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Effects of stirrups on electrochemical chloride removal efficiency

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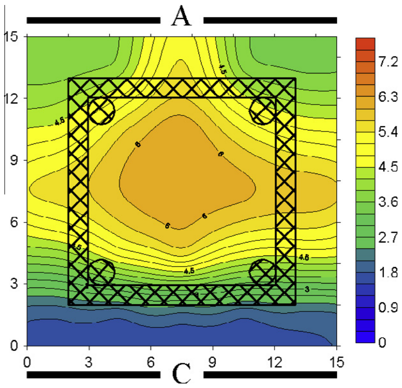
HIGHLIGHTS

- The existence of steel reinforce cage hinders the removal for chloride ions enclosed by it.
- The electrochemical signals showed that after ECR the corrosion tended to be retarded.
- The ECR may induce leaching of calcium ions inside concrete.

GRAPHICAL ABSTRACT

Chloride ions inside the cage did not move by the ECR process.



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ABSTRACT

In this paper, how the stirrups influence the efficiency of electrochemical chloride removal (ECR) is studied. The chloride removal efficiency was investigated by examining the chloride contents in concrete. In addition, the electrochemical signals for corrosion status for the rebars were recorded and analyzed. While the stirrups existed and formed a connected electric path with rebars, due to the fact that the electric potential for the steel rebar cage remains the same everywhere on the rebar cage the chloride enclosed by the steel rebar cage was difficult to be removed no matter how the electrodes were arranged. Although the chloride ions enclosed by the rebar cage were not easy to be removed, the electrochemical signals showed that after ECR the corrosion tended to be retarded.

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1. Introduction

It is well known that the existence of chloride ions in reinforced concrete structure or prestressed concrete structure influence the durability of the members. For example, concrete pavements constructed by concrete may suffer from deicing salt and serious corrosion happens while the chloride ions content is too high. The chloride ions will destroy the passive film which is formed in a very high alkaline pore solution and results in corrosion. Therefore, one needs to consider the effects of chloride ions before construction process and make a reasonable mix design to ensure the durability of the concrete structures. An overall review article about the durability of steel reinforced concrete structures can be found in [1].

Once the chloride ions content is found to be too high, one may remove unsound concrete and recast repair materials or one can consider the electrochemical chloride removal process. Assessments and guidelines for treatment were issued following the Strategic Highways Research Program (SHRP in the USA)—such as

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SHRP-S-347 and SHRP-C-620. These can be downloaded from the web. In addition, the processes were the subject of several patents which formed the basis of the "Norcure" processes that have been fairly widely known and used in USA, Europe and Japan.

The idea of ECR involves mounting an anode surrounded by a liquid electrolyte (usually NaOH or Na_3BO_3) on the surface of concrete and driving direct current into the embedded reinforcement, which acts as a cathode. (The usual electrolyte is calcium hydroxide provided as a saturated solution by mixing cellulose pulp with solid hydrated lime. This captures some fugitive chlorine and also prevents acid etching of the concrete surface). The current pushes chloride ions away from the reinforcement and extracts them towards the anode. Once reaching the concrete surface, the ions eventually pass into the anolyte and are thereby removed from concrete.

There exist numerous papers about ECR, the followings are some of them.

Garcés et al. [2] studied the effects of bar arrangements on the ECR efficiency. Five different types of bar arrangements were considered, corresponding to typical structural members such as columns (with single and double bar reinforcing), slabs, beams and footings. They concluded that the ECR efficiency was influenced by the type of bar arrangement and a uniform layer set-up favors chloride extraction.

Hassanein et al. [3] reported that factors that affected the short-term chloride removal efficiency included the resistivity of concrete, charge passed, treatment duration, initial chloride content, concrete cover, and chloride diffusion coefficient. Among these factors, the resistivity of concrete, charge passed and chloride diffusion coefficient were influenced by the water/cement (or water/binder) ratio.

Yeih et al. [4] studied the influence of the polarization parameter (defined as the desalination current density times the duration of ECR) on the ECR efficiency. They reported that as this parameter increased the chloride ion content inside concrete decreased.

Elsener and Angst [5] published a paper to discuss the mechanism of ECR. They have found that due to the removal of free chloride during the treatment, bound chloride is dissolved in order to re-establish the equilibrium between bound and free chlorides. The rate of release of bound chloride is slow compared to the rate of chloride removal and thus the ECR process quickly becomes inefficient. Current off periods allow the system to re-establish the equilibrium between bound and free chlorides. Subsequently, the process is efficient again.

Wang, Li and Page [6] used the mathematical model and numerical method to study the ECR for a 2-D member. Toumi, François and Alvarado [7] used numerical method to simulate ECR and compared results with experimental works.

Herrera et al. [8] studied the efficiency of ECR for various C_3A content, they concluded that ECE efficiency was slightly affected by C_3A because only a part of the bound chloride ions was released.

Orellan et al. [9] reported that after treatment, new cementitious phases containing rich concentrations of sodium, aluminum and potassium were formed. Moreover, alkali–silica gel was observed. They have concluded that the ECE accumulates locally high amounts of alkali ions that stimulate the alkali–silica reaction even though the concrete contained nominally inert siliceous aggregates.

Siegwart et al. [10] reported that the ECR process would result in hydrogen embrittlement thus was not suitable for prestressed concrete. They concluded that the risk of hydrogen induced brittle fracture due to electrochemical chloride extraction cannot be altered with modification of the treatment parameters, such as current density or treatment duration.

Fajardo et al. [11] reported that after ECR, about 60% to 50% of the initial chloride was removed from the concrete on average. Around 1% chloride by mass of cement remained around the steel after treatment. They also claimed that although both the chloride content and the dissolution of the steel were reduced, the repassivation of steel rebar cannot be guaranteed.

Pérez et al. [12] studied the ECR efficiency by using the conducting cement as the anode. They found that the thickness of the conductive cement paste anode has a great influence on the capacity of the anode to retain an important part of the extracted chlorides after finishing the electrochemical treatments.

Siegwart et al. [13] have reported that the pore size and pore size distribution of concrete are altered due to ECR and small pores hinder the migration of ions, which may partially be responsible for changes in concrete resistance.

Ihekwaba et al. [14] also reported that the ECR current reduced the concrete compressive strength, especially for the concrete near the cathode. By investigating the rehabilitation of several vertical structures, Ihekwaba et al. [15] concluded that circular columns containing spiral reinforcements showed better ECR performance than structures with planar surfaces. They [16] also reported that a pullout bond degradation of steel rebars in ECR concrete with a maximum decrease of 44% bond degradation was found.

Cañón et al. [17] found the sprayed conductive graphite powder–cement paste as anode not only provides electrochemical chloride removal with similar efficiency, but also is able to retain moisture even without the use of a continuous dampening system.

Miranda et al. [18] reported that if ECR is applied preventively it is an efficient procedure for delaying the start of corrosion. However, if applied too late it does not assure the repassivation of corroded RCS and is therefore useless.

Arya et al. [19] studied the factors that influence ECR and they concluded that chloride removal increased with increasing applied potential, number of reinforcing bars at a particular depth and initial chloride content of the concrete. A greater percentage of chloride was removed from prisms where the thickness of the chloride bearing layer of concrete was less than the depth of cover to the reinforcement. Where the thickness of the chloride bearing layer exceeded the cover to the reinforcement, the use of an external cathode significantly increased the total amount of chloride removed.

In this study, the effect of stirrups on ECR is examined. When stirrups and rebars together form a reinforcement cage, theoretically speaking they form a connected electric current path and the electric potential should be the same. In such a case, whether

Table 1
Concrete mix design.

w/*c*	Water (kg/m^3)	Cement (kg/m^3)	Fine aggregate (kg/m^3)	Coarse aggregate (kg/m^3)	NaCl (kg/m^3)
0.5	196	393	662	1046	11.79

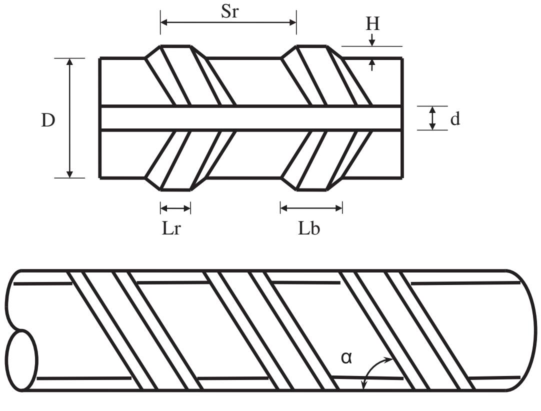
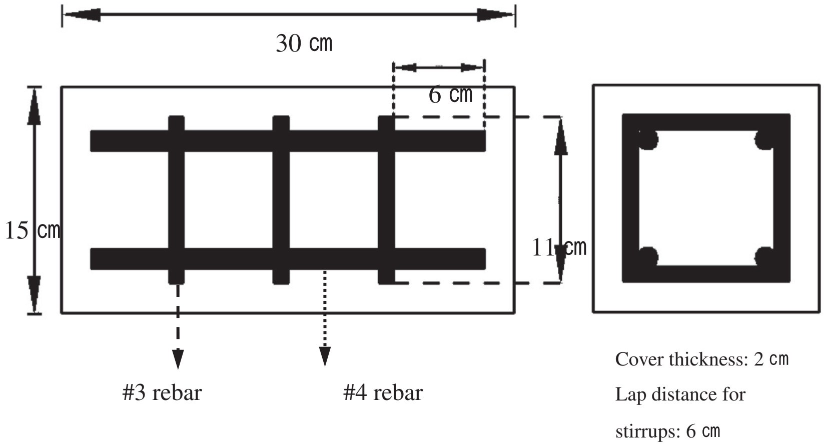


Fig. 1. Geometric diagrams of rebar.

Table 2
Geometric parameters for rebars.

Steel No.	Area (cm^2)	*D* (mm)	*d* (mm)	Lr (mm)	Lb (mm)	H (mm)	α (°)	Sr (mm)
No. 3	0.71	9.52	2.96	0.86	1.54	0.54	66	6.24
No. 4	1.29	12.30	3.99	1.36	2.24	0.74	66	8.23



Cover thickness: 2 cm
Lap distance for stirrups: 6 cm

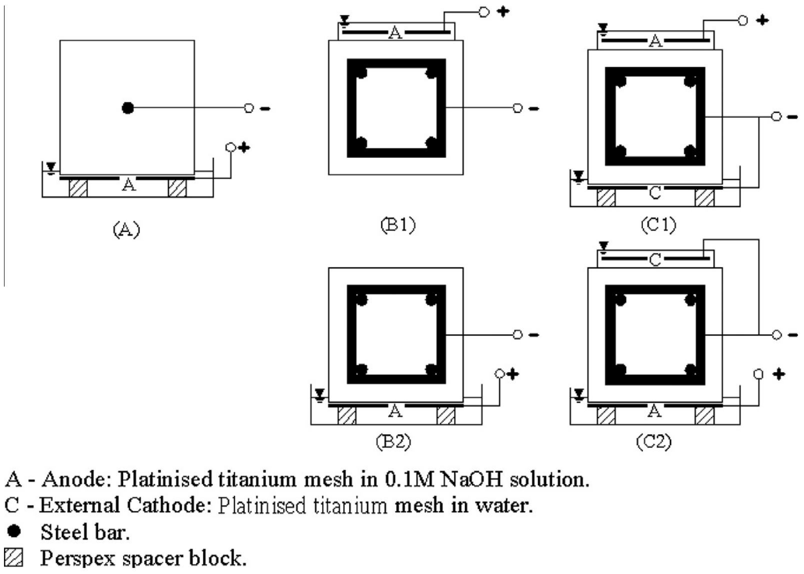
Fig. 2. Specimens with steel reinforcement cage.

or not ECR can remove chloride ions enclosed by the cage is investigated through experiments. If chloride ions enclosed by the cage cannot be effectively removed, whether or not ECR still can provide a remedy to rebar corrosion is also studied. Aside from this section, the details of materials, specimen preparations and experiments conducted will be given in Section 2. In Section 3, experimental results will be shown and some discussions will be drawn. In the final section, conclusions based on results we obtained will be given.

2. Experiments

2.1. Materials and specimen preparation

The concrete mix design is given in Table 1. Type I cement is used, the fine aggregate is river sand from local source and the properties are: fineness modulus is 2.65, SSD specific weight is 2.66 and water absorption is 2.98%. The coarse aggregate used is crush stone and the properties are listed as followings: fineness modulus is 6.36, SSD specific weight is 2.67, water absorption is 0.93% and the oven dry density is 1683 kg/m^3. The #4 rebars are used as the major reinforcements and #3 rebars are used as the stirrups for specimens. The geometrical diagrams for rebars are illustrated in Fig. 1 and the parameters are tabulated in Table 2. To simulate the contaminated concrete, an amount of NaCl for 3% cement weight is added in the concrete. Two kinds of specimens are prepared: the specimen for the first kind has a size of 15 cm × 15 cm × 30 cm with one #4 rebar locating at the center of the cross section; the specimen of the second kind has a size of 15 cm × 15 cm × 30 cm with four #4 rebars as major reinforcements and #3 rebars are used as stirrups, the lap distance of stirrups is 6 cm and the cover thickness is 2 cm. The diagram of specimens with rebar cage is shown in Fig. 2. The specimens were cast and demolded after 24 h (1 day) and then kept in saturated lime water curing environment for 27 days. After that, the age of concrete is 28-days. To conduct the ECR, the electrolyte solution used for anode cell is 0.1 M NaOH solution and for the group using additional cathode the electrolyte used for the additional cathode cell is water. The ECR lasted for 8 weeks, and for the ECR periods of 2, 4, 6 and 8 weeks various tests in Section 2.2 were carried out. For electrochemical corrosion signal measurements, they were performed after 2-week depolarization time to avoid misreading the meanings.



A - Anode: Platinised titanium mesh in 0.1M NaOH solution.
C - External Cathode: Platinised titanium mesh in water.
● Steel bar.
▨ Perspex spacer block.

Fig. 3. ECR setups.

The setup for ECR process is illustrated in Fig. 3. For setup A, only one rebar locating at the center and the anode cell is arranged beneath the specimen. This setup is used as the control specimen. The second and third setups are B1 and B2, in which the effects of steel reinforcement cage are in concern. The differences between B1 and B2 are the locations of anode cell. The fourth and fifth setup add additional cathode cells as shown in C1 and C2. Another additional setup, setup D, is to use C2 ECR setup for 4 weeks then use C1 setups for next 4 weeks. A constant voltage of 20 V was applied for ECR treatment. The current density of electrochemical chloride removal is suggested to be in the range from 1 to 10 A/m^2 to avoid possible side effects [20]. The constant voltage of 20 V is adopted according to our previous research [21].

2.2. Experiments conducted

2.2.1. Rebound strength

The rebound strength although is not reliable yet can provide a non-destructive test for evaluating the concrete surface soundness as well as the compressive strength. Since the specimens we used contained reinforcement cage such that the compressive strengths of the specimens cannot be carried out. Instead of using compressive strengths, we use rebound strengths to represent the mechanical properties of concrete. When each ECR period was fulfilled, the rebound strength tests were performed for 10 marked points as shown in Fig. 4 (red x marks). These marked points are at the centers for selected square region. The estimated concrete compressive strength then was obtained using the average of these data. In [22], the procedures for performing the rebound strength are given.

2.2.2. Chloride content

To measure the chloride content, the sampling method is first introduced in the followings. A 3-cm width slab was cut from the specimen as shown in Fig. 4 and 25 sampling points were taken as shown in Fig. 4. The chemical titration was then carried out to determine the water soluble chloride content inside the concrete. Using these 25 data for a specific ECR period, one can construct the equal chloride ion concentration contours and to see the efficiency of ECR. The contours are generated using the radial basis function approximation. The standard for measuring water soluble chloride ions can refer to ASTM C1218/C1218M-99.

2.2.3. pH values

It is known that the cathodic current will produce hydroxide ions near cathode and thus increases the pH value. The sampling method for measuring pH value is the same as mentioned above. A 3-g grounded sample which passed #50 sieve was prepared for each sampling point and it then is added into a 50 ml distilled water and heated this solution for 5 min. The container of the solution then was sealed by PVC film in order to avoid contamination and vaporization and it was placed for 24 h. The solution is then heated again for 15 min, we collected the solution passing through the filter paper. The solution was diluted into 1000 ml solution, the hydroxide ion concentration was determined by chemical titration method. Using this, the relative pH value for the original sample was obtained. It is worth mentioned here that the pH value we obtain here is not the pH value of the pore solution. The pH value we obtain here can be understood as the alkalinity the concrete may provide after ECR. If this value is low, it implies that the pH value of pore solution may be also low.

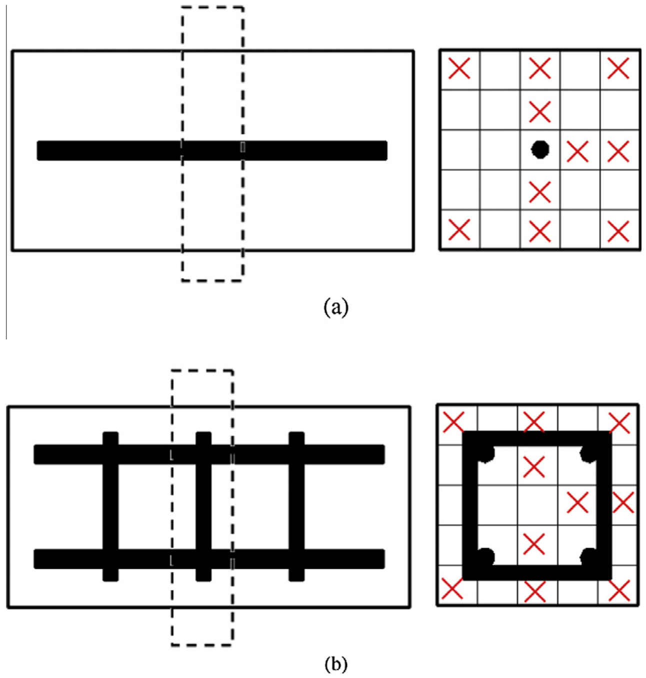


Fig. 4. Sampling methods for: (a) single rebar specimen; (b) steel reinforcement cage specimen. (X: locations for rebound strength tests).

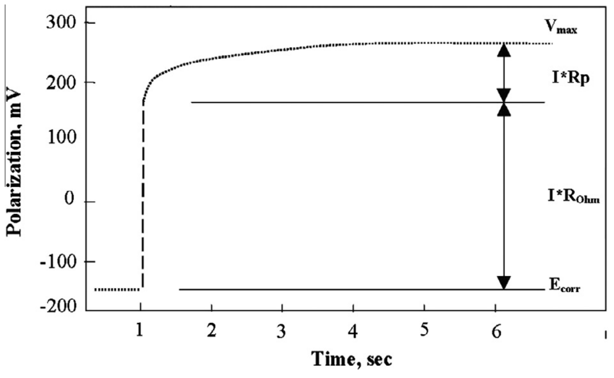


Fig. 5. A typical diagram for anodic polarization after the cutoff of current for GPM.

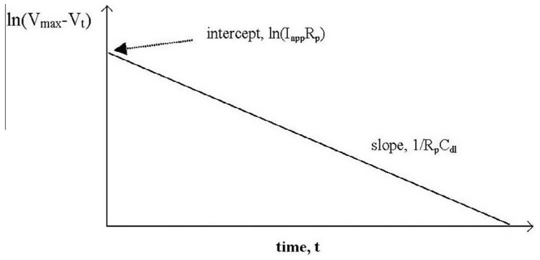


Fig. 6. The logarithm of V_{max}–V_t versus time.

Table 3
Relation between corrosion current density and corrosion status [26].

Corrosion current density (μA/cm^2)	Corrosion rate (mpy)	Corrosion status
<0.5	<0.23	Ignore
0.5–5	0.23–2.3	Low
5–10	2.3–4.6	Medium
10–15	4.6–6.0	High
>15	>6.9	Heavy

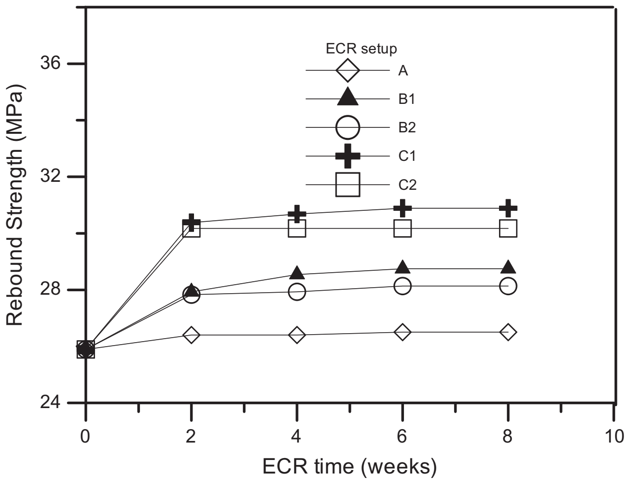


Fig. 7. Rebound strengths for various ECR setups.

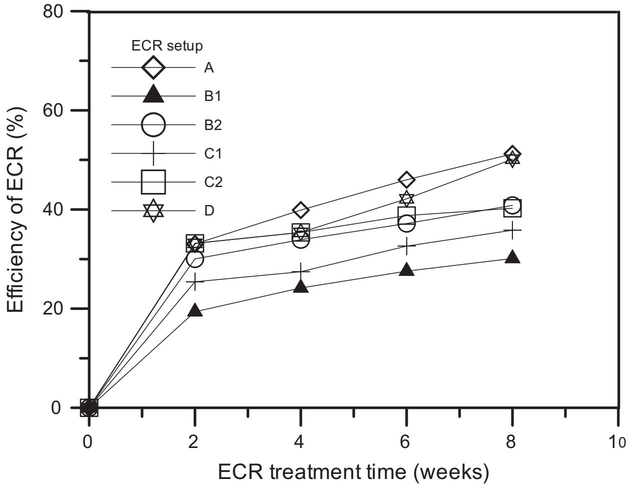


Fig. 8. ECR efficiencies for various setups.

2.2.4. Half cell potential

The half cell potential can provide thermodynamic trend for the rebar corrosion. The ASTM C876 provides a useful guide for conducting the half cell test. When each ECR period was fulfilled, the specimens then were disconnected first and 2-weeks depolarization time was required before measurement. Usually immediately after the disconnection of electric current, the half cell potential tends to a very active potential, which may lead us to make a judgment that the rebar suffers from corrosion. In order to avoid this, sufficient long enough time (at least 2-weeks depolarization time) is necessary before the measurement. Actually, the half cell potentials measured immediately after disconnection were in the region for $-950\ \mathrm{mV} \sim -1120\ \mathrm{mV}$ (CSE).

2.2.5. Instantaneous corrosion rate

The instantaneous corrosion rate was measured using the apparatus GPM-5000 manufactured by German Instruments. The GPM-5000 first measured the open circuit potential for the rebar. A pulse of current of this corrosion potential (usually 5–40 mA) was released for 10 s (suggested instrumental setup), the current was cut off. After cutoff, the measurement of potential could be carried out as shown in Fig. 5. The value for potential can be expressed by the following formula: [23]

$$V_t = I_{app}\left[R_p\left[1 - \exp(-t/R_pC_{dl})\right] + R_W\right] \tag{1}$$

where V_t (vol) is the potential at time t, I_{app} (μA/cm^2) is the difference between current densities, R_p (Ohm cm^2) is polarization resistance, C_{dl} (Columb/mol) is the double layer capacitance and R_W (Ohm cm^2) is the resistance from the surrounding environment. From Eq. (1), one can obtain

$$\ln(V_{max} - V_t) = \ln(I_{app}R_p) - t/(R_pC_{dl}) \tag{2}$$

where V_{max} is the steady-state potential.

Using the diagram as Fig. 6 and the least square method, the values of $\ln(I_{app}R_p)$ and (R_pC_{dl}) can be obtained. Since I_{app} is already known, it means that we can obtain the values of R_p and C_{dl}, respectively. After the value of R_p is known, the corrosion current density is calculated from the Stern–Geary formula:

$$I_{corr} = B/R_p \tag{3}$$

where I_{corr} is the corrosion current density, B is a constant (for active anode, $B = 26$; for passive anode, $B = 52$ [24,25]). Once again, since this method needs to know the half cell potential first the depolarization time is necessary for accurate and meaningful measurements. According to the suggestions from the manufacturer, the relations between the corrosion current density and corrosion status are tabulated in Table 3 [26].

2.2.6. Energy dispersion spectrum

It was found that after the ECR, there existed white deposit around anodic cell. The energy dispersion spectrum (EDS) was performed to know what the deposit was.

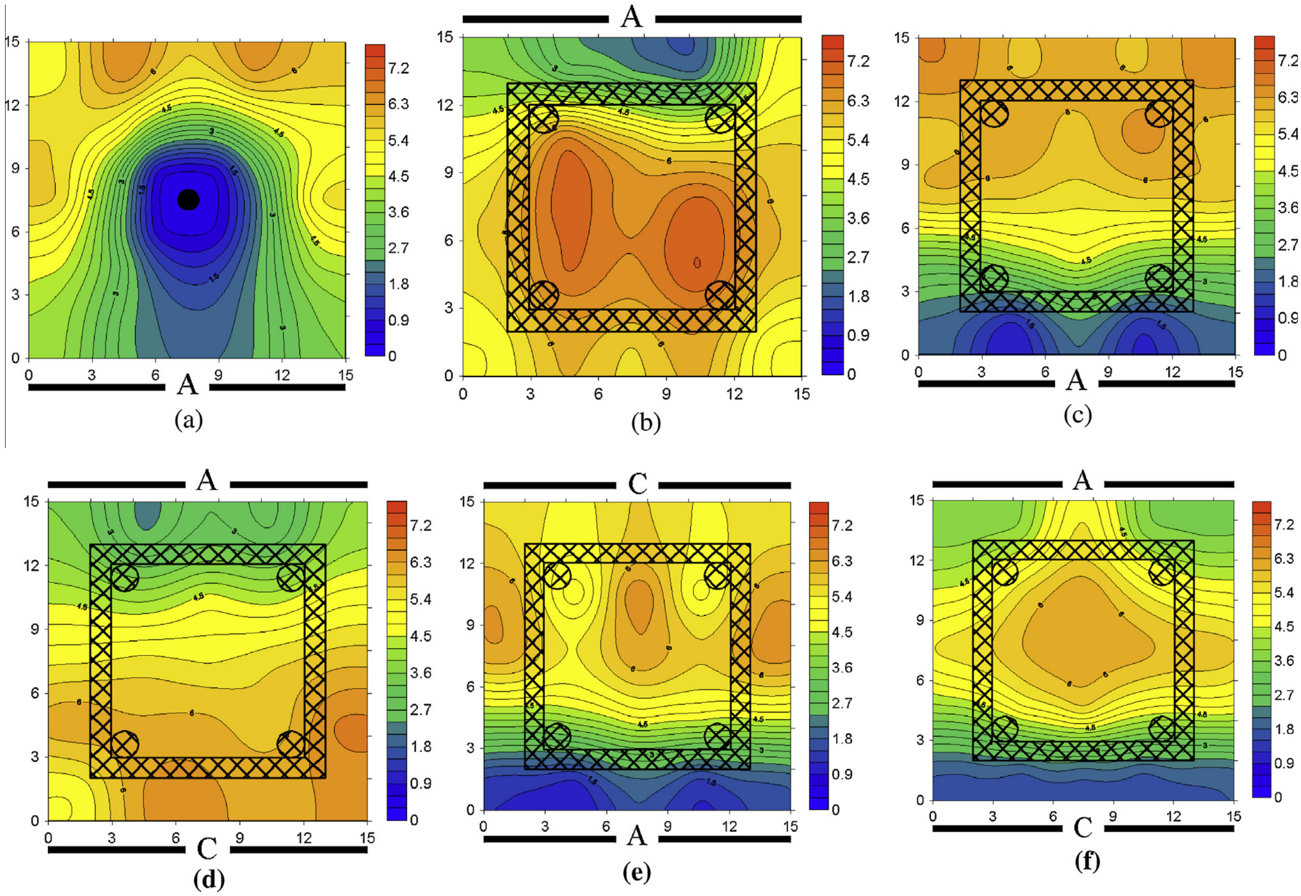
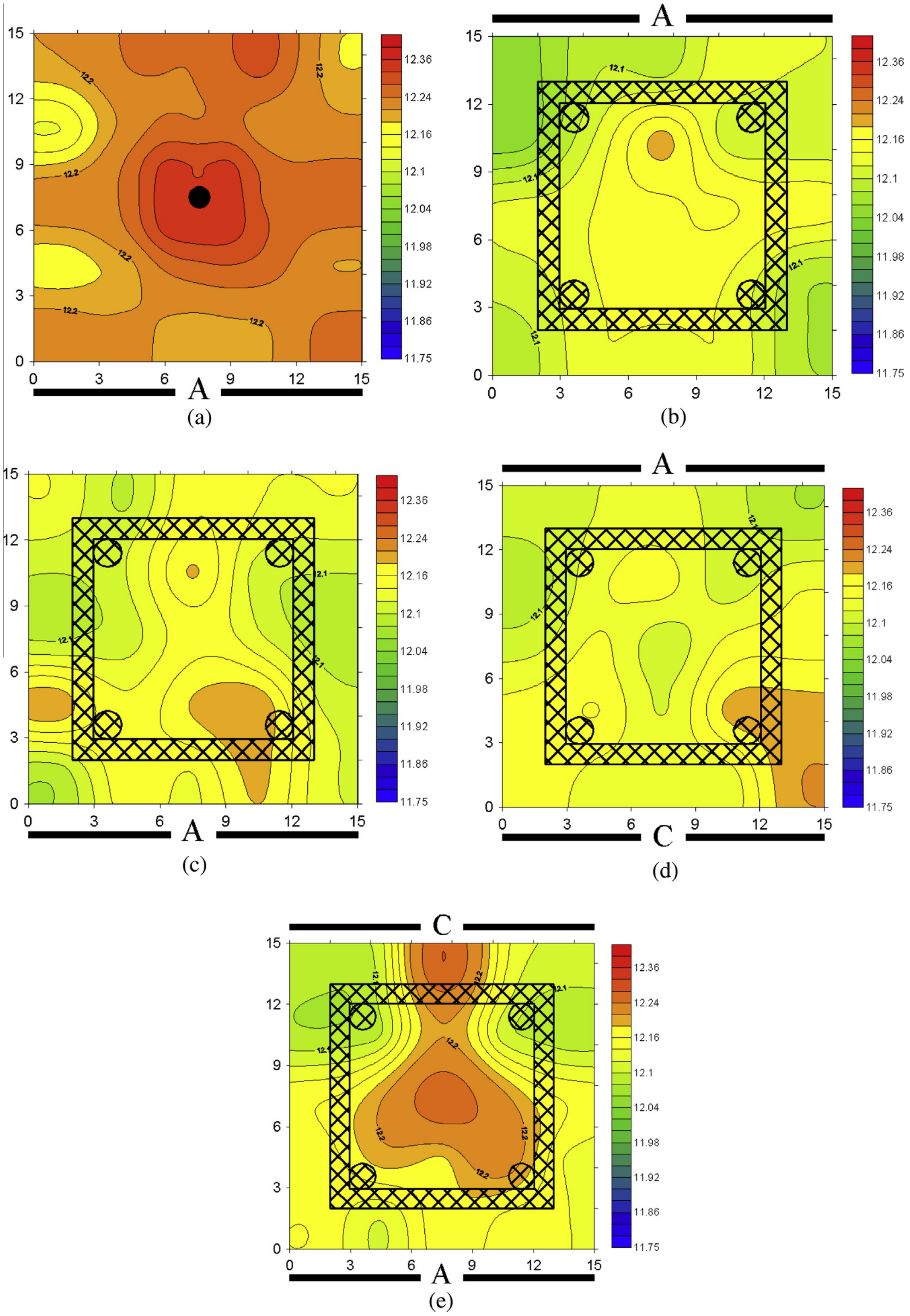


Fig. 9. The chloride ion distributions for various ECR setups after 6 week treatment: (a) setup A; (b) setup B1; (c) setup B2; (d) setup C1; (e) setup C2; (f) setup D. [Unit: kg(Cl$^-$)/m^3(concrete)].

3. Results and discussion

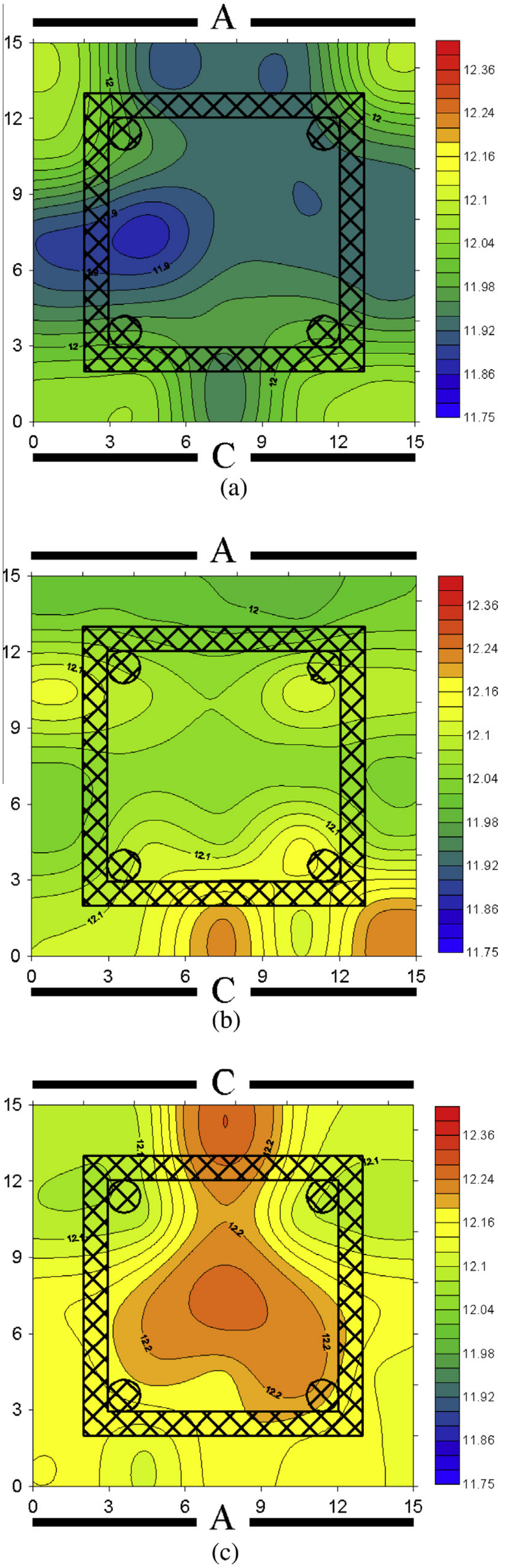
3.1. Rebound strength

The rebound strength reflects the property of concrete surface. It is well known that the rebound strengths relate to compressive strengths [27]. Since the specimens we used contained reinforcement cage such that the compressive strengths of the specimens cannot be carried out. Instead of using compressive strengths, we use rebound strengths to represent the mechanical properties of concrete. The rebound strength measurements after ECR are illustrated in Fig. 7. The 28-day rebound strength is 26 MPa. After ECR, the strength increased with respect to ECR time. The strength gain mainly happened for the first two weeks. In addition, one can observe that the strength gain was highest for setups C1 and C2, then medium for setups B1 and B2, lowest for setup A. First the



(a)
(b)
(c)
(d)
(e)

Fig. 10. The pH values after 6-week treatments: (a) setup A; (b) setup B1; (c) setup B2; (d) setup C1; (e) setup C2.



(a)

(b)

(c)

Fig. 11. The pH values for setup C1 for different treatment times: (a) 2 weeks; (b) 4 weeks; (c) 6 weeks.

strength gain may come from the curing effect from the applied electric field. After 28 days, concrete is mature; however, hydration continues. Applied electric field accelerates the hydration process. After 2-week acceleration, the hydration then slowed down due to the decrease of reactants. The reason why the strength gain for set C is highest, set B is medium and set A is the lowest is explained as follows. The effect of accelerating hydration relates to the area and locations for anode and cathode. The setup C owns the largest area for anodes as well as cathodes and due to the effect of steel reinforcement cage the arrangement for electrodes is mostly uniform. The setup B owns the second largest area for anodes and cathode and it lacks of additional cathode cell thus the effect is less significant than setup C. Setup A has the worst condition therefore it has smallest strength gain.

3.2. Chloride content

Initially the water-soluble chloride ion content for 28-day is 7.53 (kg Cl^-/m^3 concrete). This value was determined using ASTM C1218/C1218M-99.

The chloride removal percentage for a specific ECR treatment time is calculated by the following formula:

$$P_i(t) = \frac{(A - B_i(t))}{A} \times 100\% \qquad (4)$$

where P_i is the chloride removal percentage for the ith sampling region, A is the initial chloride ion content which is 7.53 (kg Cl^-/m^3 concrete) and B_i is the chloride ion content for the ith sampling region.

The efficiency of ECR, E, is estimated by

$$E = \frac{\sum_{i=1}^{25} P_i}{25} \qquad (5)$$

In Eq. (5), the value of 25 in the denominator represents the number of sampling points.

The efficiencies of ECR for various setups and treatment time are shown in Fig. 8. It can be found that the efficiency for setup B1 is smaller than that for setup B2 as well as the efficiency for setup C1 is smaller than that for setup C2. The relative humidity for environment during ECR was about 80–95%, which was apparently very high. In addition, during the ECR the current did not drop a lot which indicated that the moisture supply was enough for continuing ECR. When the water nearby the cathode decreased due to the electrolytic action, moisture supply came from concrete nearby the cathode. The gravity potential helped the moisture supply in setup B2, i.e., the migrations of moisture and chloride ions were in the same direction. It resulted in a better chloride removal efficiency for setup B2 than setup B1 shown in Fig. 8. One also can find out that the efficiency for setup A seems better than setups B1, B2, C1 and C2. Only setup D can reach similar efficiency after 8-week ECR treatment time. The reason can be explained as followings. While the steel reinforcement cage is used, the whole cage has the same electric potential. This makes the chloride ions enclosed by the cage not easy to move. It reconfirms the statement in [28]. The setup D improves the performance; however, it cannot totally resolve the abovementioned effect. The proof of this claim can be found from Fig. 9. The steel reinforcement cage hindered the ECR to remove the chloride ions enclosed by the cage as shown in Fig. 9. In this graph, the letter ‘A’ denotes the anode and the letter ‘C’ denotes the additional cathode.

3.3. pH values

The pH value contours for various ECR setups (A, B1, B2, C1 and C2) after 6 week treatment can be found in Fig. 10. It can be found that for single rebar setup the pH value near the rebar became very high comparing with other parts. However, when the steel reinforcement cage was used the pH value distributed uniformly over all parts. The hydroxide ions will yield during ECR around cathode.

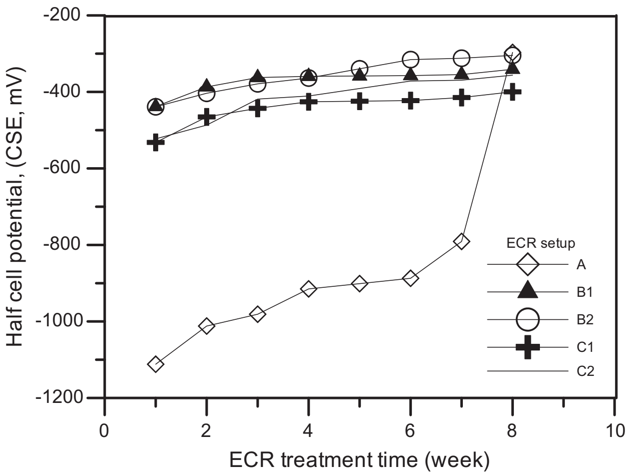


Fig. 12. The half cell potentials after ECR treatment.

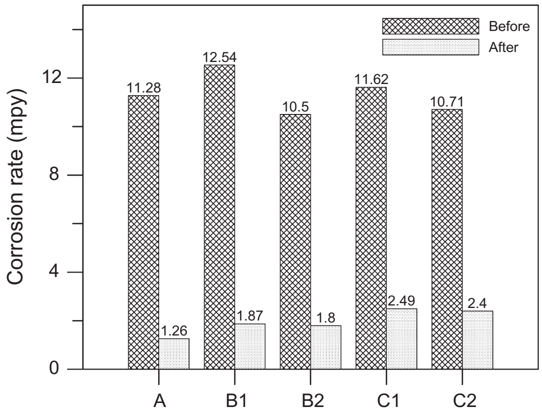


Fig. 13. Instantaneous corrosion rate for various ECR setups after 8-week treatment.

For the cage, all steel surfaces acted as cathode and thus the hydroxide ions were generated and distributed more uniformly. However, due to the increase of cathode area the concentration of hydroxide ions was not easy to be lifted up very quickly as setup A. One may doubt whether or not ECR can make pH value of concrete higher. The history of pH value for C1 setup is shown to answer this question in Fig. 11. One can find that even the steel reinforcement exists the pH value of concrete enhances gradually after the ECR treatment.

3.4. Half cell potential

To understand how the ECR can promote the ability of corrosion prevention, the half cell potential and instantaneous corrosion rate using GPM were recorded. Before measurement, 2-week depolarization time was selected to avoid possible misleading judgment. The results of half cell potentials for various ECR setups are illustrated in Fig. 12. The original half cell potential for the untreated specimen was −950 mV ∼ −1120 mV (CSE). It means that the rebar was in high risk of corrosion. No matter which setup was used, the half cell potential all went to nobler direction and this means the ECR really improve the corrosion tendency of rebar. After 8-week ECR treatment, most setups except C1 had the half potential higher than −350 mV(CSE). In addition, one can observe that for setup A in the beginning the ECR seemed not workable for making rebar nobler. Only after 8-week treatment, the setup A group showed evidence of nobler status. It should be noticed that the electric field intensity may be represented by the applied voltage divided by the distance between cathode and anode. For setup A, the distance between cathode and anode is the highest one therefore we can expect that the driving force is the smallest. Consequently, setup A could not perform better than other setups in the early stage of ECR. Overall speaking, the half cell values indicated that ECR is benefit for corrosion prevention.

3.5. Instantaneous corrosion rate

The half cell potential can only provide thermodynamic trends for rebar corrosion but it cannot really tell us how fast the rebar corrodes. To know how fast the rebar corrodes, the instantaneous corrosion rate is a better indicator. The instantaneous corrosion rates for various ECR setups after 8-week treatment are illustrated in Fig. 13. Before ECR treatment, the instantaneous corrosion rate was about 10.5–12.5 mpy. According to the manual for the GPM manufacturer this means that rebar was corroded seriously. From Fig. 13, one can see after treatment the instantaneous corrosion rate all decreased to about 2.5–1.3 mpy, which means the corrosion status is low. Remember that we have concluded that chloride ions enclosed by the cage cannot be effectively removed from concrete by ECR. However, ECR pushed chloride ions away from rebar and consequently the instantaneous corrosion rate became very small. It is not sure how long will this status last and it leaves as an open question. Theoretically speaking after disconnection of

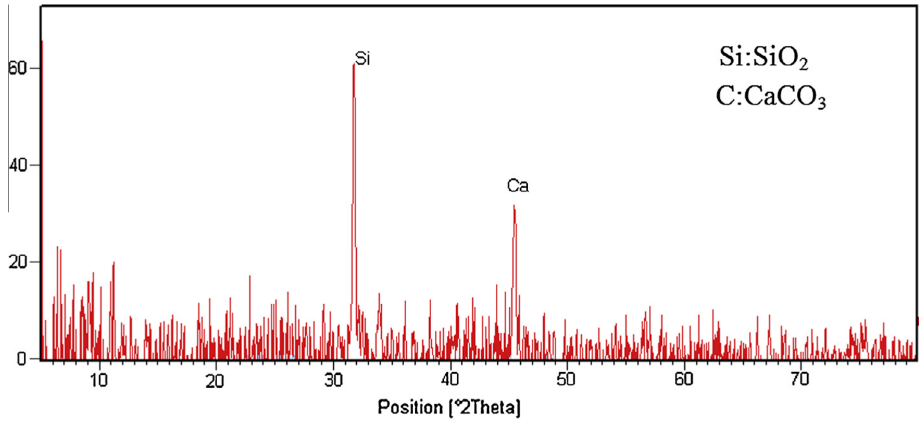


Fig. 14. EDS of the deposit near anode cell.

electric current, the chloride ions will diffuse due to the concentration difference. Whether or not the diffusion of chloride ions (inside the cage) will once again make rebar in corrosion risk is questionable and doubtful.

3.6. Energy dispersion spectrum

It was found that after the ECR, there existed white deposit around anodic cell. The energy dispersion spectrum (EDS) was performed to analyze the compositions of the deposit. The result is shown in Fig. 14. The result indicates that the composition of the deposit near anode is $CaCO_3$. Since in the anode cell the electrolyte solution is NaOH, it can concludes that the calcium ions came from the concrete. It may imply that application of ECR may induce leaching of calcium ions from concrete.

4. Conclusions

In this study, how the stirrups (or the steel reinforcement cage) influence the ECR efficiency is investigated. The results showed that while the steel reinforcement cage was used the chloride ions enclosed by the cage could not be removed from the concrete by ECR. However, the electrochemical signals showed that after ECR treatment the corrosion rate decreased and the open circuit potential became nobler. In addition, the ECR may induce leaching of calcium ions inside concrete.

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