The application of electrochemical chloride extraction to reinforced concrete - A review

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Abstract

In a context of ageing reinforced concrete structures, electrochemical chloride extraction (ECE) is an interesting treatment for structures damaged by chloride-induced corrosion. This paper summarises the state of the art by presenting the concept of ECE, assessing available measurement techniques and discussing influencing factors. The determination of ECE efficacy requires the consideration of different results, and particularly those pertaining to chloride extraction, corrosion mitigation and evolution of pH. This literature review also highlights the potential side effects and risks to the concrete structure. Further work is needed to ascertain the long-term performance of ECE.

Keywords

Reinforced Concrete, Electrochemical chloride extraction, Corrosion mitigation, pH, Long-term performance

Highlights

- This paper presents a state of the art of electrochemical chloride extraction.
- ECE is an interesting treatment for RC structures suffering from chloride induced corrosion.
- Efficiency of ECE is studied based on chloride extraction, steel corrosion mitigation and pH.
- ECE potential side effects are described.
- Long-term efficiency of ECE needs to be validated.

Table 1: Abbreviations used in this paper.

-	not determined
Add	addition of chlorides in the mix
AFNOR	Association Française de Normalisation
ASR	alkali-silica reaction
ASTM	The American Society for Testing and Materials
aSW	artificial sea water
aTi	activated Ti
Av	average (efficacy of chloride extraction)
be	beam
BFSC	blast furnace slag cement
bl	block
bwce	by weight of cement
bwco	by weight of concrete
bwmo	by weight of mortar
C	concrete
CCP	conductive cement paste
CFRP	carbon fibre reinforced polymer
co	column
COST	Cooperation in Science and Technology
CP	cathodic protection
CS	concrete surface
CSE	Copper-copper sulfate electrode
CSH	Calcium-silicate hydrate
cu	cube

cy	cylinder
DC	direct current
De	depth (efficacy of chloride extraction)
DFRCC	ductile fibre reinforced cementitious composites
DS	De-icing salt
DW	distilled water
ECE	electrochemical chloride extraction
f	depends on parameters
f	Free chloride ions
FA	fly ash
G	current control mode
GCP	graphite cement paste
GGBS	ground granulated blast furnace
Gi	intermittent current control mode
GR	graphite rod
Ing	ingressed chlorides
iTi	inert Ti
M	mortar
Mix	chloride added to the mix
OPC	ordinary Portland cement
P	voltage control mode
Pi	intermittent voltage control mode
pr	prism
Re≠	multiple rebar arrangement
Re	rebar
Re#	multiple rebar

RILEM	The International Union of Laboratories and Experts in Construction Materials, Systems and Structures
Rp	polarization resistance
RS	steel rebar surface
SCE	saturated calomel electrode
SF	silica fume
sl	slab
SRC	sulfate resistant cement
SS	stainless steel
SW	sea water
SWI	sea water immersion
SWT	sea water tide
t	Total chloride ions
TW	tap water
W/D	wetting/drying

1 INTRODUCTION

Since the second half of the last century, steel corrosion has been identified as the main cause of deterioration in reinforced concrete structures of civil engineering structures (bridges, tunnels, dams, etc.), buildings (houses, car parks, etc.), and historical monuments (churches, sculptures, etc.) [1-6]. From the moment the concrete is cast, the steel rebar it contains is passivated by the high alkaline pH levels (close to 13). During the service life of reinforced concrete structures, the presence of oxygen and water can lead steel rebar to corrode through the contamination of the rebar/concrete interface by chloride ions and/or the complete carbonation of the concrete cover. The anodic reaction occurring in the steel rebar generates corrosion products in a larger volume than that of the initial iron element, resulting in the formation of cracks in the concrete when stress exceeds its tensile strength. In terms of structural effects, the reduced diameter of the rebar and the debonding of the rebar/concrete interface can lead to decreased loading capacity. The spalling or delamination of the concrete can be a source of danger for the users. Finally, the structure may collapse.

This paper focuses in chloride-induced corrosion [1, 2, 4, 7-9], which is particularly common when the reinforced concrete structure is exposed to marine environment or de-icing salts. In these cases, chlorides penetrate the concrete cover from the outer surface and reach the inner steel rebar/concrete interface. Chloride ions may also have been added to the mix and therefore be more homogeneously distributed within the concrete cover; for example, chloride-based admixtures were sometimes used as setting accelerators (a method that is now prohibited), or marine sand was used for coastal structures.

A number of approaches to avoid steel rebar corrosion have been explored in the literature [10, 11]. Preventive solutions include (i) the use of a protective coating to insulate the steel rebar (epoxy, electroplating or galvanisation) and the use of stainless steel rebar, (ii) the use of anti-corrosion cement, the adapting of the concrete formulation or an increase in the

thickness of the concrete cover in order to delay contamination by chlorides or carbonation, and (iii) the use of corrosion inhibitors or the application of concrete cover coatings.

Maintenance and/or rehabilitation repairs must also be provided for reinforced concrete structures affected by corrosion induced by chloride ions [4, 10-13].

The extensively used conventional “patch repair” consists of replacing all the contaminated concrete and also the rebar when necessary. However, performance of patching through the European CONREPNET project [5] was poor, with a 50% failure rate that was attributed to cracking (30%), debonding (25%), continued corrosion (25%) and other causes (20%). Patch repair becomes impractical for the maintenance of infrastructures such as bridges when contamination reaches beyond the rebar depth. Moreover, the replacement of original materials is not in accordance with restoration principles for historical monuments, as defined by the Venice Charter.

When electrochemical treatments such as cathodic protection or electrochemical chloride extraction are applied to chloride-contaminated reinforced structures, it is only necessary to remove and reconstruct the concrete that no longer bonds with the rebar as polarization ensures the protection of steel rebar, and chloride ions migrate from the steel to the outside of the concrete due to the electrical field.

- Cathodic Protection (CP) of steel in concrete has been demonstrated to be successful in appropriate conditions [14]. CP is designed to decrease the corrosion rate of the steel from significant to negligible values. For this purpose, the steel/concrete potential should be lowered (made more negative) to values in the range of the protection potential. Typical levels of applied current density (with respect to the steel surface) range from 0.2 to 2 mA/m² for prevention and from 2 to 20 mA/m² for protection. CP is a lifelong treatment and requires specific monitoring to assess its efficacy according to several criteria.
- Electrochemical Chloride Extraction (ECE) is also known as desalination, electrochemical chloride removal or electrochemical chloride mitigation. A current

is applied between the steel rebar and an external anode in order to remove chloride ions from the concrete cover, and particularly from the rebar/concrete interface. The current density is usually 1 A/m² of steel rebar surface (approximately 100 times higher than the density used in CP) and is applied for a limited time (4 to 8 weeks) [15].

The aim of this literature review is to gather all available data about the application of electrochemical chloride extraction (ECE) to reinforced concrete. The following section of this paper describes ECE parameters. We then move on to presenting the different ways in which the efficacy of the treatment can be defined. The results of studies based on different parameters pertaining to specificities such as chloride extraction efficacy, corrosion mitigation and pH evolution are discussed in the fourth part of this paper. The fifth section focuses on the potential side-effects and risks of the ECE treatment. The long-term efficacy of ECE is then discussed, followed by ECE specifications and modelling. Finally, this review ends with our conclusions, recommendations for laboratory studies and practical cases, and suggestions of future research avenues.

2 ECE PRINCIPLE

2.1 History

The first trials of electrochemical chloride extraction (ECE) treatment with high DC voltages of up to 220 V were led in the United States of America by the Federal Highway Administration in the mid-70s [16-18], but this procedure was rarely used over the next 20 years. An invention was patented in Europe during the 80s by Oystein Vennesland, a researcher at Norwegian Concrete Technologies [19]. Knowledge of ECE was developed further in the 90s through research carried out in the United States of America Strategic Highways Research Program (SHRP) [20-23]. Pocock's 1994 publication [24] recognized the interest of this treatment for

the repairing of reinforced concrete contaminated by chloride ions. Different publications, state-of-the-art report, chapters on ECE treatment were published [25-27]. ECE treatment was also part of three European Cooperation in Science and Technology (COST) actions (COST 509 in 1997 [10], COST 521 in 2003 [11] and COST 534 (2007) [28]). In 2022, task group 8.1 from the federation Internationale du Béton published fib Bulletin 102 "Guide for protection and repair of concrete structures" [29] and devoted a chapter on chloride extraction/desalination [30].

The present literature review seeks to gather the different results from the 1970s until 2022.

2.2 Mechanisms

Corrosion of the steel rebar occurs by two electrochemical reactions: the anodic dissolution of iron, as shown in Equation 1, and the corresponding oxygen reduction at the cathode that uses the electrons generated by the metal dissolution in its reaction, as shown in Equation 2.

Anodic dissolution of iron	$Fe \rightarrow Fe^{2+} + 2e^-$	*Equation 1*
Oxygen reduction	$O_2 + 2H_20 + 4e^- \rightarrow 4OH^-$	*Equation 2*

Both of these reactions occur on the steel surface, resulting in the formation of corrosion products. The accumulation of corrosion products at the rebar/concrete interface (Equation 3) may induce cracks in the concrete cover.

Corrosion products	$Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2$	*Equation 3*
	$Fe(OH)_2 + nO_2 \rightarrow Fe_2O_3.nH_2O$	

ECE is designed to arrest the corrosion of steel rebar, or even recover the passivity thereof, through the following phenomena: a lowering of the thermodynamic potential of the steel, a reduction of the chloride content and an increase in the pH levels at the rebar/concrete interface.

Figure 1 shows a schematic representation of the ECE set-up when a direct current source is used[1)]. The rebar requiring protection acts as a cathode and is connected to the negative pole of the direct current source, while the anode (usually located on the surface of the concrete) is connected to the positive pole. During ECE, it is important to minimize circuit resistance between the cathode and the anode and to maintain a buffered pH. This is usually achieved with the joint use of a solution called electrolyte and one of three different methods: tanks, sprayed cellulose fibres, or synthetic felt mats.

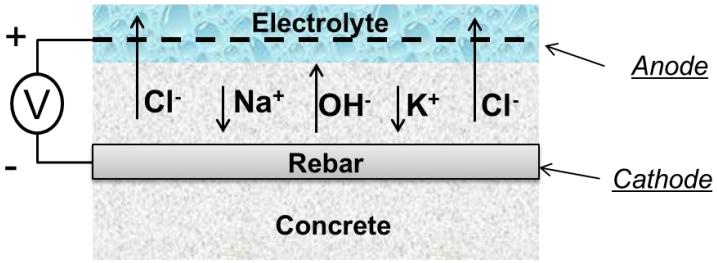

Figure 1: Schematic representation of ECE set-up.

In alkaline reinforced concrete[2)], the difference in electrical potential between the cathode and anode triggers the transport of the ions present in the electrolyte and in the concrete [25, 31-35], and may cause the following reactions:

Oxygen reduction	$O_2 + 2H_20 + 4e^- \rightarrow 4OH^-$	*Equation 4*
Water reduction	$2H_20 + 2e^- \rightarrow H_2\uparrow + 2OH^-$	*Equation 5*

The low potential of the steel at the cathode protects the rebar from corrosion. The oxygen reduction reaction (Equation *4*) takes place slowly due to the limited availability of oxygen in concrete: at the outset, there is very little oxygen available around the cathode within the concrete, and oxygen diffusion from the outside is slow. Thus the water reduction reaction

[1)] Sacrificial anodes are not considered in this paper.
[2)] Because concrete is an alkaline medium (sound concrete pH~13 and carbonated concrete pH~9), hydroxyl ions (OH^-) >>> protons (H^+).

(Equation 5) predominates [34]. The production of hydrogen gas can increase local pressure, eventually leading to concrete cracking and hydrogen embrittlement of steel (§5.2.2).

At the anode, the oxidation of hydroxyl ions forms oxygen and water (Equation 6). No dissolution of the anode occurs if the metal is inert (Equation 7). In the case of electrolyte acidification, the oxidation of chloride ions in chlorine gas (*Equation 8*) should be avoided for safety and environmental concerns.

Water oxidation	$4OH^- \rightarrow O_2 + 2H_2O + 4e^-$	*Equation 6*
Anode dissolution	$Me \rightarrow Me^{n+} + ne^-$	*Equation 7*
Chlorides	$2\ Cl^- \rightarrow Cl_2 + 2e^-$	*Equation 8*

In addition to these oxidation-reduction reactions, at typical current densities for ECE, migration dominates with the transport of ions contained in the concrete and in the electrolyte. The transport of ions involves different processes based on concentration gradients, electrical migration, lixiviation and diffusion. Due to the electrical field of the rebar and the anode, anions such as free chlorides (Cl^-), hydroxyls (OH^-), carbonates (CO_3^{2-}), and sulfates (SO_4^{2-}), are mostly repelled from the vicinity of the rebar and migrate away from the cathode region towards the anode. Similarly, cations such as calcium (Ca^{2+}), potassium (K^+), sodium (Na^+) and lithium (Li+) ions migrate towards the cathodic steel region. Electrical neutrality is maintained on a macroscale despite the modified distribution of specific ions within the concrete. It is important to note that the accumulation of alkali ions can cause an alkali-silicate reaction if the concrete contains reactive aggregates (§5.2.4).

Figure 2 provides a schematic representation of the electrical field obtained during the treatment, and shows that the maximal extraction efficacy is obtained in the area in between the rebar and the anode. Moreover, chloride ions present at the deepest levels close to the rebar cannot be easily extracted [25].

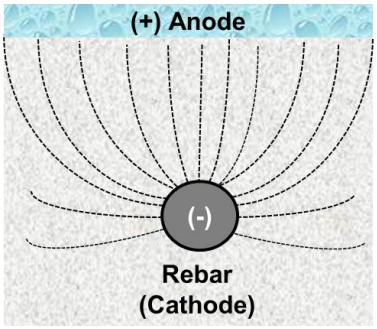

Figure 2: Schematic representation of streamlines in an inhomogeneous field between rebar (cathode) and anode at the concrete surface.

2.3 Process parameters

In this section, each component of the ECE treatment (Figure 1) is presented and some important features are explained.

2.3.1 **Cathode**

In almost all cases, the cathode is the rebar (simple or deformed rebar, mild steel plate, rebar plus stirrups or structural reinforcement).

Arya et al. [36] used an additional external cathode for cases in which chloride was present at a deeper level than the rebar. Setup design affected chloride extraction, which increased from 40% (obtained with the conventional setup) to 55% when both the rebar and external platinised titanium mesh were connected as cathodes.

2.3.2 **Anode**

In the context of ECE treatment, the anode has to satisfy various criteria to be fully functional [4]. It must distribute the chosen current throughout the area requiring protection for the entire duration of the treatment. This element must also be corrosion resistant and should not affect the appearance of the concrete surface. Finally, its installation should be inexpensive.

During their preliminary study in 1976, Slater et al. [16] tested platinized titanium, titanium and graphite anodes in a 0.1 N calcium hydroxide solution. Platinized titanium was found to be the most appropriate anode material; it satisfied the test criteria, unlike graphite (which disintegrated at high current densities) and titanium (which had poor electro catalytic activity). The most reliable and widely used anode today is composed of titanium that has been activated with oxides or coated with different metals. This anode can take the form of mesh, wire or strip. It is suitable for the protection of different concrete structures, and can be used to treat vertical and horizontal structures and both sides of curved surfaces. Ueda et al. [37] used a titanium mesh cast in a ductile fibre (PE or PVA)-reinforced, cementitious composite (DFRCC), which improved its adhesion to a prepared concrete surface. Although chloride extraction efficacy at the rebar level was close to 75%, chlorides were accumulating in the DFRCC layer and could therefore spread back into the concrete cover.

Steel was tested given its low price, however its use is not recommended because it is rapidly consumed during the treatment (Equation 9) and causes rust spots on the concrete surface [20].

Different anode systems to replace titanium anodes have been studied and referenced in the literature. Pérez et al. [38] worked on the viability of replacing the classical Ti-RuO_2 mesh anode with a graphite anode and a conductive cement paste (CCP) combination, applied superficially as a plaster. Results showed that although the chloride extraction was similar for both types of anodes, an accumulation of chloride ions appeared in the CCP layer that could eventually spread back into the concrete cover. Canon et al. [39] and Carmona et al. [40] studied the feasibility of using a sprayed conductive graphite powder cement paste as an anode, which would make it possible to work on various surface shapes and inclinations and therefore facilitate the application of ECE. The results showed that the chloride extraction efficacy was similar to that observed in the titanium-based anode. Moreover, unlike the anode suggested by Pérez et al. [38] this sprayed layer retained moisture for up to 10-12 h but failed to retain chlorides. The durability of this anode layer has been questioned following damage by anodic reactions. Finally, Zhu et al. [41] studied the application of a conductive carbon-fibre-

reinforced polymer (CFRP) anode in the form of a mesh with good mechanical properties, durability, conductivity and electrochemical stability. The results for chloride extraction were similar to those obtained with an activated titanium mesh anode. It should be noted that values are homogenized when a conductive anode layer is used meaning that half-cell potential mapping measurements cannot be carried out for inspection.

2.3.3 **Electrolyte**

The electrolyte plays three main roles in ECE treatment:

- First, it decreases the electrical resistance by wetting between concrete and reinforcement (hydration and dissolution of the ions in the concrete), thus lowering the voltages used and improving ionic migration,
- Second, the pH of the electrolyte should prevent any modification of the concrete pH,
- Lastly, it should trap chlorine gas, which is hazardous to health.

Usually, renewal or circulation of the electrolyte is needed. Tanks or sprayed cellulose fibre can be used for the treatment when applied on horizontal or vertical surfaces.

The two most widespread electrolytes for ECE are water [34, 39, 40, 42-52] and saturated calcium hydroxide ($Ca(OH)_2$) solution [16, 20, 41, 43, 44, 51-66]. Saturated calcium hydroxide solution with a pH equal to 12.4 is consistent with the requirement for a buffered medium and the need for a relatively high pH value to prevent acidification and chlorine gas.

Other electrolyte solutions containing sodium ions were studied, namely sodium hydroxide (NaOH) [36, 43], sodium carbonate (Na_2CO_3) [65] and sodium borate solutions (Na_3BO_3 [56], $Na_2B_4O_7$ [67] and $Na_2B_4O_7$ $10H_2O$ [68, 69]. Sodium ions can sometimes be detrimental in terms of Alkali-Silica Reaction (ASR) (§5.2.4). Finally, lithium borate solution was also used as an electrolyte, as lithium ions have already demonstrated their ability to limit ASR [23, 37, 70, 71]. Moreover, Ueda [70] demonstrated that increasing the temperature of Li_3BO_3 electrolyte solution resulted in a higher percentage of chloride extraction.

2.3.4 Current flow

The current flow between the rebar and the anode must ensure the cathodic protection of the steel rebar. Current flow can be obtained in two modes: constant voltage (voltage control) and constant current (current control) in a continuous or intermittent ways. In the voltage control mode, the current flow is expressed in voltage [16-18, 20-23, 36, 49, 58, 72-75]. In the current control mode, the current flow is expressed in current density in A/m² of steel surface or concrete surface that must be indicated. Current density is expressed in A/m² of steel rebar surface in references [35, 40, 41, 43, 45, 47, 48, 51, 54, 55, 57-60, 63, 65, 67, 69, 76-85], and in A/m² of concrete surface in references [21, 34, 37-39, 44, 46, 53, 62, 70, 71, 86, 87]. No surface information is clearly indicated in references [61, 88, 89].

Moreover, some authors attempted to increase the efficacy of the treatment through the use of intermittent rather than continuous current flow, via voltage control mode [18] or current control mode [34, 45, 46, 48, 55, 81, 86, 90, 91].

It is important to note that it is very difficult if not impossible to compare the results if the ECE modes are not the same or if the current density units are different.

2.3.5 Duration

ECE is a short-term treatment (vs Cathodic Protection). It is often applied for periods ranging from 2 to 8 weeks [23, 32, 34, 36-39, 41-44, 46-48, 51-55, 57-61, 65-67, 70, 71, 75, 79, 80, 82, 84, 87, 88, 92-95]. Larger duration than 15 weeks [42, 74, 76, 88, 96] has also been considered particularly when using lower current density or for testing prestressed concretes.

3 TECHNIQUES TO ASSESS ECE EFFICACY AND POTENTIAL SIDE EFFECTS

This third section describes the different non-destructive and destructive techniques used by researchers to characterize the efficacy of ECE, and seeks to verify whether the ECE treatment

induces chemical, microstructural and/or mechanical changes that could be detrimental to the reinforced structure.

3.1 Visual inspections

A visual inspection of the appearance of the rebar and its surrounding concrete can provide general information such as corrosion product colours, layer of deposits, cracks, etc. [45, 49, 57, 59, 71, 76-79, 83, 87, 96].

3.2 Chloride analyses

The efficacy of the treatment varies according to the chloride content before, during, after and long after ECE (in case of re-diffusion). In concrete, chlorides are either freely dissolved in the pore solution or exist in a bound form with cement hydration products such as hydrated C3A or C4AF phases and CHS gel. Moreover, penetrated chloride content varies according to the depth at which it is located in the concrete. Usually specimens (from the laboratory or from cores obtained on the field) are sliced, crushed, sieved and dissolved in water where the chloride content profile is determined as water soluble (free chlorides) or acid soluble (total chlorides) based on potentiometric titration using silver nitrate or spectrophotometric analysis or chemical analysis [43, 48, 49, 53-55, 59, 70, 97-99].

It is important to note that the measurement of free chloride ions in the interstitial solution is only possible for pastes [32, 34] where pressure needed for extraction can be achieved. Finally, Elsener et al. [48] developed a chloride ion-selective electrode as an embedded sensor in concrete.

The concentration of removed chloride ions can also be determined from electrolyte analysis [43, 45, 51, 55, 70, 83]. Polder et al. [43] found a good general correlation between the amount of chloride calculated from electrolyte analysis and concrete analysis, respectively.

3.3 Electrochemical characterizations

Non-destructive electrochemical measurements such as half-cell potential, linear polarization resistance and corrosion rate can be used to characterise the corrosion state of the rebar when the latter is in a steady state (no polarization) [16, 20, 23, 37, 42-46, 48, 49, 51-54, 59, 60, 67, 70, 73-75, 79, 82, 84, 100]. According to the literature, the efficacy of the treatment that will eventually lead to a passive state can be assessed by performing electrochemical measurements before ECE and after ECE once the polarization of the rebar is over. It is important to note that the time at which depolarization of the steel occurs cannot be known in advance. During or immediately after the treatment, these electrochemical measurements should confirm that the rebar is polarized (i.e. high electronegative half-cell potential and high current densities values are observed). This is an interesting means to verify that different treated specimens are reacting in the same way, or to prove that no area has been forgotten when the treatment is applied on-site, for example.

Half-cell potential measurements are the most commonly used method because they are very easy to conduct [20, 37, 43, 45, 46, 48, 49, 51-54, 59, 60, 67, 70, 73-75, 79, 82, 84, 100]. However, it should be noted that the results are strongly dependent on parameters such as concrete resistivity (moisture content), chloride content, pH, temperature or climatic conditions. Half-cell potential measurements aim to evaluate the probability of rebar corrosion (when all the other parameters remain constant) and/or to locate corroding zones in larger specimens and in on-site structures, where climatic conditions are not controlled. For reinforced concretes in atmospheric conditions, test procedures and guidelines are described in the ASTM C-876 standard (1991 and 2009) [101, 102] and in the RILEM TC 154 recommendation (2003) [103]. After ECE, more positive potentials and smaller gradients are expected (versus before ECE). Despite the availability of these tools, result analysis for reinforced concrete structures in a marine environment can still be difficult because of the tide that modifies humidity and oxygen content.

The measurement of polarization resistance (Rp) before, during and after treatment with a potentiostat [54, 57, 60] permits the evaluation of the corrosion rate at a given time. Corrosion rate can also be measured on site using a GECOR 6 [32, 55] or a GPM-5000 [73]. The results obtained can be compared to the established corrosion rate classification [104]. The RILEM TC 154-EMC document provides important recommendations for on-site corrosion rate measurements [103]. It is important to note that corrosion rate measurements using Rp do not give realistic values at short timescales after current interruption. This is likely to be due to the chemical changes induced close to the rebar (i.e. high alkalinity and low O_2 activity) [53].

Miranda et al. [59] and Marcotte et al. [79] used a non-stationary method by performing full polarization curves to assess the corrosion rate of reinforced concrete slabs before and after ECE (with ECE performed on both sides of the slabs).

3.4 pH determinations

During the ECE process, pH evolution at the rebar/concrete interface, in the concrete and in the electrolyte can be analysed. This analysis can involve the use of phenolphthalein, a colour acid/base indicator that will turn purple when sprayed on concrete with a pH level of 9-10 or above [35]. Other authors titrated the pH of concrete from concrete samples dissolved in water [58, 73, 75, 82]. Finally, pH values for the electrolyte were reported in [43, 45, 55].

3.5 Concrete characterizations

3.5.1 Ion migration

The performance of atomic absorption spectrophotometry on solutions obtained from dissolved concrete provides quantitative information on the presence of ions such as potassium, sodium, calcium and lithium and their migration through the electrical field during ECE, and thus provides information about the evolution of the concrete composition [45, 52, 57, 71].

3.5.2 Microstructure transformation

Petrographic examination can be performed [23, 43] to study the microstructural evolution of the concrete (i.e. micro cracks, pores, etc.).

Scanning electron microscopy can be used to observe the microstructure of concrete and identify any new phases (petrographic) formed during ECE by comparing treated or untreated specimens [35, 41, 47, 57, 60, 69, 80, 83, 88].

X-ray diffraction is also used to analyse additional crystalline materials present in the concrete or in concrete surface deposits [60, 87, 100].

The Mercury Intrusion Porosimetry technique can be used to determine the pore size distribution within the concrete cover [83] and evaluate ECE impact.

3.6 Steel characterizations

Microstructural observations of the rebar/concrete interface by scanning electron microscopy coupled with dispersive X-ray spectroscopy are used to identify the corrosion products and their location at the rebar/concrete interface after the corrosion process and after the ECE treatment. X-ray diffraction was used by Miranda [59] to determine the nature of the corrosion products.

Raman microspectroscopy can identify the iron corrosion products during and after treatment and help to ascertain the corrosion state of the rebar. Tissier et al [69] developed a specific electrochemical cell set-up without any oxygen contact to perform in-situ observations in the laboratory.

3.7 Mechanical tests

Compressive strengths were studied in [55, 80]. Pull-out tests were performed [41, 50, 58, 76, 78, 96] to determine bond stress changes. For bigger specimens like beams, flexural strength tests were conducted [55].

4 LABORATORY RESULTS

All the published studies considered in the present evaluation are presented in three tables (Table 2, Table 3 and Table 4) according to the electrochemical chloride extraction mode used. All the abbreviations can be found in Table 1.

Table 2 summarises all the published studies regarding voltage control mode. The voltages are in the range of 5 to 100 V. High voltage (100 V) led the temperature of the concrete to increase drastically (from 25 °C to 50-84° C) and resulted in concrete cracks in some cases.

The major disadvantage when using a voltage control mode is that the current received by the rebar will decrease over time as the resistance of the medium increases (due to the extraction of chloride).

Using current control mode allows a better repeatability of the experiment. Table 3 summarises all the published studies where the current density is based on the area of the concrete surface (A/m² CS), whereas Table 4 concerns studies where the current density is based on the area of the steel rebar surface (A/m² RS). The type of surface was not clearly indicated in papers [61, 88].

Current densities based on the area of the concrete surface (Table 3), are calculated on the assumption that the ratio of the rebar area to concrete area in the treated structure is equal to one [88]. However, this is not always correct. For example, 1 A/m² of CS can be equal to 5 A/m² RS [88] or to 9.5 A/m² RS [78]. Unnecessarily high current densities can lead to detrimental side effects including increased temperature and concrete cracking. The aim of ECE is to protect the steel from corrosion in concrete; the current density must therefore be based on the area of the steel rebar surface.

Elsener et al. [48] proposed to use an intermittent current control mode in which bound chlorides were released as free chlorides during the OFF period. This led to increased efficacy of chloride extraction during the next ON period. The authors even reached 100% efficacy in this experimental program.

Several papers studied the use of an intermittent voltage control to increase the efficacy of ECE [45, 48, 55, 81, 90]. An interruption of current [48] may have the following advantages:

- Bound chlorides may become free chlorides.
- A redistribution of ions (Cl^-, OH^-) may occur within the concrete. The current interruption period should be long enough to achieve the depolarization of the rebar and therefore permit the expected re-distribution of ions.
- Risks of undesirable side effects arising from the application of electrochemical techniques can be reduced.

Table 4 describes the current control mode and applied current density based on the steel rebar surface. Here, 1A/m² RS is the most frequently used value [41, 43, 45, 47, 51, 54, 55, 57, 61, 63, 65, 67, 69, 81, 82, 85, 90]. Lower densities of applied current (0.25-0.75 /m² RS) were chosen to study the influence of cementitious binder on chloride extraction under electrochemical treatment in concrete [63, 65, 76, 84, 90]. The application of higher current densities was also reported in ranges of >1-4 A/m² RS [40, 41, 43, 48, 51, 59-61, 63, 65, 77, 82, 83, 85] and ≥ 5-28.6 A/m² RS [59, 60, 77-80, 82]. The highest densities of applied current were mainly used to study the influence of the ECE treatment on the mechanical properties of reinforced concretes and particularly the loss of bond between the rebar and the concrete. Treatment most often lasts 28 or 56 days, but other periods in the range (3-112 days) were also reported.

From here on, our discussion will solely concern the ECE results obtained with a current control mode and a current density expressed in A/m² of steel rebar surface (RS), described in Table 4. Since these data were obtained from a variety of experimental setups, the corresponding details must be considered when comparing them.

The determination of ECE treatment efficacy requires the simultaneous consideration of different results, particularly those pertaining to chloride extraction (§4.1), corrosion mitigation

(§4.2), evolution of pH (4.3) and visual aspects (§4.4). Miranda et al. [59] argued that the determination of chloride profiles, the use of the phenolphthalein test, or the simple measurement of potentials before and after the application of ECE cannot guarantee its efficacy, and underlined the importance of using techniques that provide quantitative determinations of corrosion kinetics. Orellan et al.[54] added that a polarization resistance technique is needed to confirm that the steel has re-passivated after treatment.

1 *Table 2: Published laboratory studies on ECE treatment performed using a voltage control mode (continuous P or intermittent Pi).*

2

First author [Reference]	Binder	Sample	w/c ratio	Cement (kg/m3)	Dimensions (mm) [shape]	Steel rebar (diameter mm)	Cover (mm)	Initial chloride content [mode] / [content Cl^-]	Cathode	Anode	Electrolyte	Voltage control mode	Duration (days)
Lankard [18]	OPC	C	0.44	360	76dx150 [cy]	10	50	[Mix, NaCl] / [(De) (t) 0.019-0.144%bwco]	Re	Ti-Pt	[$Ca(OH)_2$+IER] / [top pond]	[P] / [V 50, 100]	1-3
Lankard [18]	OPC	C	0.45	360	1170x2280x230 [sl]	20	50	[Mix, NaCl] + [ponding 3%NaCl 5 months]	Re	Ti/Pt	[$Ca(OH)_2$+IER] / [top pond]	[P, Pi] / [V 100]	1-2
Chang [73]	OPC	C	0.5	393	150x150x300 [pr]	Re# 9.52, 12.30	20	[Mix, NaCl] / [(Add), 1.28%bwce]	Re#, External cathode	TiPt	[NaOH] / [tank]	[P] / [V 20]	14, 28, 42, 56
Yeih [75]	OPC	C	0.5	393	150x150x300 [pr]	R# 9.52, 12.30	20	[Mix, NaCl] / [(Add), 1.82% bwce], [(Av), (f), 1.79%bwce]	Re	Ti mesh	[NaOH anode cell, TW cathode cell] / [acrylic cell]	[P] / [V 20]	56
Arya [36]	OPC	C	0.65	400	200x200x120 [pr]	10	30, 50, 80	[Mix, NaCl] / [(Add) 2, 3, 4%bwce] / [2 layers: bottom 0%bwce and top layer 75mm: 1.87%bwce]	Re#, External cathode	iTi	[NaOH] / [tank]	[P] / [V 10, 20, 30]	33, 83
Chang * [58]	OPC	C	0.66	307	100dx200 [cy]	12.3	40	[Mix, NaCl] / [(Add), 1.28 %bwce; (Re), 3800ppm bwco]	Re	Ti	[$Ca(OH)_2$] / [tank] [NaOH] /	[P] / [V 5, 10, 20, 30]	7, 14, 21, 28, 35, 42

											[tank]		
Sanchez [49]	OPC	M	0.5	-	20x55x80 [pr]	6	24.5	[Mix, NaCl] / [(Add), 1%bwce]	Re, External cathode	-	[DW] / [tank]	[P] / [V 12]	5
Martinez [74]	OPC	M	0.55	450	70x70x70 [cu]	6	30	[Mix, NaCl or $CaCl_2$] / [(Add), 1.52 or 2.35%bwce]	Re	MMO aTi	[W] / [sponge]	[P] / [V 8, 12 ,16, 20]	0-93, 115, 133, 143

*) Authors said that a voltage control of 5 V corresponds to a 18.8A/m² CS current control

Table 3: Published laboratory studies on ECE treatment performed using a current control mode (continuous G or intermittent Gi) with current densities expressed as A/m² of concrete surface (CS).

First author [Reference]	Binder	Sample	w/c ratio	Cement (kg/m^3)	Dimensions (mm) [shape]	Steel rebar (diameter mm)	Cover (mm)	Initial chloride content [mode] / [content Cl^-]	Cathode	Anode	Electrolyte	Current control mode	Duration (days)
Tritthart [34]	-	C	-	-	50d, 200d [cores]	2 rebar layers	45-55 and 60-70	[Ingress, DS] / [(t), (De) 0-170mm: 15-0.5%bwce; (De) 0-10mm: 12-15%bwce, 40-60mm (overlapping bar depth): 7.8-9%bwce; 90-100mm: 3-4.5%bwce; 150-160mm:0.5%bwce]	Re	Ti	[W] / [paper pulp]	[G, Gi] / [A/m² CS 1]	40, 83, 96, interruption 98d, 139d
Canon [39]	-	C	0.6	350	200dx1000 [co]	Re# d8 Stirrups d6	40	[Mix, NaCl] / [(Add), 2%bwce] / [(t) 1.5%bwce]	Re	Ti-RuO_2, CCP	[TW] / [irrigation or dampening]	[G] / [A/m² CS 5]	6, 8, 13.5, 14.6
Ihekwaba [86]	OPC	C	0.3	432	300dx1200 [co]	16	50	[Mix, NaCl] / [(Add), (De) 1.7%bwce inner 15cm, 3%bwce outer 15cm] / [(Add), (Av), 5000 (#1), 6000 (#2) ppm bwco]	Re#	iTi	$[Ca(OH)_2]$ / [cellulose fiber]	[G, Gi] / [A/m² CS 1]	56, 70 8w ON then 2w OFF, then a second ECE on 1 of the specimens for 2w ON
Ihekwaba [86]	OPC	C	0.3	432	300x300x1200 [co]	16	50	[Mix, NaCl] /	Re#	iTi	$[Ca(OH)_2]$ / [cellulose fiber]	[G, Gi] / [A/m² CS 1]	56, 70 8w ON then 2w OFF,

								[(Add), (De) 1.7%bwce inner 15cm, 3%bwce outer 15cm] / [(Add), (Av), 5000 (#3), 6000 (#4) ppm bwco]					then a second ECE on 1 of the specimens for 2w ON
Ueda [70]	OPC	C	0.45 0.55	384 340	100x100x200 [pr]	d13 deformed	43,5	[Mix, NaCl] / [(Add), 8% bwce] / [(t) 2.35, 2.08%bwce]	Re	Ti	[Li_3BO_3] / [tank]	[G] / [A/m² CS 1]	56
Sharp [87]	OPC	C	0.4 0.45 0.5 0.55	377	(92, 105, 118)x127x305 [bl]	16	44, 57, 70	[Mix + Ponding] / [(Add), 0.25%bwco]	Re	aTi	[$Ca(OH)_2$ or CaO] / [felt]	[G] / [A/m² CS 1]	50
Sharp [87]	OPC	C	0.45 0.5 0.55 0.6	377 331 301 276	(177, 190, 203)x610x608 [bl]	16	38, 51, 64	[Mix] / [(Add), 0.25%bwco]	Re	aTi	[$Ca(OH)_2$ or CaO] / [felt]	[G] / [A/m² CS 1]	50
Bennett [20]	OPC	C	0.5	363	600x600x200 [sl]		50	[Mix, NaCl] / [(Add), (De), bottom 8cm: 0.0%bwco, median 8cm: 0.16%bwco, top 4cm: 0.39%bwco] / [(t), (Tit), (De), 0.015-0.410%bwco]	Re	Steel	[$Ca(OH)_2$] / [cellulose fiber]	[G] / [A/m² CS 1]	91
Ihekwaba [56]	OPC	C	0.5	370	500x500x300 [bl]	2 steel mats (uniform or displaced) d19	60	[Pond, NaCl, 18m] / [carbonated blocks, (Av), (t) 1500-3000 ppm bwco ; non-carbonated blocks, (Av), (t) 3500-8000 ppm bwco]	2 steel mats	iTi	[Na_3BO_3] / [top pond]	[G] / [A/m² CS 1]	-

Buenfeld [35]	OPC	C	0.5	400	102dx254 [cy]	32	35	[Mix, NaCl] / [(Add), 1.5%bwce] / [(Add), (De 0-30mm), (t) 0.48-0.32%bwce]	Re	aTi	[wet cellulose fibre] / [tank]	[G] / [A/m² CS 1]	28, 56
Ueda [71]	OPC	C	0.55	338	100x100x300 [pr]	13	43	[Mix, NaCl] / [(Add) 8%bwce] / [(Av), (t) 2.37%bwce]	Re	Ti	[LiOH, Li_2CO_3, Li_2SiO_4, $LiNO_3$, $Ca(OH)_2$] / [tank] / [30, 40°C]	[G] / [A/m² CS 1]	56
Ueda [37]	OPC	C	0.55	318	100x80x400 [be]	d10 deformed	15	[Mix, NaCl] / [(Add), 8%bwce] / [(t) 2.52%bwce]	Re	Ti in DFRCC	[Li_3BO_3] / [DFRCC]	[G] / [A/m² CS 1]	56
Green [53]	OPC	C	0.66	288	130x100x100 [bl]	16	42	[Mix, NaCl] / [(Av), (t) 0.26% bwco = 2.17%bwce]	Re	Ti-Pt	[$Ca(OH)_2$] / [tank]	[G] / [A/m² CS 1, 3]	18
Chang * [58]	OPC	C	0.66	307	100dx200 [cy]	12.3	40	[Mix, NaCl,)] / [(Add), 1.28%bwce (Re), 3800ppm bwco]	Re	Ti	[$Ca(OH)_2$] / [tank] [NaOH] / [tank]	[G] / [A/m² CS 1.88, 18.8]	7, 14, 21, 28, 35, 42
Abdelaziz [44]	OPC SRC	M	0.5	-	80x80x300 [pr]	10	35	[Mix, NaCl 1.2%bwce + W/D NaCl 1 year] / [(De 0-35mm) (t) 4.2-1.7%bwce]	Re	SS mesh	[Ca(OH)2] / [tank] [TW] / [tank]	[G] / [A/m² MS 1, 2]	14, 28, 56

Climent [91]	CEM II/A-L	C	0.5	342	100dx100 [cy]	Re d10 deformed	45	[Mix, NaCl] / [(Add) 1%bwce] / (Add) (t) 0.55%bwce]	Re	aTi-RuO_2, Graphite	[DW] / [immersion or sandwich]	[Gi] / [A/m² CS 3]	?
Climent [91]	CEM II/A-L	C	0.5	342	200dx1000 [co]	6 Re d12 deformed	30	[Mix, NaCl] / [(Add), 1%bwce] / [(De) (t) 1.3-1%bwce]	Re≠	aTi-RuO_2, Graphite	[TW] / [immersion or sandwich]	[Gi] / [A/m² CS 5]	280
Perez [38]	CEM II/B-L	C	1(?) 0.5	195 342	180x180x120 [bl]	2	60	[Mix, NaCl] / [(Add), 1.2%bwce (Av), (t) 0.24, 0.31%bwco]	Re steel mesh	Ti-RuO_2 or CCP	[DW] / [top pond]	[G] / [A/m² CS 2]	139h 556h

* 5 V corresponds to 18.8A/m² CS

Table 4: Published laboratory studies on ECE treatment performed using a current control mode (continuous G or intermittent Gi) with current densities expressed as A/m² of steel rebar surface (RS).

First author [Reference]	Binder	Sample	w/c ratio	Cement (kg/m^3)	Dimensions (mm) [shape]	Steel rebar (diameter mm)	Cover (mm)	chloride content before ECE [mode] / [content Cl^-]	Cathode	Anode	Electrolyte	Current control mode (G or Gi] / [A/m^2 RS]	Duration (days)
Monteiro [81]	-	C	0.45 0.65	500 380	1180x560x1200 [pr] 1220x960x1200 [pr]	6.3	20, 40	[SS, 4m] / [(De 0-40mm), (t), 1.5, 0.5%bwce]		SS mesh	[TW] / [tank]	[Gi] / [RS 1]	2w ON, 1w OFF, 2w ON, 1w OFF, for a total cycle of 6w
Elsener [48]	-	C	0.5 0.6	340 303	320x245x70 [bl]	8	40, 50	[Cycles+I, NaCl] / [(De 0-50mm), (t) 0.45-1.45%bwce, (f) *0.02-0.15%bwce]*	SSR#	SS plate	[TW] / [foam plastic]	[Gi] / [RS 2]	18 (7d ON,12d OFF, 4d ON, 3d OFF, 7d ON)
Miranda [59, 60]	-	M	0.5	-	1330x1330x70 [sl]	8	31	[Mix, $CaCl_2$ 3% bwce] / [(Add), (De 0-60mm), 1000-3500 ppm]	R	Ti	[$Ca(OH)_2$] / [cloth soaked]	[G] / [RS 5] 1 or 2 sides	42 (per side)
Zhu [41]	OPC	C	0.32 0.4 0.5	480	100dx100 [cy]	10	45	[Mix, NaCl] / [(Add), 1.82%bwce]	R	CFRP	[$Ca(OH)_2$] / [tank]	[G] / [RS 0, 1, 2, 3]	28
Siegwart [47]	OPC	C	0.4	460	1000x100x100 [pr]	22	39	[Mix, NaCl] / [(Add), 1.5%bwce]	R+PSC	Ir	[W] / [bath]	[G] / [RS 1]	42

Kim [84]	OPC	C	0.40	-	70x70x150 [pr]	10	30	[Mix, NaCl] / [(Add), 1.82%bwce]	R	Ti	none	[G] / [RS 0.25, 0.50, 0.75]	28
Orellan [57]	OPC	C	0.45	400	110dx100 [cy]	10	50	[W/D, NaCl, 10m] / [(De 0-50mm), (t) 4.5-6.7%bwce, (f) 3.75-5.7%bwce] / [(Re), (t) 5 %bwce, (f) 4.5 %bwce]	R	Ti	$[Ca(OH)_2]$ / [tank]	[G] / [RS 1]	50
Polder [43]	OPC	C	0.4 0.54	420 300	500x100x100 [pr]	5	15, 30, 46	[SWI, 16y] / OPC 0.54 [(Av), (t), 2.7-4.4%bwce] / OPC 0.4 [(Av), (t), 2.5-4.1%bwce]	R	aTi	$[Ca(OH)_2]$ / [tank] [TW] / [tank]	[G] / [RS 1, 4]	39
Stoop [51]	OPC	C	0.4 0.54	420 300	200x100x100 [pr]	5	15, 30, 46	[SWI, 16y] / OPC 0.54 [(Av), (t),2.7-4.4%bwce] / OPC 0.4 [(Av), (t), 2.5-4.1%bwce]	R	aTi	$[Ca(OH)_2]$ / [bath] [TW] / [bath]	[G] / [RS 1, 4]	39
Buenfeld [35]	OPC	C	0.5	400	102dx254 [cy]	32	35	[Mix, NaCl] / [(Add), 1.5%bwce]	R	cTi	[W] / [cellulose fibre] / [tank]	[G] / [RS 0.75]	up to 112
Ihekwaba [78]	OPC	C	0.5	370	150x150x150 [cu]	20	65	[Mix, NaCl] / [(Add), 1.7, 3%bwce]	R	aTi	$[Na_3BO_3]$ / [tank]	[G] / [RS 9.5, 28.6]	56
Tang [90]	OPC	C	0.57 0.60 0.70 0.72	285	200x200x25 [sl]	16	50	[Mix, NaCl] / [(Add), 0, 0.2, 0.7 and 2%bwco]	R	Ti	[TW] / [running]	Gi RS - 0.7; 1.0 and 1.3	56 2w ON then 2w OFF for a total of 8w
Vennesland [77]	OPC	C	0.7	-	- [pr]	-	-	[Mix, NaCl] / [(Add), 2%bwce]	R	Ti	[W] / [tank]	[G] / [RS 1.6, 4, 8]	7, 14, 26, 56

Polder [85]	OPC	C	0.45 0.55 0.65	380 345 310	500x400x140 [pr]	8 16	15 30	[W/D, NaCl, 20 to 77w] / [(Av 0-45mm), (t) 0.3-1.6%bwce]	Re#	aTi	$[Ca(OH)_2]$ / [tank] [TW] / [tank]	[G] / [RS 1, 4]	21-42
Orellan [54]	OPC	C	0.7	300	50dx100 [cy]	10	20	[W/D, NaCl, 10m] / [(De 0-20mm), (t) 4.4-6.5%bwce, (f) 2.2-3.8%bwce]	R	Ti	$[Ca(OH)_2]$ / [tank]	[G] / [RS 1]	10, 20, 30, 40, 50
Fajardo [45]	OPC	C	0.6	310	50dx110 [cy], 110dx110 [cy]	10	20, 50	[W/D, aSW, 10m] / [(Av) 4.6%bwce]	R	Ti	[DW] / [tank]	[G, Gi] / [RS 1]	90 (G), 5d ON 2d OFF until 90d (Gi)
Swamy [55]	OPC	C	0.6 0.75	350 383	100x 165x2300 [be]	6mm² (sq)	20	[Mix, NaCl] / [(Add), 2.4, 6%bwce] / [(De), (t) 1.5-8.2%bwce]	R	TiPt (2 sides)	$[Ca(OH)_2]$ / [tank]	[Gi] / [RS 1]	45
Yeih [82]	OPC	C	0.66 0.4	295 488	100dx200 [cy]	9.5	25 50	[Mix, NaCl] / [(Add), 1.8%bwce] / [(Re), (f), 1.7%bwce]	R	Ti	[NaOH] / [tank]	[G] / [RS 1, 2, 4, 8]	7, 14, 21, 28, 35, 42
Carmona [40]	OPC	C	0.65	250	180x180x80 [sl]	15.8	20	[Mix, NaCl] / [(Add) 2%bwce] / [(Av), (t) 1.97%bwce]	R#	GCP	[DW] / [top pond]	[G] / [RS 3.4]	8.8
Tissier [69]	OPC	µC	0.65	387	100dx40 [cy]	5	17.5	[Mix, NaCl 5% bwce + Carbonation] [Carbonation + W/D NaCl]	R	TiPt	$[Na_2B_4O_710H_20]$ / [tank]	[G] / [RS 1]	3, 7,14, 28, 56
Marcotte [79, 80]	OPC	M	0.5	-	51dx141 [cy]	9.5	20, 65	- [Mix, NaCl] / [(Add), 2%bwce] = [(Add), (-), 0.25%bwmo] - [Ing, NaCl] /	R	TiPt	$[Na_3BO_3]$ / [tank]	[G] / [RS 8.37]	56

								[(Add), (-), 0.38%bwmo] - [Mix, NaCl + Ing, NaCl] / [(Add), (-), 0.36%bwmo]					
Shan [83]	OPC	M	0.45 0.55	M	40x40x160 [pr]	10	15	[Mix, NaCl] / [(Add), 2%bwce]	R+Ti	Ti	$[Ca(OH)_2]$ / [tank]	[G] / [RS 2]	28
Bouteiller [67]	CEM II/A-S (6-20%slag)	C	0.53	400	2500x200x200 [be]	6	16, 41	[SWT, 40y] / [(De 0-41mm), (t) 6.6-3.8 %bwce, (f) 3.9-1.3%bwce]	R#	TiPt	$[Na_2B_4O_7]$ / [top pond]	[G] / [RS 1]	56
Kim [84]	GGBS (40%OPC+ 60%GGBS)	C	0.40	-	70x70x150 [pr]	10	30	[Mix, NaCl] / [(Add), 1.82%bwce]	R	Ti	none	[G] / [RS 0.25, 0.50, 0.75]	28
Tissier [69]	CEM III/A (65%slag)	µC	0.65	387	100dx40 [cy]	5	17.5	[Mix, NaCl 5% bwce + Carbonation] [Carbonation + W/D NaCl]	R	TiPt	$[Na_2B_4O_710H_20]$ / [tank]	[G] / [RS 1]	3, 7,14, 28, 56
Stoop [51]	BFSC (70%slag)	C	0.4	420	200x100x100 [pr]	5	15, 30, 46	[SWI, 16y] / BFSC 0.4 [(Av), (t), 1.0-2.8%bwce]	R	aTi	$[Ca(OH)_2]$ / [bath] [TW] / [bath]	[G] / [RS 1, 4]	39
Polder [43]	BFSC (70%slag)	C	0.4	420	500x100x100 [pr]	5	15, 30, 46	[SWI, 16y] / BFSC 0.4 [(Av), (t), 1.0-2.8%bwce]	R	aTi	$[Ca(OH)_2]$ / [tank] [TW] / [tank]	[G] / [RS 1, 4]	39
Bertolini [61]	BFSC (70%slag)	C	0.4	420	500x100x100 [pr]	5	15, 30, 46	[SWI, 16y] / BFSC 0.4 [(Av), (t), 1.0-2.8%bwce]	R	aTi	$[Ca(OH)_2]$ / [tank] [TW] / [tank]	[G] / [RS 1, 4]	39
Polder [85]	BFSC (70%slag)	C	0.45 0.55 0.65	380 345 310	500x400x140 [pr]	8 16	15 30	[W/D, NaCl, 20 to 77w] / [(Av 0-45mm), (t) 0.3-1.6%bwce]	Re#	aTi	$[Ca(OH)_2]$ / [tank] [TW] / [tank]	[G] / [RS 1, 4]	21-42

Polder [63, 65]	BFSC (70%slag)	C	0.55	-	150d [cores]	16	30, 45	[Splash, 8y] / [(De 0-58mm), (t), 1.45-0.02% bwce]	R	aTi	$[Ca(OH)_{2}]$ / [bath] $[Na_2CO_3]$ / [bath]	[G] / [RS 0.7, 1, 4]	19, 21, 42
Kim [84]	Ternary (40%OPC+ 40%GGBS+ 20%FA)	C	0.40	-	70x70x150 [pr]	10	30	[Mix, NaCl] / [(Add), 1.82%bwce]	R	Ti	none	[G] / [RS 0.25, 0.50, 0.75]	28

4.1 Chloride extraction

4.1.1 **Ion migration**

The current flowing from the rebar to the anode allows the migration of ions (anions Cl^-, OH^- migrate from the rebar to the external anode and cations Na^+ and K^+ migrate towards the rebar). The extraction of chloride ions from the vicinity of the rebar and from the concrete cover is expected to decrease corrosion. The efficacy of chloride extraction depends on the chloride transport number t_{Cl} that depends on the chloride concentration and on the amount of other ions present in the medium. The transport numbers of the different ions [26] are indicated in Table 5.

Table 5: Transport numbers t of ions (from [26]).

Ion	OH^-	Cl^-	Na^+	K^+	Ca^{2+}
C (mol/L)	0.5	0.2	0.035	0.58	10^{-4}
t	0.62	0.1	0.01	0.27	0

The quantity of chloride ions extracted (Cl_{ext}) per m² per second is obtained using Equation 9.

$$Cl_{ext} = i.t_{Cl.}G/F \qquad \textit{Equation 9}$$

Where i is the current density, t_{Cl} is the transport number of chlorides, G is a geometrical constant (*f* (rebar-concrete interface)) and F is the Faraday constant.

Assuming that the current remains constant, the quantity of chloride ions extracted will vary according to the transport number of the different ions present in the concrete pore solution [63, 86].

4.1.2 **Efficacy of ECE**

The authors calculated the efficacy of chloride extraction (Cl_{eff}) according to Equation 10 based on the available published results using current control mode.

$$Cl\ Efficacy\ = 100\ x \frac{Chloride\ content\ before\ ECE - Chloride\ content\ after\ ECE}{Chloride\ content\ before\ ECE} \quad \text{Equation 10}$$

Figure 3 presents the efficacy of ECE using a current control mode and current densities expressed in surface of steel rebar (RS) applied to reinforced concretes or mortars contaminated by chloride ions from different sources (studies from Table 4). The authors calculated the chloride extraction efficacy close to the rebar (Re) based on free (f) or total (t) chloride ions. In some papers, it was not clear if the chloride concentration after ECE was determined close to the rebar or averaged from the concrete cover. We therefore considered the data as the average chloride extraction in the concrete (Av). Figure 3 plots chloride efficacy versus total charge in the range of 168 to 39 782 A.h/m² RS. The total charge is indicated on the abscissa axis, as well as duration and applied current density, which are noted in brackets. The authors highlight the need to include these three types of information (applied current density, duration and total charge) as part of the design. When the efficacy is related to a parameter of the ECE set-up, it is indicated in Figure 3 as *f* (parameter). Finally, results pertaining to OPC binder are presented on the right-hand side of the graph, whereas the results for the other binders are presented on the left.

For OPC binder, which is the main cement studied in literature, chloride extraction efficacy in areas located close to the rebar increases from 25 to 97% as the applied total charge increases from 168 to 1344 A.h/m² RS. The application of higher total charges ranging from 2016-39 782 A.h/m² RS with current densities above 2A/m² RS can cause detrimental mechanical degradations [59].

Kim et al. [84] found that an increase in the total charge resulted in a decrease in the chloride concentration around the steel rebar (i.e., at a depth of 27-30 mm) for current densities that varied from 0.25 to 0.75 A/m² RS and were applied for 28 days, whatever the binder concerned. Yeih et al. [82] studied the efficacy of chloride extraction using different current densities varying between 1 to 8 A/m² RS and different durations (7, 14, 21, 28, 35, 42 days) applied on different reinforced concretes (w/c ratio (0.4 or 0.66)) and concrete covers (25 or 50 mm). They proposed a unified regression curve describing the evolution of the chloride content near the rebar versus the total charge [82]. Looking at the overview of many data (Figure 3), the authors can recommend for ordinary Portland cement a total charge of 1 000 – 2 000 A.h/m² RS for achieving a 80% efficacy.

On the left-hand side of Figure 3, chloride extraction is also shown to increase with total charge for non-OPC binders such as BFSC [43, 51], binary (40%OPC+60%GGBS) or ternary binders (40%OPC+40%GGBS+20%FA) [84] and CEM II/A-S (6-20% slag) [67].

Finally, the results of Tissier et al [68, 69, 105, 106] are original because the reinforced concretes made with OPC or CEM III/B binders were contaminated by both chloride ions and carbonation prior to ECE. The excellent extraction efficacy reported was due to the free form of most chloride ions after carbonation and the thinness of the concrete cover (17 mm).

From a practical perspective, clear criteria are required to ensure that a satisfactory chloride extraction has been achieved. The literature proposes the following criteria: (i) attaining a certain percentage of extracted chlorides and/or (ii) obtaining chloride content values in the vicinity of the rebar that are below a threshold content, such as 0.4% bwce.

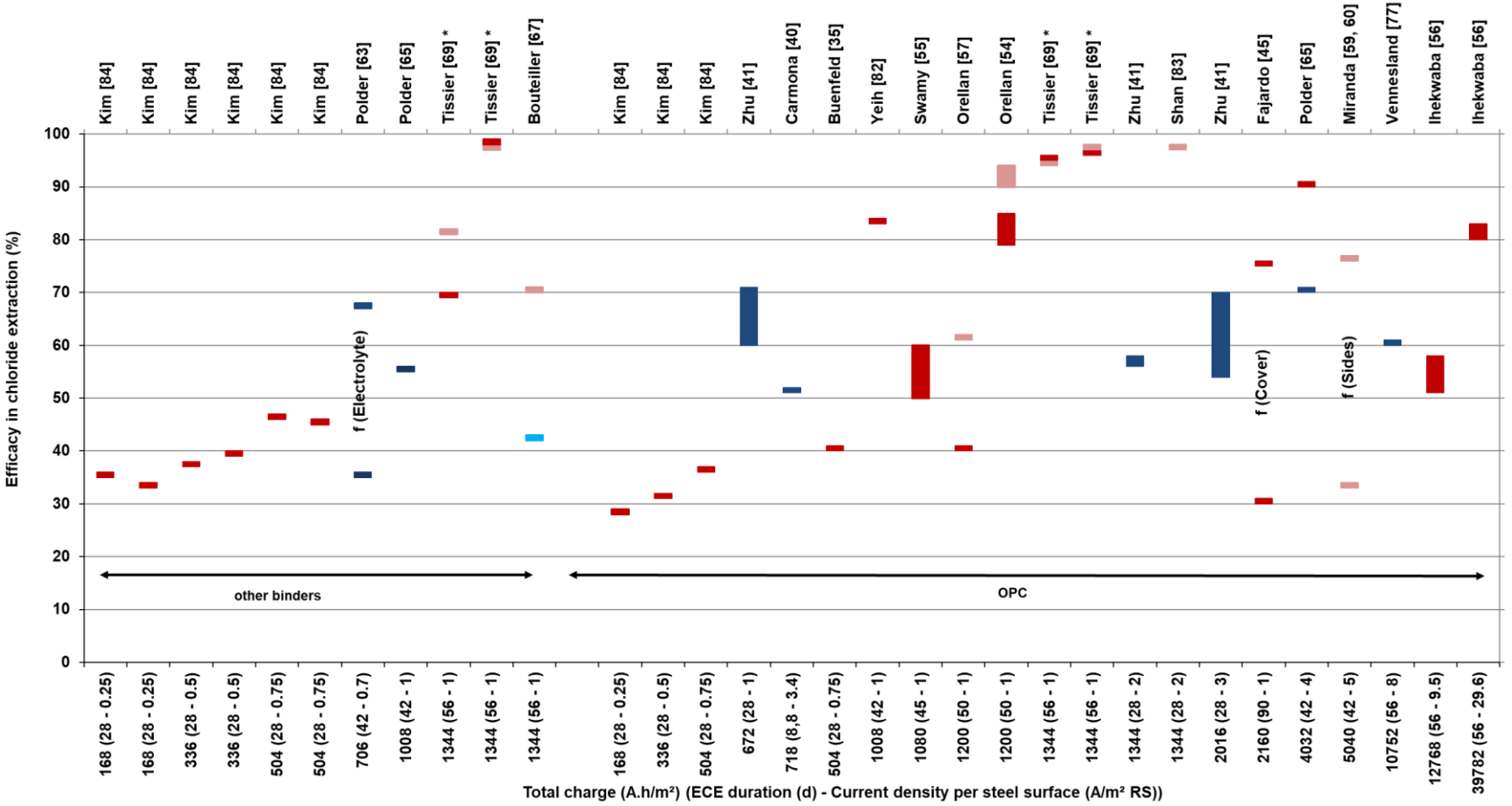

Figure 3: Efficacy of chloride extraction applied to reinforced concrete specimens (chloride concentration close to the rebar (Re for rebar (red)) or chloride concentration of concrete cover (Av for Average, (blue)). Efficacy was calculated based on total or free chloride ions (shown in dark or pale colours, respectively). A star following the reference (top axis) indicates that the reinforced specimens were contaminated by both chlorides and carbonation prior to ECE.

4.1.3 **Influencing factors on chloride extraction**

The authors used the published results to analyse the factors influencing chloride extraction efficacy, i.e. concrete properties, chloride characteristics, electrolyte type and duration of treatment.

4.1.3.1 Concrete properties

Concrete properties such as w/c ratio, concrete cover, binder type and contamination can have an impact on ECE efficacy.

4.1.3.1.1 Water/cement ratio

Polder et al. [43, 85] found no impact on the extraction efficacy when ECE was applied to concretes with different w/c ratios (0.4 up to 0.65). Zhu et al. [41] did not find a clear trend between w/c ratios (0.32, 0.4 and 0.5) and chloride extraction efficacy. Other authors [48, 81-83, 104] found that specimens with high water/cement ratio exhibit a higher chloride extraction efficacy due to their greater porosity. In his thesis, Orellan [104] showed that the w/c ratio (0.45 or 0.7) influences the efficacy of the average chloride extraction value but does not influence chloride extraction efficacy in the vicinity of the rebar. Swamy et al.[55] observed that the efficacy of chloride removal from reinforced concrete beams was neither consistent nor concurrent with the w/c ratio (0.6 or 0.75).

4.1.3.1.2 Concrete cover

Several authors found that a thinner concrete cover permits a more effective chloride extraction due to lower electrical resistance and shorter chloride transport distances [45, 81, 104]. As an example, Fajardo et al. [45, 107] obtained a chloride extraction efficacy that varied between 75% and 30% respectively for specimens with a 20 mm and 50 mm concrete cover after applying a current density of 1 A/m^2 RS for 90 days.

4.1.3.1.3 Binder type

When Polder [43, 63, 85] applied ECE to reinforced concretes cast with OPC or BFSC, no impact on chloride extraction efficacy was observed. However, it is important to note that the driving voltages needed to obtain the required current density were significantly different given the resistivity of each binder: whereas OPC specimens with resistivity in the range of 100-200 ohm.m required 3-5 V to obtain a current density of 1 A/m² RS and 16-30 V for 4 A/m² RS, BFSC specimens with resistivity in the range of 400-1000 ohm.m needed 16 V (1A/m² RS) and 38 V (4A/m² RS) to obtain the same density. Kim et al. [84] found that it was easier to remove chlorides from concretes made with Portland cement than from cements containing GGBS (40%OPC+60%GGBS) or ternary mixtures (40%OPC+40%GGBS+20%FA). A higher binding capacity of chlorides in concrete may therefore result in a lower chloride extraction efficacy.

4.1.3.1.4 Concrete contamination

Tissier et al. [68, 69] obtained a chloride efficacy of above 90 % for specimens that had been carbonated and chloride contaminated (via different methods) prior to ECE. The high efficacy was obtained for all binder types (OPC or CEM III/A) This very good result was explained by two factors, namely the thinness of the concrete cover (17.5 mm), and the fact that carbonation tends to release the bound chlorides into the matrix as free chlorides.

4.1.3.2 Chloride characteristics

The efficacy of ECE treatment is affected by chloride characteristics, namely the method of contamination, the initial content and the type of chlorides analysed (total, free).

4.1.3.2.1 Method of chloride contamination

Chloride contamination can result from different methods:

- Mix addition is the most common means of contamination, because the chloride contamination is immediate. Studies have used NaCl (Cl^- range 1 % - 8 % bwce) [35,

40, 41, 46, 47, 55, 61, 69, 75, 77, 79, 80, 82-84] or $CaCl_2$ (Cl^- range 2.35 % - 3 %bwce) [59, 60],

- Wetting/drying cycles in NaCl solution [54, 57, 69] or in artificial sea water [45],
- Submersion in NaCl [48],
- Salt spray chamber [81],
- Outdoor exposure conditions or real structures: splash zone (8 years) [43], sea water (16 years) [43, 51], sea water tide (40 years) [67].

Marcotte et al. [79] used rather strong ECE conditions (8.37 A/m² RS for 56 days) and found that admixed chlorides are more difficult to extract than ingressed chlorides: chloride extraction efficacies in the vicinity of the rebar were respectively 79 %, 74 % and 3 % when chloride contamination was achieved by mix + ingress additions (initial Cl^- 0.36 % bwmo), ingress (initial Cl^- 0.38 % bwmo) or mix (initial Cl^- 0.25 % bwmo). The study did not clearly indicate whether the chloride was free or total.

4.1.3.2.2 Initial chloride content

Several authors stated that chloride extraction is greater for concretes with a higher initial chloride content [43, 48, 78, 90, 108, 109]. Tang [90] concluded that ECE treatment has a limit beyond which the further removal chloride becomes difficult. Finally, no correlation was found between the initial chloride and the remaining chloride after treatment.

4.1.3.2.3 Type of chloride analysed (free, total or bound)

Elsener [48] described the equilibria between chemically bound, physically adsorbed and free chlorides in the ECE treatment (Figure 4). Part of the remaining chloride measured was in an insoluble form after one or more ECE treatment cycles [48]. On the other hand, Orellan et al. [54] found that ECE efficacy on concrete was slightly affected by the C_3A content (4.3 or 9.5%) in OPC cement because only a part of the bound chloride ions was released.

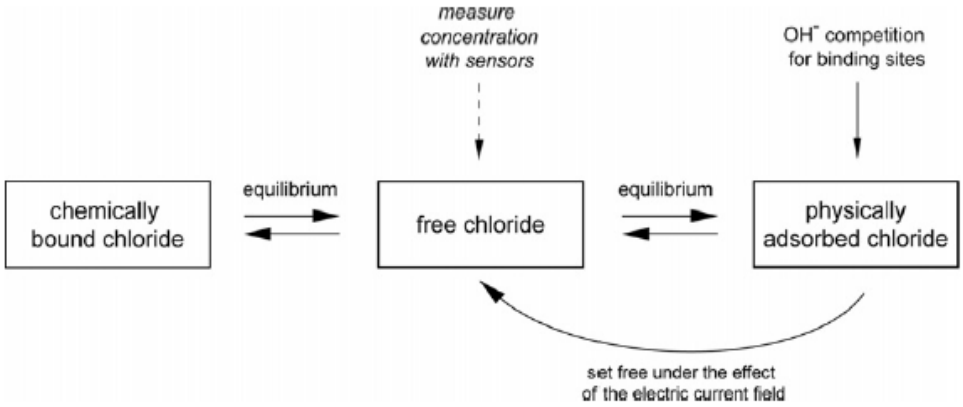

Figure 4: Equilibria between bound, adsorbed and free chloride in the ECE treatment (from [48]).

4.1.3.3 Electrolyte

A study by Monteiro et al. [81] reported that the use of tap water as the electrolyte caused an increase in the resistance and thus also in the polarization of the anode. Polder found no difference in chloride extraction efficacy between tap water and saturated calcium hydroxide solutions [43]. Polder et al. [63] compared sodium carbonate (Na_2CO_3) and calcium hydroxide ($Ca(OH)_2$) solutions, and found that extraction with calcium hydroxide was more efficient. The use of $Ca(OH)_2$ electrolyte produced a 67% extraction of total chlorides (0.7 A/m² RS, 42 days), whereas the proportion of chloride extracted over the same period with Na_2CO_3 electrolyte was respectively only 35 % and 55 %, with current densities of 1 and 4A/m² RS respectively. Using an electrolyte with Na causes Na^+ migration into the pore solution, thus reducing the Cl^- transport number. Bouteiller et al. [67] used sodium tetraborate ($Na_2B_4O_7$) solution to extract chloride from a reinforced concrete beam (4 cm cover) that had been exposed to a marine environment in France for 40 years. The treatment allowed the removal of up to 70 % of the free chlorides located in the vicinity of the rebar, 50 % of those in the intermediate cover and 5 % of those located at the surface of the concrete. In his thesis, Orellan [104] used identical experimental conditions and ranked different electrolytes from the most to the least effective

in terms of chloride extraction, namely tap water (60 %), sodium-based electrolyte (54 %), and calcium and lithium-based electrolytes (40 %).

4.1.3.4 Duration

Although most ECE treatments last 28 or 56 days, other periods were reported ranging from 3-112 days. The extraction of chlorides was reported to be at its highest at the beginning of the treatment [35, 41, 45, 48, 54, 83, 90]. This phenomenon is easily explained: at the beginning of the process, chloride ions are the main negative ions that migrate through the current flow. After some time, the chloride transport number decreases (due to the extraction) and the hydroxyl ions that are formed at the cathode compete as negative charge carriers. Tang et al [90] suggested that there is a limit beyond which the further removal of chloride ions becomes difficult.

4.2 Steel corrosion mitigation

The characterization of ECE efficacy requires evidence that steel rebar corrosion has been mitigated following the extraction of chloride ions (Table 4). Table 6 presents published results on resistivity, half-cell potential and corrosion current density.

The resistivity of the concrete is a key parameter that affects both corrosion behaviour (a concrete resistivity below 10 kohm.cm is linked to a high corrosion probability [110]) and ECE treatment conditions (the voltage increases with resistivity).

Half-Cell Potential (Ecorr) indicates the probability of corrosion. When the measured potential is more positive than -250 mV CSE, there is a 90 % probability that rebar corrosion is absent [102]. An increase of potential values after treatment indicates a lower probability of corrosion [111]. Miranda et al. [59, 60] warned, however, that this approach might give rise to serious errors when evaluating the active or passive state of the rebar if the saturation degree of the pores is not identical. A significant ennobling of potentials after the application of ECE cannot

therefore be deemed sufficient evidence to demonstrate the efficacy of the treatment; evidence of a decrease in corrosion current density is also needed.

Corrosion current density (Icorr) is an instantaneous value. A value in the range of above 1, 1-0.5, 0.5-0.1 or less than 0.1 µA/cm² respectively indicates a high, moderate, low or negligible corrosion level [103]. A decrease in corrosion current density after treatment will therefore highlight mitigation of corrosion.

The efficacy of ECE treatment is dependent on the mitigation of corrosion. The latter must be evaluated by comparing electrochemical characterizations before ECE, immediately after ECE and after the depolarization of the steel rebar.

It is important to highlight the following conditions:

a) The comparison of electrochemical results is only valid if the "steel rebar in concrete" system is at its equilibrium (i.e.; not polarized) and
b) The comparison of electrochemical results should only be carried out for the same sample if conditions of temperature, humidity and oxygen availability remain identical.

Finally, it is important to note that increasing Ecorr and decreasing Icorr can indicate a mitigation of corrosion, but this in itself does not suffice to conclude that the rebar is passivated. No clear demonstration of re-passivation has been published to date.

222 *Table 6: Published results for electrochemical chloride extraction (italic values for non-treated samples)*

223

Reference	Condition	Resistivity		Half-cell potential					Corrosion current density			
		Before ECE (kohm.cm)	Just after ECE (kohm.cm)	Reference Electrode	Before ECE (mV)	Immediately after ECE (mV)	Depolarization time (days)	After depolarization (mV)	Before ECE (µA/cm²)	Immediately after ECE (µA/cm²)	Depolarization time (days)	After depolarization (µA/cm²)
Buenfeld [76]		<206> (223; 89; 255; 255)	<1245> (541; 509; 745; 3183)									
Polder [43]				Ag/AgCl	*-400 to -260* -420 to -230		39	*-400 to -330* -130 to +140				
Marcotte [79]	Ingressed			SCE	-	-1200		-375 1year after ECE (but no O2)	<0,1 (18 (5% surface))	7		
	Admixed + ingressed								<0,1 (or 27 (5% surface))	19		
Monteiro [81]				SCE		-900	30	-157				
Orellan [54]				SCE	-647 (10d) -626 (30d) -589 (50d)	-1123 (10d) -1075 (30d) -1053 (50d)	61	-335 (10d) -153 (30d) -172 (50d)	5.76 (10d) 6.73 (30d) 6.26 (50d)	5.02 (10d) 5.08 (30d) 4.70 (50d)	61	2.65 (10d) 2.19 (30d) 2.89 (50d)
Fajardo [45]	Cover 20			SCE	-570	» -1000 (90d)	42	-380 (42d) up to -300 (147d)	9		90	4

	Cover 50				-600	» -1000 (90d)	36	-400 (36d) up to -400 (142d)	3		90	2
Bouteiller [67]	B4B3			SCE	*» -500;* -500	-800 to -700	60	*-550 to -500* ; -600 to -500	*» 0.6*; 0.6 to 1	2 to 3.5	60	*» 0.6*; 2 to 2.5
	B2B1				*-600 to -550* -650 to -600	-900 to -700	60	*-600 to -550* ; -550 to -500	*» 0.6*; 0.6 to 1.5	3 to 4	60	*» 0.6;* 2 to 2.75
Yeih [82]	Cover 25	6.3 to 9.6	11.1 to 13.3	CSE	-500 to -450	-1110	60	» -230 ; -200	0.45		60	1.04
	Cover 50	7.1 to 10.7	10.8 to 13.3		-450 to -390							0.17
Miranda [59, 60]				SCE	-650	-350	90	-50	» 10	» 10	90	» 10
* Elsener [48]				SCE		-1100 to -800	12 (first current off phase)	-600 to -400				
Shan [83]				SCE		» -1225	30	-200 (after 30d of immersion after ECE)	10		30	<0.1
Kim [84]	OPC			SCE	-650		?	-475	110		?	8.0
	GGBS				-500			-300	100			2.5
	Ternary				-700			-320	100			4.3
Tissier [68, 69, 105, 106]	OPC, GGBS			SCE	-600 to -550	-1200	70-90	-450 to -200 (14d) -300 to -150 (28d) -250 to -160 (56d)	10	10	70-90	0.01 to 1 (14d) 0.04-0.1 (28d) < 0.03 (56d)

224 * Stainless steel rebar

225

4.2.1 **Electrical resistivity**

Polder [43] states that no major changes occurred during the ECE process, resulting in the relatively stable driving voltages needed to obtain the fixed currents. In contrast, Miranda [59] found that the voltage increased from 7 to 40 V due to the progressive elimination of the chlorides and the increase in mortar resistivity. Buenfeld [76] and Yeih [82] also reported an increase in resistivity due to the extraction of chlorides.

Despite obvious differences between studies, resistivity is not conclusive with regard to corrosion mitigation.

4.2.2 **Half-cell potentials**

Half-cell potential is an interesting measurement for evaluating ECE. Immediately after the treatment, potential values are highly electronegative due to the polarization of the steel rebar [45, 48, 54, 67, 68, 79, 81-83]. This confirms that the treatment has been applied correctly. It is impossible to predict how long it will take for the rebar to depolarize, and the results from Table 6 show this process to take 1 to 4 months. As depolarization occurs over time, the half-cell potential gradually becomes nobler [45, 54, 59, 60, 69, 82, 84] and reaches a stable value. If the latter is less electronegative than the value obtained prior to ECE, it can indicate a decrease in corrosion activity that is consistent with the reduction in chloride ions around the rebar.

4.2.3 **Corrosion current density**

Before ECE, corrosion current densities are in the range of 5 to 27 µA/cm² (Table 6), with the highest density reported at 100 µA/cm² in reference [84]. Higher corrosion current densities immediately after ECE can be explained by (i) the polarization of the steel, (ii) the accumulation of hydroxyl ions at the rebar (iii) a decrease in oxygen at the rebar/mortar interface [79]. After depolarization (over 1 to 4 months), decreases in corrosion current density ranging from fifty percent [45, 54] to even 1 or 2 orders of magnitude were obtained [69, 83, 84].

4.2.4 **Influencing factors on steel corrosion mitigation**

4.2.4.1 ECE conditions

At a chosen ECE current flow, the increase in duration (and thus an increase in the total charge) increases the efficacy of corrosion mitigation [68, 82, 83]. Kim et al. [84] studied the optimisation of current density applied during ECE (varying from 0.25 to 0.75 A/m² RS). They observed that an increase in current density resulted in an increase in the half-cell potential and a decrease in the corrosion rate.

4.2.4.2 Binder types

Kim [77] reported that in ECE treatment carried out at 0.75 A/m² RS, the corrosion rates were 2.5 and 4.3 µA/cm² for GGBS and ternary mixes respectively. These rates are substantially lower than those found for OPC mix (8 µA/cm²), presumably due to a lower supply of water and oxygen (i.e. cathodic reactions) through concretes that contain pozzolanic materials.
Tissier et al. [68, 69] found that corrosion rates decreased by two orders of magnitude for micro-concrete specimens that had been contaminated by both carbonation and chloride ions prior to ECE, whatever the binder used (OPC or CEM III/A (max 68% slag)).

4.3 Evolution of pH

The evolution of pH has been studied in two main contexts, namely the pH of the electrolyte, which is central to determining a suitable treatment, and the pH of the rebar/concrete interface, which should provide evidence of improved treatment efficacy.

4.3.1 **pH of the electrolyte**

The pH of the electrolyte has been studied throughout the ECE treatment to ensure that the electrolyte does not become acid or differ too greatly from the concrete pH. Maintaining a high

pH also allows monitoring of Cl in the electrolyte; at low neutral to pH it is oxidised and disappears as Cl_2 (Equation 8).

Water has a pH of 7 and is not a suitable electrolyte; during ECE, its pH will decrease to 3.5 (and produce a chlorine gas smell) without circulation, whereas the use of circulation will lead pH to decrease to 4-6 [43], 6.5 [45] and 5-5.5 [40]. Using calcium hydroxide ($Ca(OH)_2$) electrolyte (pH=12.5), Polder et al. [43] observed a local acidification in which the pH dropped to about 5.5. While performing ECE on beams, Swamy et al. [55] also observed the acidification of the calcium hydroxide electrolyte and chlorine smell in the laboratory for 2-8 weeks. A renewal of the calcium hydroxide electrolyte prevented these reactions (stable 12.5 pH and no chlorine smell).

4.3.2 **pH of the rebar/concrete interface**

During ECE, pH values increase at the rebar/concrete interface because of the formation of hydroxyl ions at the cathode (Equation 5). This should play a role in corrosion mitigation and durable protection. In an ECE set-up with water electrolyte, Buenfeld [35] used a phenolphthalein indicator and found that a uniform purple staining was obtained 48 h after the spraying on areas that included the concrete adjacent to the rebar. Zhu et al. [41] used calcium hydroxide electrolyte and found an increase in pH close to the rebar. The use of sodium hydroxide (NaOH 0.1M) solution in a study by Yeih et al [82] led pH values to increase to 12.4, with the total charge attaining 8064 A.h/m². These authors suggested that a pH value of 11.5 in the vicinity of the rebar might be one of the performance criteria for determining ECE efficacy. Finally, in the case of a realkalisation treatment, an increase in pH at the rebar was also consistent with halting the corrosion [112, 113].

4.4 Visual observations and gravimetric analysis performed on the rebar

Table 7 summarizes the published results of visual observations and gravimetric analysis before and after ECE treatment.

300

301 *Table 7: Visual observations and gravimetric analysis before and after ECE*

302

Reference	Before ECE	After ECE
Buenfeld [35, 76]	- Substantial amount of corrosion particularly associated with entrapped air voids in the concrete at the surface of the rebar	- Less evidence of corrosion - A rusty brown coloration was revealed beneath the brittle black surface layer - Air voids were partially filled with a soft black deposit
Marcotte [79]	- Corrosion pits covering 5 % of the surface for samples exposed to ingressed chlorides - No observable corrosion pits for samples with admixed chlorides	- Fine white products cover the steel surface - All steel samples had large areas (50 to 100 % of the surface area) of black-brown discolouring
Vennesland [77]		- Blackish deposits at the concrete/rebar interface
Ihekwaba [78]		- Accumulation of whitish deposits around the rebar treated at high impressed cathodic current densities
Orellan and Fajardo [45, 57]	- Substantially corroded	- Slight white deposit on the surface of the rebar
Miranda [59, 60]	- Corrosion products have a reddish colour	- Corrosion products are dark brown

Kim [84]	- Weighable rust was present on the surface of steel rebar rather than pit corrosion	- Rust stain was still present on the surface rather than pit corrosion - Mass loss: 0.48-0.55 %, 0.27-0.32 % and 0.26-0.29 % respectively for 2.5, 5 and 7.5 µA/cm² RS (Mass loss 0.79-0.88 % without ECE treatment).

Corrosion products are observed before treatment: they do not usually cover the entire surface of the steel rebar and they do not always appear as pits, although corrosion is induced by chloride ions (Table 7). After ECE, whitish deposits [57, 78, 79] or black-brown products [35, 59, 76, 77, 79] were observed on the surface of the steel rebar (Table 7). Ihekwaba [78] reported that the whitish deposits are due to the alkali hydroxides formed as a result of cathodic action during ECE application. Vennesland [77] studied the blackish products and found that ECE induced an accumulation of cations (sodium and potassium) close to the rebar. Marcotte [79] explained the black products by the formation of magnetite: although $HFeO_2^-$ and FeO_2^{2-} ions are likely to have been formed in the high pH/ low potential conditions of the treatment, the steel would become more noble as the effect of the cathodic polarization decreased over time and, with the redistribution of the OH^- ions, magnetite (Fe_3O_4) would become the dominant product formed. Miranda [59, 60] considered that the changing appearance of the corrosion products from reddish to dark-brown was due to the ECE-induced reduction of ferric compounds in magnetite (very small portion of goethite), as determined by X-ray diffraction. Finally, based on gravimetric analysis, Kim [84] found that the mass loss of corrosion products on samples with no treatment was in the range of 0.79 to 0.88 % whereas this value was significantly reduced in samples treated with different strengths of cathodic current.

5 SIDE EFFECTS

ECE treatment may also modify the characteristics of the reinforced concrete. It is therefore important to know to what extent this occurs, and whether such effects can be detrimental to the concrete or the steel rebar.

5.1 Modifications of concrete characteristics

5.1.1 **Porosity**

Buenfeld [76] and Monteiro [81] did not observe any change in the porosity induced by the treatment. In contrast, other authors found that ECE treatment modifies the concrete porosity: Castellote [114] reported an increase in total porosity as the electrical current was applied through the concrete. Siegwart et al. [47]'s study based on visual observations and scanning electron microscopy images of concrete treated samples (1 A/m² RS 42 d) at four different locations found that the number of pores (2309-4296) was larger and the average pore size (0.35-0.47 µm) was smaller than those observed in the control sample (1479 and 0.7 µm). Shan [83] also found a modification in the pore size distribution in mortar samples after ECE: the number of small pores (<100 nm) increased whereas the number of larger, harmful pores (≥100 nm) decreased.

5.1.2 **Ion migration**

Polarization applied during ECE treatment induces an ionic redistribution. A decrease in chloride concentration is observed in regions close to the steel rebar, as well as an increase of hydroxyl ions. Moreover, the accumulation of alkali ions (K^+, Na^+) is found to increase with time and/or current and varies according to the concrete cover. Orellan [57, 104] observed an increase of alkali ions in Na_2Oequiv/m^3 from 12 to 27 kgs of concrete located close to the steel rebar, and attributed this event to the introduction of Na+ during the immersion-drying cycles (NaCl was used to contaminate the specimens). This phenomenon was more pronounced

when the concrete cover was thin and the alkali ion concentration levels were sufficiently high. Fajardo [45] also observed an increase compared to the control specimen, according to the thickness of the concrete cover (20 or 50 mm) for Na^+ (20 to 40%), for K^+ (42 to 30%) and for Ca^{2+} (30 or 17%). Ihekwaba [56] found that the accumulation of alkali ions at the rebar/concrete interface is higher in carbonated than in non-carbonated concrete.

5.1.3 **New phases formed**

Marcotte et al. [80] used environmental scanning electron microscopy and energy dispersive X-Ray microscopy to study the effect of ECE on the microstructure of concrete, and observed that chlorides were no longer detectable at the surface of the steel after treatment. They also noted the absence of C-S-H and suggested that it had decomposed into cationic (calcium-rich) and anionic (silicate-rich) species under the impressed current. The formation of new cementitious phases (i.e., calcium-aluminium-rich, sodium-rich, iron-rich and silicon-poor phases) and their type depended on the source and magnitude of the chloride exposure.

Orellan [57, 104] monitored the evolution of concrete microstructure at different times after treatment. Immediately after treatment, he observed the systematic formation of a new thin, cementitious layer at the steel/concrete interface. This layer was composed of Ca, Si, Na, Al, and K. New compounds including Na, Ca and Si were observed, and the formation of secondary ettringite was reported in some cases. Two months after treatment, small amounts of alkali-silicate gel had formed near the steel rebar but no cracks were observed. Six months after treatment, large hexagonal crystals composed of Ca, Al and Cl had formed, as well as a large amount of alkali-silicate gel.

5.2 Potential risks to reinforced concrete

Although ECE is shown to be an effective treatment for extracting chlorides, mitigate corrosion and increase pH, there are a number of questions concerning potential harmful effects on the reinforced concrete.

5.2.1 Concrete cracking

Hydrogen gas produced during the treatment causes localised increases in concrete pressure and may promote concrete cracking. Polder et al. [43] observed a slight increase in micro crack density that appeared to be related to the material composition rather than the applied current density (1 and 4 A/m² RS). Indeed, it seemed that the high w/c ratio of OPC concrete made it sufficiently permeable to allow the gas to escape at a relatively low pressure without causing crack formation/expansion.

5.2.2 Steel embrittlement

Part of the hydrogen gas formed during the ECE process can be adsorbed on the metal surface as atomic hydrogen and may then enter the steel, contributing to its embrittlement. This risk can be monitored: no potential more negative than -1 100 mV with respect to Ag/AgCl/0,5 M KCl shall be permitted for plain reinforcing steel or -900 mV for prestressing steel for an alkaline concrete. Ordinary reinforcing steel does not suffer embrittlement [4] on the contrary to prestressed steel [28, 88, 115]. Siegwart et al. [88] observed hydrogen embrittlement in their study of prestressed concrete, using tendons stressed to the same degree. They investigated the role of stress in hydrogen-induced stress corrosion cracking (HISCC) on the central wire of the 7-wire strand of pre-stressed concrete samples which was post-tensioned to 90% of its proof stress and subjected to ECE treatment at 2 A/m² RS for 40 days. The test was repeated three times, but none of the samples failed prematurely due to HISCC. Other authors think that the risk of HISCC depends on the type of prestressed steel and on levels of prestressing [28, 115]. Therefore, it is not advised to use ECE on prestressed concrete structures.

5.2.3 Modification of mechanical properties

The chemical changes occurring during electrochemical chloride extraction, and particularly those in the vicinity of the steel rebar, can lead to modifications of concrete mechanical properties. These changes deserve some attention.

5.2.3.1 Loss of bond at the steel rebar/concrete interface

The bond strength between steel rebar and concrete occurs through adhesion, friction and the mechanical interlocking of surface deformation. Several authors have studied the impact of ECE on bond strength [35, 59, 76-78, 80, 82]. Marcotte et al. [80] observed a change in the microstructure of the mortar at the rebar/concrete interface (no C-S-H, an increase in the proportion of Ca(OH)2 and the formation of new cementitious phases) that may explain a loss of bond. Miranda et al. [59] suggested that a treatment-related reduction of certain ferric compounds in magnetite probably weakens the bond at the interface.

Buenfeld et al. [35, 76] studied plain rebar treated with a current density of between 0.02 and 0.75 A/m² RS for periods of up to 32 weeks. They found that prior to ECE the bond strength of specimens containing chlorides was around 57 % higher than that of specimens with no chlorides. This was attributed to corrosion enhancing the bond through a combination of prestressing and mechanical interlock. After 2 weeks of ECE, the enhanced bond strength was eliminated. The authors explained this result by the removal of corrosion products, as confirmed by visual examination. They also noted that the bond strength was heavily dependent upon the time interval between the ECE current being turned off and the bond test being carried out. Vennesland et al. [77] monitored bond stress evolution for samples subjected to ECE for 26 days and for different applied currents (1.6, 4 and 8 A/m² RS) after 1, 2, 7, 17 and 27 days. They noted extreme decreases in bond strength when higher currents were applied. After the end of ECE, they first observed a drop in bond stress between 7 and 17 days post treatment, followed by an increase when compared to the level of the reference specimen. Chang [116] reported significant bond loss and softening around rebars due to applied charge at ECE levels and proposed a parameter based on the impressed current density and polarization time. Ihekwaba et al. [78] performed pull-out tests and found that the specimens that were electrochemically treated with a very high impressed current density of 28.6 A/m² RS consistently showed increased degradation in bond strength and rebar slippage compared with specimens that had been impressed with a lower current density of 9.5 A/m²

RS. Yeih et al.[82] based their research on the “pushing-away concept”, which does not aim to completely remove free chloride but rather seeks a compromise between corrosion prevention and bond degradation.

5.2.3.2 Loss of compressive strength

Marcotte et al. [80] found that a reduction of compressive strength is one of the consequences of the cement phase alteration (composition and morphology) at the concrete/steel interface. Swamy et al. [55] also noted a loss in compressive strength and attributed it to ASR.

5.2.4 **Alkali-silica reaction (ASR)**

Sodium or potassium cations migrate towards the steel rebar during ECE (§5.1.2). If the concrete contains reactive aggregates, this increase in alkaline content may lead to alkali silica reaction (ASR). To avoid adding sodium or potassium ions to concrete, a $Ca(OH)_2$ solution is usually chosen as the electrolyte. Work by Swamy et al [55] demonstrated that ECE could trigger ASR in concretes that contain a reactive aggregate, i.e. synthetic fused silica (5 % or 15 % by mass of total aggregates). The concrete beams studied in the aforementioned article exhibited significant cracks after an 8-week treatment at a current density of 1 A/m² RS.
Miller [117] states that ECE will not cause ASR in the field mainly due to ECE being a short term treatment and ASR development is slow. On the other hand, Banfill [89] based on a mechanistic model for ASR found ECE to enhance the reaction, depending on the presence of chloride. It is therefore important to consider aggregate ASR reactivity before performing ECE.

In the case of structures contaminated by chlorides and already suffering from ASR, the literature reported that lithium-based electrolytes could be suitable for use in ECE treatment.
In Ueda's studies [70, 71], the current flow concerns the concrete surface and not the steel surface, it was found that ECE treatment could inject Li^+ into the concrete from the electrolyte solution (Li_3BO_3, 40° C) and produce an accumulation of Li^+ around the rebar. Indeed, Li^+ ions

competed with the alkali ions (Na^+ and K^+) and would therefore limit or suppress/prevent the ASR-induced expansion of concrete. Ueda [71] studied the penetration of Li^+ ions using different electrolyte solutions (Li_3BO_3, LiOH, Li_2CO_3, Li_2SiO_4, $LiNO_3$) at various temperatures (30 or 40 C). Results showed that the transference number of Li^+ increases with temperature and that Li_2CO_3 and $LiNO_3$ would be the most effective in terms of concrete penetration. $LiNO_3$ at 40 °C was the most suitable electrolyte for the prevention of ASR-related expansion.

6 LONG-TERM PERFORMANCE

Remaining chloride ions in concrete may redistribute and migrate back to the rebar and eventually result in the (re)initiation of corrosion. The protective effect of ECE treatment would thus be limited in time. Therefore, long-term performance of ECE treatment needs to be evaluated. Very few laboratory studies report the long-term durability of ECE treatment; the majority of publications describe periods ranging from 2 months to two years after the end of treatment [40, 48, 51, 55, 67, 79, 80, 83, 118].

Marcotte et al. [79] observed significant corrosion rates one month after treatment, and attributed this to the reduction of the passive film along with any existing corrosion products during the treatment. This would lead to general corrosion, and the reduction of dissolved oxygen in the concrete pore solution would prevent repassivation. They added that the increase in pH at the steel surface puts it in the alkaline corrosion region of the Pourbaix diagram ($HFeO_2^-$ and FeO_2^{2-} formation). However, it is important to note that samples were maintained in solution in this study, thus limiting the oxygen supply and preventing repassivation.

Elsener et al. [48] used chloride sensors to confirm that only a negligible amount of bound chloride was released two years after the end of ECE treatment.

Bouteiller et al. [67] reported no difference between steel potential values obtained 2 months after ECE and those obtained after 17 months of immersion of the beam in sea water. They

also noticed that the free chloride concentration remained stable close to the steel rebar for both the treated area and the control area.

Carmona et al. [40] tested chloride penetration sensitivity after treatment. They found that the capacity of chloride absorption during the 24 weeks of contamination was 17.4 % lower in the treated specimens than in non-treated samples.

Shan et al. [83] immersed the samples in a salt solution after ECE treatment. EIS, Ecorr and Icorr values showed that corrosion of the rebar resumed after 50 days.

Finally, Stoop et al. [51] studied the chloride ion redistribution within the cover over a one-year period, then developed a model to predict redistribution and thus risks of corrosion re-initiation over a 10-year period.

Swamy et al. [55] studied the effect of ECE on structural strength and bond for 400 days following the end of treatment. They did not report any adverse effect, although some of the treated specimens did contain fused silica (which is ASR reactive).

7 REGULATIONS

Different projects completed in North America have followed guidelines developed by the Strategic Highway Research Program (SHRP) [20-23] and by the Federal Highway Administration [72, 87]. A NACE international publication on electrochemical chloride extraction from reinforced concretes was published in 2001 [119] and re-approved in 2018 [120].

In Europe, the technical guidelines written in 2003 in CEN TS 14038-2 were replaced in 2018 by an AFNOR draft standard (PR NF EN 14038-2) entitled "Electrochemical realkalization and chloride extraction treatments for reinforced concrete - Part 2: Chloride extraction" [15].

8 MODELLING

Although experiments are a relevant approach when investigating ECE, this type of

498 investigation comes at a significant cost and requires time. It therefore appears interesting to
499 approach the processes through modelling. This approach requires a good knowledge of the
500 materials and the chemical and electrochemical mechanisms involved in ECE, and makes it
501 possible to determine influencing factors, carry out a first approach for the planned objectives
502 and predict the evolution of the process over time.
503
504 Several authors used the Nernst-Planck equation to model the migration of chlorides during
505 extraction. Yu et al. [121] presented a computer simulation of multicomponent ionic migration
506 that did not take binding and desorption effects into account, leading to a description of
507 experimental features that was qualitative but not quantitative. Andrade et al. [33] presented
508 the different mathematical models that were available related to the treatment conditions. They
509 first compared the mathematical solutions provided in the literature for constant initial chloride
510 concentrations in order to define the chloride concentration gradient throughout ECE treatment
511 under voltage or current control modes (easier mathematical calculation is possible with the
512 former). They then suggested a new computer program that could calculate the treatment
513 duration in the case of an initial gradient of chloride concentration (the model is based on
514 diffusion coefficient determination as a function of electrolyte conductivity, and on Fick's
515 second law). Only voltage control mode was considered. Andrade et al. [33] finally showed
516 that mathematical modelling can be performed. However, few data were available to validate
517 their proposal at the time of their study.
518 Polder [85] used diffusion modelling of chloride remaining after ECE to approach the time until
519 corrosion might reappear.
520 Sa'id-Shawqi et al. [122] used the Nernst-Planck and Laplace (potential and current
521 distributions determination) equations to predict the amount of extracted chloride and the
522 remaining chloride profile in a ECE treatment (voltage mode). The study solely considered free
523 chloride removal from concrete. Despite some differences between experimental and
524 modelling data, they highlighted that the chloride transport number decreases with increasing

525 treatment time or chloride concentration and found that the quantity of free chloride removed
526 from behind the rebar can increase with treatment duration.
527 Bound chlorides were not considered in the models in the second and third studies presented
528 above [25, 112], yet this phenomenon could have an influence on mechanisms.
529 Hassanein et al. [123] developed a model to predict chloride extraction during a constant
530 current ECE, in a two-step process. The first step corresponds to the rapid removal of free
531 chloride ions and the second concerns the release of bound chloride ions. In their experiments,
532 they found that a significant amount of chloride was removed during the first 4 weeks of
533 treatment and very little during the next 4 weeks. The rate of release of bound chloride was
534 determined using the difference between the chloride concentration at 4 and 8 weeks of
535 treatment, assuming that all chlorides were all either initially bound or free (an intermediate
536 value for their model was chosen). Several other input parameters were also used for chloride
537 ions (diffusion coefficient, initial concentration, percentage of initially bound, etc.), concrete
538 (pore solution fraction, density, cover thickness, etc.) and the electric field. They validated their
539 model based on 12 cases and obtained reliable accuracy despite the use of estimated values
540 for many parameters.
541 In 2007, Toumi et al. [124] also used the Nernst-Planck equation, with concrete porosity
542 density, pore solution composition, chloride-binding isotherm, and diffusion coefficients as
543 input data. They found good accordance between the model and the experimental results and
544 highlighted the important role played by the binding isotherm in the prediction of chloride
545 removal.
546 Li et al. [125, 126] investigated the effect of ionic concentrations on diffusion and migration of
547 ions in a concentrated pore solution when a constant current is applied. They revealed that the
548 higher the current density applied or the concentration of the species, the more the “activity
549 coefficient” (chemical potential change arising from ion-ion and ion-solvent interaction) can
550 affect ionic diffusion and migration. The authors then presented a finite element model and
551 performed numerical investigations in order to show the effects of applied current density,
552 treatment duration, the diffusion coefficient of different ions, ionic binding, boundary conditions

and medium porosity. They highlighted that even if the removal of chloride ions increases with both applied current density and time, a long-term treatment is likely to be less effective in decontaminating the zone at the steel/concrete interface and would even be counterproductive: their results show that during the treatment, the potential gradient decreases near the steel but increases near the anode. They also show the influence of chloride binding on chloride removal. The same authors (Wang et al. [127]) presented a two-dimensional computer model that considered the electrostatic coupling of ions and other factors (ionic adsorption, porosity and tortuosity of pore structure). Based on their results, they underlined that when possible, the use of anodes on two opposite sides of the structure improves the removal of chloride in the area behind the rebar. This is in accordance with results obtained during experimental studies [36, 52]. Castellote et al. [108], sought to identify an optimum treatment duration beyond which no further chloride is extracted due to the decrease in chloride transfer number through the increase of hydroxyl ions near the rebar. This theoretical optimum duration was defined as a function of the electrical charge passed, and was calculated using an empirical equation that considered the chloride diffusion coefficient decrease during the treatment. They also predicted chloride profiles after treatment according to the current applied.

While the previous works considered electro-neutrality condition, works conducted at the University of Plymouth [128-130] used Poisson's equation to determine the electrostatic potential. However, Poisson's equation is theoretically applicable when no electrical field is applied. When ECE treatment is applied, the electrolysis of water ensures the electro neutrality. In the study [128], the concrete was treated as a heterogeneous composite structure with two phases (mortar and aggregates that were assumed to be impermeable). The finite element model was developed for a voltage control mode and provided information about the migration of four ionic species (Cl^-, Na^+, OH^-, K^+) during the treatment. The analysis of the results highlighted the variation and the influence of migration speed, distribution and total amounts of ionic species. They also found that local diffusion could have a significant influence

on the evolution of migration speed. In the study [129], the concrete was presented as a three-constituent composite (i.e. a mortar matrix, aggregates and the interfacial zone between them). This two-dimensional model was also used to study how current flow pattern and the evolution of chloride concentration are affected by the use of different configurations of multiple rebars as cathodes. The authors underlined that the configuration of the rebars (cathode) in ECE is very important; a single well-located rebar provided a better extraction than the same rebar coupled with another rebar that was located closer to the anode. As described in numerous other experimental and numerical studies, although the amount of removed chloride increased with applied current density and treatment time, the daily removal rate decreased with treatment duration. Finally, they found that the two opposite effects on chloride extraction that resulted from the presence of aggregates, namely an increase in current and a change in porosity and tortuosity, almost cancelled each other out. Liu et al. [128] also studied different parameters (aggregates and their effect on interfacial zone effect of rebar position, etc.) using a finite element model based on inverse parameterization of chloride diffusion coefficients. They notably found that Na^+ and OH^- were mainly located around the rebar at the end of the treatment, while K^+ was widely distributed between the rebar and the anode. They also studied the influence of cracks and found that chloride extraction was slower when cracks were perpendicular to the electric field (the study excluded aggregates for this analysis).These recent types of modelling were developed to facilitate the study of structures with complicated geometries and of numerous physical parameters, but have not always been validated with experimental data.

An experimental analysis by Cheng et al. [131] of how ECE affects the evolution of porosity in concrete structure observed a decrease in porosity with treatment time close to the cathode and anode regions of the concrete, with a higher decrease noted near the anode region. They proposed a numerical simulation of this aspect and highlighted that taking the evolution of porosity into account led to a better accuracy in ECE treatment prediction.

Following this analysis of some of the attempts to model the ECE treatment of concrete, we can conclude that there are a great number of parameters which can (or should) be considered. Some authors proposed interesting models but did not always validate them by a comparison with experimental data. The use of some of the models can currently be considered to define or validate certain issues that are specific to concrete or inherent to the application of the treatment, but much work remains to be done. In particular, the type of used cement has a major influence on the porosity and the characteristics of the porous network (pore sizes and connectivity) and this aspect must be better defined in future models. Moreover, still in relation to the nature of the cement, the quantity of bound-free-total chlorides can be very different depending on cement and the kinetics of these chloride forms must be better evaluated. Finally, models involving electrochemical data would be a step forward.

9 CONCLUSIONS

Based on this literature review the following conclusions and recommendations can be drawn:

1. Electrochemical chloride extraction has proven to be an efficient treatment for the extraction of chloride ions from the concrete cover. In order to continuously protect the rebar, constant current mode is preferred (when using a constant voltage mode the current received by the rebar will decrease over time as the resistance of the medium increases). Moreover, as ECE seeks to protect the steel rebar from corrosion in concrete, the current density must be calculated according to the area of the steel rebar surface (A/m² RS) and not the concrete surface. Finally, it is important to specify the duration of the treatment in order to calculate the total charge (in A.h/m²), which is useful data when comparing results.
2. Design is an important step to achieve successful ECE (steel rebar surface, choice of electrolyte, position of the anode, continuous or intermittent treatment, etc.)

3. ECE efficacy should be determined according to multiple criteria: extraction of chlorides (free and total), mitigation of corrosion (half-cell potentials, corrosion current density, resistivity, etc.), pH and visual inspection.
4. ECE efficacy has been demonstrated based on analyses performed before and immediately after the ECE, and after steel rebar depolarization, with full consideration of treated and control samples.
5. Optimisation of ECE (current density and duration) is needed to successfully demonstrate its efficacy without causing detrimental effects (such as altered composition and morphology at the rebar/concrete interface, loss of bond, etc.) and to propose performance criteria.
6. Limitations of ECE are:
    - Chloride ions penetrated behind the rebar (in this case Cathodic Protection should be preferred to ECE).
    - Prestressed concrete (hydrogen embrittlement)
    - Reactive aggregates (ASR risk)
7. ECE-related rebar repassivation should be studied in greater depth using in-situ techniques such as micro-Raman spectroscopy in laboratory. The only evidence provided in the literature concerns corrosion mitigation; no evidence of repassivation has been presented to date.
8. Models still need to be validated with experimental data.
9. Further work is needed to demonstrate the long-term efficiency of ECE 1, 2, 5 and 10 years after treatment.

Finally, this literature review clearly highlights the urgent need to transfer technology from a state of laboratory knowledge (experimental and modelling) to functional use in the practical field.

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