binding, boundary condition and medium porosity on chloride extraction efficiency were studied. Xu, Jun et al. (Xu and Li, 2019b) established the fitting formulas of chloride removal efficiency of unilateral-anode concrete with current density and treatment time. Liu, Q., *et al.* (Liu *et al.*, 2015) discussed the effects of aggregate shape, boundary conditions and double electrode layers on ECR efficiency and ion concentration distribution.

Many researchers have conducted a large quantity of fundamental studies on ECR, providing a theoretical and experimental basis for follow-up ECR researches (Liu *et al.*, 2014). However, all of the abovementioned researches are based on uniform chloride distribution in concrete. While in practical engineering, the distribution of chloride in concrete is nonuniform. The concentration of chloride ions near the erosion surface is higher, while the chloride concentration away from erosion surface is lower. In this paper, the distribution of free chloride in practical engineering is simulated through two stages, *i.e.*, the diffusion of free chloride and electric migration. The external electric field is applied to remove chloride ions diffused into the concrete. The numerical electrochemical repair model with diffusion-electric migration is established, and the chloride distribution laws in the concrete are investigated.

2. Theoretical background

2.1 Chloride diffusion

In offshore engineering structures, chloride ions in the environment diffuse into the concrete due to the chloride concentration difference (Chen *et al.*, 2020, 2021). The studies of many researchers indicates that chloride diffusion conforms to Fick's second Law, and the chloride diffusion equation in the concrete is as follow (DHIR, JONES and MCCARTHY, 1993; Jin *et al.*, 2015; Zhu and Zi, 2017; Mazer, Lima and Medeiros-Junior, 2018; Wang *et al.*, 2018):

$$\frac{\partial C}{\partial t} = \nabla \left(D_{cl} \nabla C \right) \tag{1}$$

where t is the erosion time of chloride, C is the chloride concentration (by weight of concrete, %), and D_{cl} is chloride diffusion coefficient, respectively. It can be seen from Eq. (1) that D_{cl} is a decisive factor for chloride concentration in concrete. The chloride ion diffusion coefficient not only relates to the concrete itself (such as concrete type, pore structure, water-binder ratio, hydration degree and so on), but also relates to the external environment, such as temperature (Bažant and Najjar, 1972), humidity (Muthulingam and Rao, 2014), maintenance age (Martín-Pérez, Pantazopoulou and Thomas, 2001) and other factors. In this paper, only the effects of water-binder ratio, temperature, humidity, and age are considered in the chloride diffusion coefficient. And the

chloride diffusion coefficient can be expressed as (Jin *et al.*, 2015; Wang *et al.*, 2018; Chen *et al.*, 2020):

$$D_c = \frac{2 \times \varphi_p^{2.75} D_p}{\underbrace{\varphi_p^{1.75}(3-\varphi_p) + n(1-\varphi_p)^{2.75}}_{\text{Water-cement ratio}}} \cdot \underbrace{\exp\left[\frac{U_c}{R}\left(\frac{1}{T_{ref}} - \frac{1}{T}\right)\right]}_{\text{Temperature}} \cdot \underbrace{\left[1 + \frac{(1-h)^4}{(1-h_c)}\right]^{-1}}_{\text{Humidity}} \cdot \underbrace{\left(\frac{t_{ref}}{t}\right)^m}_{\text{Curing age}} \tag{2}$$

where U_c is the chloride diffusion activation energy ($44.6 KJ/mol$), t_{ref} is the reference exposure time (28 d), t is the chloride ion erosion time (s), T is the ambient temperature (K), R is the gas constant ($8.314 J/K \cdot mol$), T_{ref} is the reference temperature ($298 K$), h is the humidity and h_c the critical humidity (0.75). D_p is the chloride diffusion coefficient in water (1.07×10^{-10} m^2/s), n is the empirical parameter (14.44) (Du, Jin and Ma, 2014), m is the time attenuation index. And φ_p is the porosity of cement paste, which can be expressed as (Sun *et al.*, 2011)

$$\varphi_p = \frac{wc - 0.17\alpha}{wc + 0.32} \tag{3}$$

where α is the degree of hydration of cement paste, and wc is the ratio of water to binder.

2.2 Electrochemical migration

As is known, the concrete pore solution contains various ions. Under the action of external electric field, positively charged ions move towards low potential, and negatively charged ones move towards high potential (Liu *et al.*, 2015; Chen *et al.*, 2021). Assuming that the concrete pore solution is diluted solution and the internal ions do not react with each other, the ion concentration in the concrete under the action of electric field can be calculated according to Nernst-Planck equation. The material equilibrium and ion transfer are defined by combing electric neutrality condition (Xu and Li, 2019a; Chen *et al.*, 2021; Fang *et al.*, 2021).

$$\frac{\partial C_i}{\partial t} = \nabla\left(D_i \nabla C_i + z_i D_i \left(\frac{F}{RT} \nabla \phi\right) C_i\right) \tag{4}$$

$$I = F \sum_{i=1}^{n} z_i \left(-D_i \nabla C_i - z_i D_i \left(\frac{F}{RT} \nabla \phi\right) C_i\right) \tag{5}$$

$$\sum_{i=1}^{n} z_i C_i = 0 \tag{6}$$

where C_i, D_i, and z_i represent the concentration, diffusion coefficient and charge number of the *i*-th ionic species in concrete, respectively. $F=9.648 \times 10^4 C \cdot mol^{-1}$ is the Faraday constant, ϕ is the electric potential.

3 Numerical simulations

During concrete pouring process in the actual projects, the concrete pore solution contains various ions, mainly including K^+, Na^+, Ca^{2+} and OH^- ions. These ions are formed during the pouring

132 process and are uniform distribution in the concrete. The chloride content is very small and can be

133 ignored during the pouring process. However, during the service process of concrete, chloride ions

134 in the environment gradually penetrated concrete. The free chloride diffusion process is simulated

135 by Eq. (1), in which the external electric field is not applied. After chloride diffusion for a certain

136 time, an external electric field is applied to remove chloride ions from concrete. The ion

137 concentration distributions at different time are calculated by Eqs. (4) ~ (6).

138 Wu *et al*. (Wu, Li and Yu, 2017) performed on-situ chloride sampling and testing at 13# wharf

139 of Fangcheng Port, Beibu Gulf in Guangxi Province. The results showed that the chloride

140 concentration in the outmost layer was about 0.67%, which was converted into Molar concentration

141 of 471.8 mol/m^3. Therefore, the boundary concentration of chloride ions on the erosion surface is

142 taken as 471.8 mol/m^3 in this paper. In the 10th year after chloride ion free erosion, electrochemical

143 repair is started. The chloride concentration in the concrete is CL_{10}, and the chloride diffusion

144 coefficient is D_{cl10}. The boundary concentrations and diffusion coefficients of other ions can refer

145 to references shown in **Tab. 1**. **Fig. 1** is the schematic diagram of chloride attack on concrete and

146 electrochemical repair. The diameter of the reinforcing bar is 12 mm, the thickness of the concrete

147 cover is 60 mm, and the geometric dimension of concrete section is 150 mm x 150 mm, respectively.
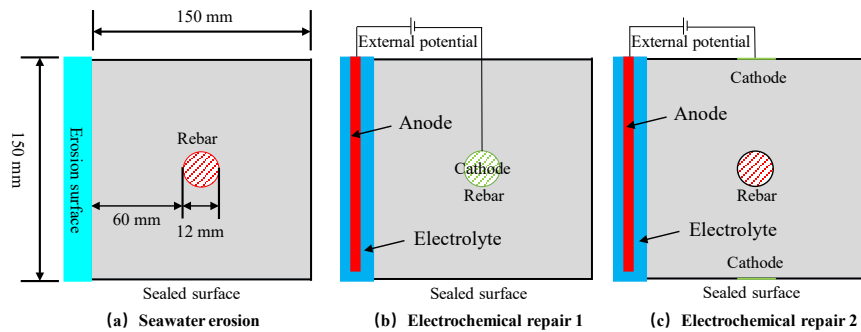
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(a) Seawater erosion   (b) Electrochemical repair 1   (c) Electrochemical repair 2

149 **Fig. 1.** Numerical simulation diagram.

150 **Table 1** Boundary conditions, initial values, and diffusion coefficients of various ion (Wang, Li and Page, 2001;

151 Liu *et al.*, 2012; Xia *et al.*, 2020).

Field variables	Position	K$^+$	Na$^+$	Ca^{2+}	OH$^-$	Cl$^-$
Charge number	--	+1	+1	+2	-1	-1
Boundary concentration (mol/m^3)	Anode	5	5	25.2	12.6	10
Flux boundary condition	Sealing	0	0	0	0	0
Initial concentration (mol/m^3)	Internal	100	Cl_{10}	Cl_{10}	100	0

Diffusion coefficient (m²/s)	--	3.9×10^{-11}	2.7×10^{-11}	1.6×10^{-11}	5.28×10^{-10}	D_{cl}

4. Result analysis and discussion

4.1 Chloride concentration distribution

(a) Before repair   (b) Repair 1 week   (c) Repair 4 weeks
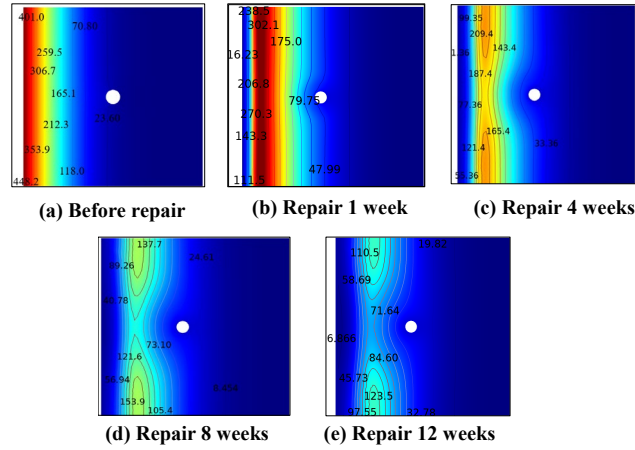
(d) Repair 8 weeks   (e) Repair 12 weeks

Fig. 2. Chloride concentration distribution cloud map.

Fig. 2a shows the free chloride concentration distribution in the 10th year of chloride diffusion. In the electrochemical repair process, the reinforcement bar is used as cathode, as shown in **Fig. 1b**. And the applied electric potential is 12 V. Additionally, the distributions of free chloride with different electrochemical repair time are shown in **Fig. 2b-e**. It can be seen from **Fig. 2a-e** that under the action of external electric field, chloride ions are removed from concrete. The chloride distributions in the concrete after electrochemical repair (as shown in **Fig. 2b-e**) are very different from that in the free diffusion stage (as shown in **Fig. 2a**). Compared with that before electrochemical repair (as shown in **Fig. 2a**), the chloride concentrations near the erosion surface (as shown in **Fig. 2b-e**)are lower after electrochemical repair. With the increase of erosion depth, chloride concentration first gradually increases and then decreases. Chloride ions migrate outward rapidly due to the applied low electric field around the reinforcing bar. With the increase of electrochemical repair time, the total chloride content in concrete gradually decreases, indicating that electrochemical repair is an effective method to remove chloride ions from concrete.
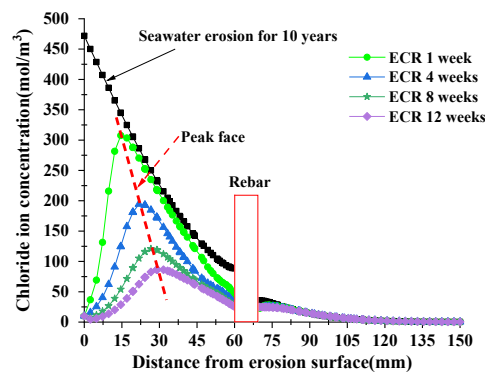
Fig. 3 shows the chloride concentration distribution curves in concrete at 1week, 4 weeks, 8 weeks, and 12 weeks of electrochemical repair. It is obvious from **Fig. 3** that the peak value of chloride concentration in concrete appear at 15 mm~30 mm away from the erosion surface. With the increase of the electrochemical repair time, the peak concentration of chloride gradually decreases and gradually moves towards the interior of concrete, which indicates that chloride ions in concrete are continuously remove from concrete under the action of external electric field. After 12 weeks of electrochemical repair, the peak value of chloride concentration decreases to 86.55 mol/m^3. Additionally, the chloride concentration near the erosion surface changes obviously, while the change of chloride concentration on the other side of erosion surface is not obvious. Similar conclusions were also found in the study of Chen *et al* (Chen *et al.*, 2021).

4.2 Chloride concentration on reinforcing bar surface

For reinforced concrete, the chloride concentration on the reinforcing bar surface is one of critical factors influencing corrosion of reinforcing bar. The chloride concentration on the reinforcing bar surface under the action of external electric field will be analyzed in this section. It should be noted that the position and applied potential of the cathode are the same as those in Section 4.1. *i.e.,* the reinforcing bar is used as cathode and the applied electric potential is 12 V. **Fig. 4** shows the chloride concentration curves at three different points on the reinforcing bar surface. It is apparent from **Fig. 4** that the chloride concentrations at three positions decrease with the increase of electrochemical repair time. Moreover, in the first 14 days (14 d) of electrochemical repair, the chloride concentrations decrease sharply and then tended to be stable (about 24 mol/m^3). This indicates that the electrochemical repair efficiency cannot be improved by prolonging the repair time when the electrochemical repaired has been carried out for a certain time.
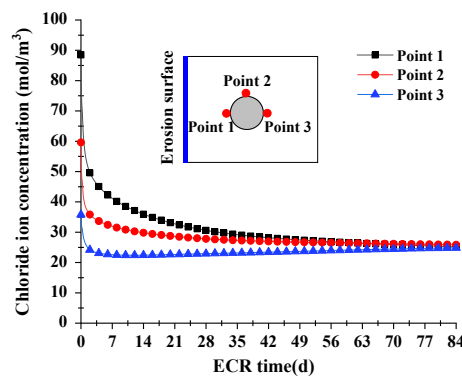

Fig. 4. Chloride concentration curves on the reinforcing bar surface.

4.3 ECR efficiency

Chloride diffusion is a very slow process, the total chloride content in the concrete has a great influence on the redistribution of chloride after the ECR. Therefore, the total chloride reduction rate

198 in concrete is employed in this paper as an evaluation index measuring the repair effect. The

199 calculation formula is as below:

$$P = \frac{M_0 - M_t}{M_0} \times 100\%$$ （7）

201 where P is the chloride ion reduction rate (%), M_0 is the total amount of initial chloride ion in

202 concrete (mol), M_t is the total concentration of chloride ion in concrete at the time of

203 electrochemical repair (mol).

204   The reinforcing bar is used as cathode and the applied electric potential is 12 V **Fig. 5** shows

205 the histogram of chloride removal rate in concrete at different repair time. It is obvious from **Fig. 5.**

206 that at 1 week, 4 weeks, 8 weeks and 12 weeks of the electrochemical repair, the chloride removal

207 rates are 24%, 45%, 58% and 66%, respectively. This indicates that the electrochemical repair could

208 effectively remove chloride ions in concrete. Meanwhile, with the increase of repair time, the

209 increase amplitude of chloride removal rate gradually decreases. Similar phenomenon has also been

210 found by Saraswathy *et al*. (Saraswathy *et al*. 2017) and Chen et al. (*Chen et al.,* 2021).
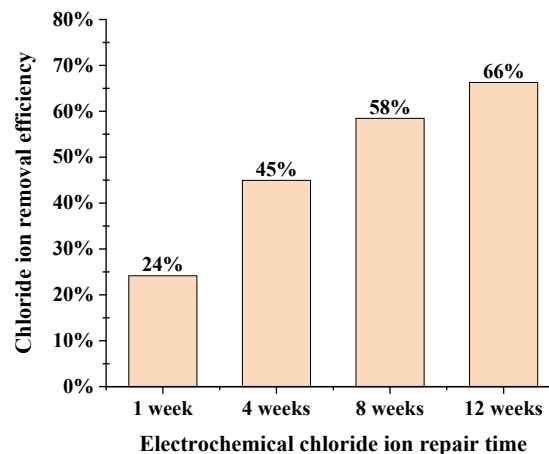

Fig. 5. ECR efficiency at different repair times.

4.4 Effect of different potentials on ECR

214   According to formula (4), the potential gradient is the main driving force for chloride migration.

215 Therefore, the potential difference between anode and cathode plays a very important role in

216 chloride removal rate. **Fig. 6a-c** shows the distributions of chloride concentration at 12 weeks of the

217 electrochemical repair under potentials of 5 V, 12 V and 24 V, respectively. It can be seen from **Fig.**

218 **6a-c** that the chloride concentration in concrete gradually decreases with the increase of potential.

219 For different potentials, the overall chloride concentration are different, while the chloride

220 distribution trend in concrete doesn't changed significantly. It is obvious from that **Fig. 6a-c** two

221 chloride concentration peak points are formed on the side near the erosion surface. Through

222 comparing **Fig. 6a**, **b** and **c**, it is apparent that the chloride concentration difference between **Fig. 6a**

223 and **b** is very large, while the chloride concentration difference between **Fig. 6b** and **c** is relatively

224 small. This indicates that as the potential increase, the influence of potential on chloride
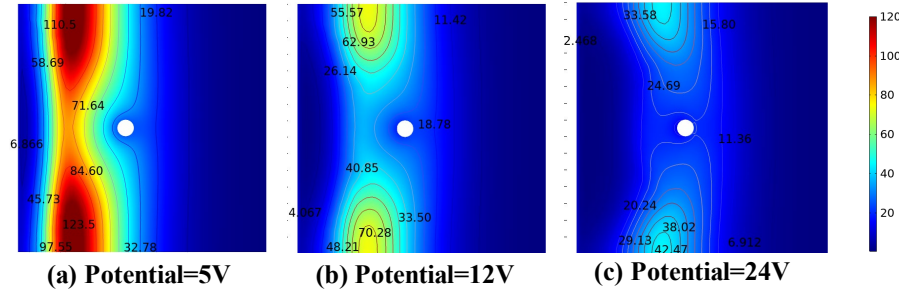
225 concentration distribution decreases.

(a) Potential=5V   **(b) Potential=12V**   **(c) Potential=24V**

226

227 **Fig. 6.** Cloud images of different potential potentials.

228   **Fig. 7** shows the chloride removal rate histograms at 1 week, 4 weeks, 8 weeks, and 12 weeks

229 of electrochemical repair under different potentials. It can be seen from **Fig. 7** that increasing

230 potential can effectively improve the chloride removal rate. However, too high potential may cause

231 hydrogen evolution reaction on the reinforcing bar surface and make the reinforcing bar more fragile

232 (Mao *et al.*, 2019; Chen *et al.*, 2021). At 12 weeks of the electrochemical repair, the chloride removal

233 rates under external potentials of 5 V, 12 V and 24 V are 66%, 81% and 88%, respectively.,

234 respectively. Compared with the electrochemical repair time of 8 weeks, the chloride removal rates

235 under external potentials of 5 V, 12 V and 24 V at the electrochemical repair time of 12 weeks

236 increase by 8%, 6% and 3%, respectively. While compared with the electrochemical repair time of

237 4 weeks, the chloride removal rates under external potentials of 5 V, 12 V and 24 V at the

238 electrochemical repair time of 8 weeks increase by 3%, 13% and 10%, respectively. These results

239 indicate that the increase of ECR efficiency decreases gradually with the increase of electrochemical

240 repair time. The chloride migration is mainly driven by electric migration in the early stage of

241 electrochemical repair, while it is mainly driven by the concentration difference in later stage of
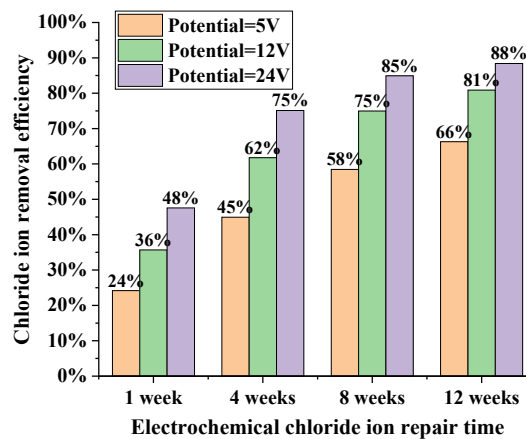
242 electrochemical repair.

243

4.5 Effect of cathode location on ECR

According to the above analysis, the electrochemical repair can effectively remove chloride ions in concrete. However, many studies show that ECR will not only eliminate harmful chloride ions, but also have adverse effects on reinforced concrete structure, such as reducing the bonding strength between reinforcing bar and concrete (Nguyen *et al.*, 2021), making the reinforcing bar more fragile (Saraswathy, H. S. Lee, *et al.*, 2018). Meanwhile, Na^+ and K^+ ions in the concrete solution will also aggregate near the reinforcing bar, causing concrete ASR harm. In this section, the chloride distributions in concrete and the ECR efficiency are analyzed when the reinforcing bar is used as cathode and both sides of concrete are used as cathodes. **Fig. 8a-b** show the chloride distributions in concrete at 8 weeks of electrochemical repair. It is apparent from **Fig. 8a-b** that there is great difference in the chloride distributions in concrete with these two cases. Generally, the reinforcing bar is taken as cathode, as is shown in **Fig. 8a**. And the chloride ion concentration near the reinforcement is the lowest, and two peaks of chloride ion concentration are formed in the concrete. When both sides of concrete are used as cathodes, chloride ions move from both sides of the concrete (*i.e.*, cathodes) to the erosion surface (*i.e.*, anode), as is shown in **Fig. 8b**. This results in the lowest chloride concentration on both sides of the concrete (*i.e.*, cathodes) and the highest chloride concentration near the surface of the reinforcement, as is shown in **Fig. 8b**. And a chloride ion concentration peak appears near the reinforcement surface in concrete. Moreover, by comparing **Fig. 8a** and **b**, the peak values of the two cases are almost the same, *i.e.*, about 100 mol/m³.

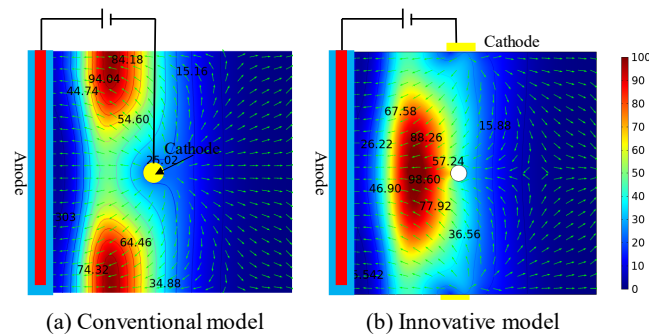(a) Conventional model   (b) Innovative model

Fig. 8. Chloride distributions in concrete with different cathode positions at 8 weeks of electrochemical repair. (a) Reinforcing bar is used as cathode, and (b   both sides of concrete are used cathodes.

Fig. 9 shows the distributions of chloride ion concentration on the surface of reinforcement with erosion time when reinforcement is used as cathode and both sides of concrete are used as cathode, respectively. It can be seen from **Fig. 9** that under the same external potential of 12 V, the chloride concentration on the reinforcing bar surface in the model with cathodes on both sides of concrete (red dot curve) is much higher than that with cathode on the reinforcing bar (black square

curve). Moreover, in the model with reinforcing bar as cathode, the chloride concentration on the surface of reinforcing bar (black square curve) decreases rapidly and tends to be stable after 14 d of electrochemical repair. However, for the model with both sides of concrete as cathodes, the chloride concentration (red dot curve) decreases slowly with electrochemical repair time. Additionally, the chloride concentration curves with both sides of concrete as cathodes under external potentials of 12 V, 24 V and 36 V are also depicted in **Fig. 9**. It is obvious from **Fig. 9** that the chloride concentration on the reinforcing bar surface can be effectively reduced by increasing the external potential. However, the overall electrochemical repair effect in the model with cathodes on both sides of concrete is still worse than that in the model with reinforcing bar served as cathode. Nevertheless, from the curve declining trend analysis, the model in which the two sides of concrete served as the cathode can achieve better chloride removal effect with the increase of electrochemical repair time.
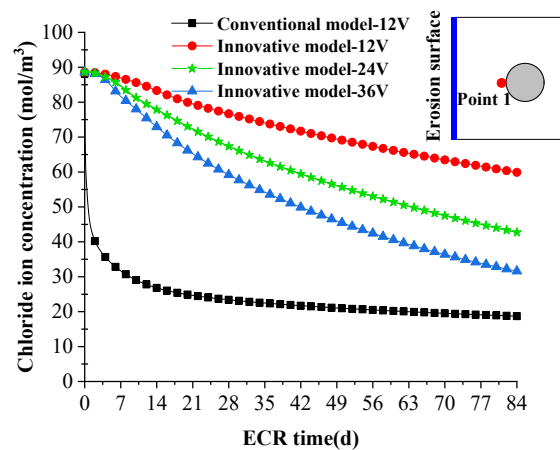

Fig. 9. Chloride ion concentration curves on the surface of steel bars for different repair models.

5 Conclusions

In this paper, a numerical chloride migration-diffusion model is established by Nernst-Planck equation to simulate the chloride erosion process in the external environment and the electrochemical repair process. Based on a systematic study, the following conclusions can be drawn:

(1) Electrochemical repair changes the chloride distribution in concrete. After chloride ions in the external environment diffuse into the concrete, the chloride ions in the concrete can be effectively removed by electrochemical repair. After electrochemical repair, the chloride concentration peak appears at 15 mm ~ 30 mm away from the erosion surface. With the increase of electrochemical repair time, the peak value of chloride concentrate gradually decreases, and the peak of chloride concentrate moves toward the interior of concrete.

(2) Electrochemical repair can rapidly reduce the chloride concentration on the reinforcing bar surface. The chloride concentration on the reinforcing bar surface decreases sharply in the beginning of electrochemical repair and tends to be stable after 14 d of electrochemical repair. Furthermore, with the increase of external potential, the stabilization time is shortened.

301   (3) Electrochemical repair can effectively reduce the chloride concentration in concrete. When

302   the ratio of chloride ion removal in concrete to the total amount of initial chloride ion is used to

303   measure the ECR efficiency, the ECR efficiency can reach more than 60% after 12 weeks of

304   electrochemical repair. Moreover, the ECR efficiency increases with the increase of external

305   potential.

306   (4) When the cathodes are set on both sides of concrete, the ECR efficiency can be improved

307   by increasing the external potential and lengthening the repair time. From a comparison between

308   the situation with cathodes on both sides of concrete and that with cathode on the reinforcing bar

309   surface, it can be found that there is a great difference in chloride concentration after the

310   electrochemical repair. Moreover, the decreasing curves of chloride concentration on the surface of

311   reinforcement is also different. However, the ECR efficiency with cathodes on both sides of concrete

312   can be improved by prolonging the repair time and increasing the external potential.

313

314 **Author Contributions:**

315 **Xuandong Chen**: Conceptualization, Methodology, Writing- Original draft preparation, Software,

316 Investigation. **Qing Zhang:** Writing-reviewing and Editing, Methodology, conceptualization,

317 Supervision. **Feng Fu**: Data curation, Writing- Reviewing and Editing. **Yang Ming**: Software, Data

318 curation. **Hua Rong**: Methodology, Software, Conceptualization.

319

327 **Conflicts of Interest:** The authors declare no conflict of interest.

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