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Additional Information

Effect of macrocell currents on rebar corrosion in
 reinforced concrete structures exposed to a marine
 environment

4 J.R. Lliso-Ferrando<sup>a,</sup>\*, I. Gasch<sup>a</sup>, A. Martínez-Ibernón<sup>a</sup>, M. Valcuende<sup>b</sup>

<sup>a</sup> Research Institute for Molecular Recognition and Technological Development (IDM),
Universitat Politècnica de València, Camino de Vera, s/n., 46022 Valencia, Spain.

- <sup>b</sup> Department of Architectural Constructions, School of Architecture, Universitat Politècnica de
   València, Camino de Vera, s/n., 46022 Valencia, Spain.
- 9 \*Corresponding author. Tel.: +34 655633238; E-mail address: jollife2@arq.upv.es
- 10 Address: Electrochemistry Laboratory, 1B-ETSIE, Universitat Politècnica de València, Camino de
- 11 Vera, s/n., 46022 Valencia, Spain

### 12 Abstract

13 Autonomous corrosion monitoring systems in reinforced concrete structures tend to use 14 sensors that are electrically isolated from rebars. This monitoring strategy ignores the 15 macrocell currents that take place between active and passive areas of reinforcement. The 16 effect of these currents has been studied on rebar corrosion in structures exposed to a marine 17 environment. The results showed the existence of macrocell currents generates greater anodic 18 polarisation in rebars' active areas, which favours chloride migration towards steel surface. As 19 a result, corrosion onset takes place sooner, the concentration of the chlorides is higher, and 20 the corrosion rate increases. When a rebar is in passive state, steel mass loss is negligible due 21 to macrocell currents. However, after corrosion onset, the influence of these currents is 22 greater the worse concrete quality is, and the smaller cover is. The corrosion rate showed 23 differences of almost two orders of magnitude in concrete C30 and of one order of magnitude 24 in concrete C50. When the total corrosion rate is determined only by measuring microcell 25 currents, the obtained values tend to underestimate the mass loss due to corrosion by 90%. 26 Conversely, when the analysis includes macrocell currents, a good fit between the theoretical 27 and real values is obtained.

## 28 <u>Keywords</u>

29 Reinforced concrete, corrosion, macrocell current, chlorides, marine environment.

## 30 **<u>1-. Introduction</u>**

The corrosion of rebars embedded in reinforced concrete structures (RCS) is one of the main causes of premature deterioration and failure when they are located in marine environments (Vélez et al., 2011; Qiao et al., 2016; Balestra et al., 2019; Liang and Wang, 2020; Pushpakumara and Thusitha, 2021; Torres-Acosta et al., 2021; Torres-Acosta and Castro-Borges, 2021). This phenomenon reduces their service life and leads to huge economic losses (Moradian et al., 2012).

Rebars are protected by a concrete cover that acts as a physical barrier against aggressive agents (Poursaee, 2016). Thanks to the cementitious matrix's high alkalinity, cover also acts as a chemical barrier because a layer of oxides forms on rebars that reduces ionic mobility and steel dissolution with negligible values (passive film) (Carnot et al., 2003; Bagheri and Rastegar, 41 2019). These conditions cannot be sustained perpetually when structures are located in 42 marine environments given concrete's permeability: chloride may reach rebars and favour 43 steel depassivation (Montemor et al., 2003). As a result of concrete's heterogeneity, chloride 44 ions do not reach rebars uniformly, but locally, which triggers the appearance of pitting. In 45 these regions, anodic and cathodic areas co-exist (Evans, 1978; Mansfeld, 1971; Bertolini et al., 46 1996) whose generated current (microcell curent) is added to the current that occurs between 47 the corroded and uncorroded areas (macrocell current) that are electrically connected to one 48 another (Soleimani et al., 2010; Valipour et al., 2014). This means that the real rebar corrosion 49 affected by this phenomenon corresponds to the sum of both current intensities (Qian et al., 50 2006; Andrade et al., 2008; Andrade, 2019) (Eq. 1) Figure 1.

$$i_{CORR} = i_{CORR,MICRO} + i_{CORR,MACRO}$$
(Eq. 1)

51 In accordance with Faraday's law, this current can also be expressed in corrosion rate terms 52  $(V_{CORB})$ , which is steel section loss by corrosion:

$$V_{CORR} (\mu m/year) = 1.17 \cdot i_{CORR} (\mu A/cm^2) \qquad (Eq. 2)$$





Figure 1- Microcell and macrocell corrosion

55 The seriousness and cost of the damage caused by corrosion in RCS, and the increasing use of 56 reinforced concrete in harbour and offshore structures (Fernández and Pardob, 2013), have 57 led many researchers to design embedded monitoring systems to determine the corrosion rate 58 without having resort to test boring or in situ inspections. Furthermore, these systems are 59 really interesting if we consider that most structures located in marine environments present 60 partial or complete inaccessibility to take on-site corrosion measurements. Furthermore, 61 according to the codes for durability design in many countries, it is compulsory for projects of 62 structures to include an inspection and maintance plan, which makes correct monitoring very 63 important.

64 In recent years, much progress has been made in developing embedded sensors capable of 65 detecting the depassivation of reinforcement (Andrade, 2020; Daniyal and Akhtar, 2020; 66 Rodrigues et al., 2021), and of measuring the electrochemical potential of adjacent rebars 67 (Karthick et al., 2018; 2019) or the resistivity of concrete cover (Ramón et al., 2021). Other 68 authors have designed sensors to determine the presence of chlorides or concrete carbonation 69 (Gandía-Romero et al., 2016a; 2016b; Ramani and Kuang, 2020), as well as to quantify other 70 corrosion kinetic-related parameters; for example, humidity or oxygen concentration 71 (Martínez-Ibernón et al., 2021). Nonetheless, the most interesting parameter to properly 72 evaluate and control a structure's durability is the corrosion rate because it provides 73 quantitative information about steel section loss owing to corrosion. One of the first examples 74 of this kind of sensors was developed by Martínez and Andrade (2009), in which corrosion rate 75 measurements were taken by the galvanostatic pulse method. Figueira (2017) presented a 76 similar system, which estimated the corrosion rate by the polarisation resistance technique. 77 Another monitoring system was designed by Ramón et al. (2016), who simplified existing 78 models. The measuring technique is set up as a 2-electrode arrangement to avoid resorting to 79 a reference electrode (Martínez-Ibernón et al., 2019; Ramón et al., 2016; 2020). A common 80 characteristic of all the above-cited sensors is that the measurement technique is applied to 81 the working electrode electrically isolated from rebars while corrosion is being measured. 82 Therefore, sensors do not measure the macrocell current, and only local corrosion is recorded 83 (microcell current).

84 Not many research works have analysed the importance of macrocell currents on rebar 85 corrosion. Most have focused on solution studies (Andrade et al., 1992, 2008; Dong and 86 Poursaee, 2020, Chen and Su, 2021), but the obtained results are not representative of the 87 situation that actually occurs in RCSs (Lliso-Ferrando, 2021). The few conducted studies about 88 concrete have obtained very different results. For instance, Valipour et al. (2014) pointed out 89 that macrocell currents are only a small part of the corrosion rate (5-10%), while François 90 (2022) considered that these currents correspond to about 100% total corrosion. Other studies 91 indicate that macrocell currents represent about 30% total corrosion (Hansson et al., 2006; 92 Subramaniam et al., 2010). Revert et al. (2019) obtained values between 20% and 55%, while 93 Rodríguez et al. (1999) reported values of 70%. These different results in the bibliography are 94 also conditioned by the testing conditions under which these studies were performed.

95 As the uncertainties about the importance of macrocell currents in RCSs' corrosion processes 96 are substantial, it is necessary to analyse this aspect because autonomous monitoring systems 97 do not take them into account. To correctly quantify rebar corrosion rates, that is, their section 98 loss due to corrosion, it is essential to ensure the structure's security, evaluate its service life 99 and avoid costly repair processes. Consequently, the objective of this research work was to 100 study both the influence of macrocells on rebar corrosion and the importance of 101 contemplating them in corrosion monitoring systems for RCS exposed to a marine 102 environment. To do so, rebars were embedded in concretes with different compressive 103 strengths and distinct covers. Specimens were partially submerged in 3% NaCl solution for 104 almost 1 year and the corrosion rate of the embedded rebars was monitored. Research was 105 conducted using a large cathodic surface that simulates the conditions that would take place in 106 densely reinforced areas like those in marine structures. A gravimetric comparison between 107 the mass loss estimated according to the corrosion monitoring results and real mass loss was 108 made for the study validation. Chloride diffusion inside the material was also studied to 109 estimate the time that chlorides take to reach rebar surface.

# 110 **<u>2-. Experimental plan</u>**

## 111 <u>2.1.-Materials</u>

112 In order to manufacture the specimens herein used, four concrete types with different 113 characteristics and properties were prepared: low-strength concrete 30 MPa (C30); high-114 strength concrete 50 MPa (C50); very high-strength concrete (C90); ultra high-performance 115 fibre-reinforced concrete (UHPFRC, UH-150). The characteristics of each mix appear in Table 1. 116 All the concretes were made using CEM I 42.5 R/SR. For concretes C90 and UH-150, silica fume 117 (Elkem Microsilica®) was also included in the mix. For concrete type UH-150, siliceous flour 118 with a similar particle size distribution to the cement and steel fibres in 2%(vol.) (Ø0.2 mm; 13 119 mm) was used. All the specimens were cured at 20±2°C and 95% relative humidity (RH) until 120 the age of 28 days.

	C30	C50	C90	UH-150
Cement	292	450	500	800
Water	190	225	178	160
Superplasticizer Sika <sup>®</sup> -20HE	2.80	1.37	3.50	30
Silica fume			55	175
Siliceous flour				225
Siliceous sand fine (0/0.5)				302
Siliceous sand medium (0.6/1.2)				565
Limestone sand (0/4)	1256	880	914	
Limestone gravel (4/8)		880	779	
Limestone gravel (4/12)	707			
Steel fibres (Ø0.2mmx13mm)				175
w/c ratio	0.65	0.50	0.36	0.20
w/b ratio	0.65	0.50	0.32	0.16
fc (28 days) (MPa)	30.67	49.88	88.86	135.43

#### 121 <u>Table 1</u>- Mixture proportions of concretes $(kg/m^3)$

122

## 123 <u>2.2.-Test methods</u>

124 In order to know resistance to chloride diffusion, the diffusion coefficient was analysed in the 125 non-stationary state for each concrete type. At the same time, the corrosion on the embedded 126 rebars was followed up. The compressive strength at the age of 28 days was determined from 127 each mix on cylindrical specimens ( $\emptyset = 100$  mm; height = 200 mm) (Table 1).

# 128 <u>2.2.1.- Chloride diffusion test</u>

129 Chlorides penetrate through the cementitious matrix mainly by diffusion (De Medeiros-Junior 130 et al., 2015; Balestra et al., 2020). The unidirectional diffusion coefficient of each concrete type 131 was obtained according to European Standard EN 12390-11 (AENOR, 2019). Testing was done 132 with cylindrical samples ( $\phi$  = 100 mm; height = 100 mm) exposed to a unidirectional chloride 133 diffusion flow. For this purpose, a water tank (300 mm high) with 3% NaCl solution was placed 134 on one of the specimen sides and epoxy coating was applied to the others, as described in the 135 standard method (AENOR, 2019). Specimens were left under these conditions for 90 days. 136 After this period, the chloride concentration was measured at different depths. The chloride content of each sample was obtained as set out in Standard NT Build 208 (NORDTEST, 1996). 137 138 For concrete UH-150, the steel fibres in each sample were removed by an electromagnet before running the chloride content analysis. Testing was done on three samples for eachconcrete type and the result for each mixture was taken as the arithmetic mean of the threerecorded values.

142 <u>2.2.2.- Corrosion monitoring</u>

143 The monitoring of macrocell and microcell corrosion ( $i_{CORR,MACRO}$  and  $i_{CORR,MICRO}$ ) was done

on cubic specimens ( $150x150x150 \text{ mm}^3$ ) with three embedded rebars ( $\emptyset 10 = \text{mm}$ ; 150 mm long), as described in Figure 2.



146 147

<u>Figure 2</u>- Test specimens' configuration

Rebars were partly covered by epoxy and a PVC pipe (Figure 2) to delimit the surface that came into contact with concrete (working area) at 15.71 cm<sup>2</sup>, and were arranged on the specimen with different covers: 5 mm, 10 mm, 20 mm and 30 mm. Two specimens for each concrete type and cover were made. After a curing period in a humid chamber, the upper side was sealed with epoxy coating. Then specimens were partly immersed in 3% NaCl solution and were left under these conditions for almost 1 year.

154 Along with each specimen, a carbon fibre meshing piece was placed (600 cm<sup>2</sup> surface area). 155 This piece was electrically connected to the two lateral rebars (Figure 3). The rebars in each 156 specimen were divided into two groups: two rebars connected (C) to the carbon fibre meshing 157 piece and one was not connected (NC). Using carbon fibre mesh as a cathode allows the 158 oxygen present in solution to be adsorbed on its surface and to undergo a reduction process 159 with the electrons from rebars to simulate rebar behaviour in the passive state. Another 160 advantage of using carbon fibre mesh is that no oxidising takes place under dissolution 161 conditions. In this way, a macrocell current was established between rebars (active area) and 162 mesh (passive area).

For each concrete type and cover, the microcell current of two rebars (NC rebars) was monitored, as were the macrocell and total currents in four other rebars (C rebars). In all, 96 rebars were monitored. <u>Table 2</u> lists the number of rebars and the nomenclature used in this work.

167 <u>Table 2</u>- Rebars, configuration and nomenclature of specimens.

Concrete type	c	C30 C50		C90		UH-150		
Electric configuration	(C)	(NC)	(C)	(NC)	(C)	(NC)	(C)	(NC)
Cover: 5 mm (+5)	4	2	4	2	4	2	4	2
Cover: 10 mm (+10)	4	2	4	2	4	2	4	2
Cover: 20 mm (+20)	4	2	4	2	4	2	4	2
Cover: 30 mm (+30)	4	2	4	2	4	2	4	2

**Total rebars** 

96 rebars

I 168 (C-Connected and NC-Not Connected to carbon fibre mesh)



169

170

### Figure 3- Electrical configuration

171 When rebars C were connected to carbon fibre mesh, the net current flow between it and the

172 two rebars of each specimen was measured by a Zero Resistance Ammeter (ZRA; Keithley 2000

173 Tektronix model). The value obtained for each rebar was normalised by its surface (15.71 cm<sup>2</sup>).

This current corresponds to the macrocell current ( $i_{CORR,MACRO}$ ). The value was recorded 1 174 175 minute after measurements commenced to ensure that the recorded signal was stable enough

176 (Figure 4).





Figure 4- Macrocell measurement configuration

179 Moreover, rebar corrosion current density ( $i_{CORR,MICRO}$ ) was periodically measured in all the rebars (C and NC) by the potentiostatic step voltammetry (PSV) technique described in 180 181 (Martínez-Ibernón et al., 2019; Ramón et al., 2019, 2020). This measurement system allows 182 rebar corrosion to be analysed by studying the response of steel to potentiostatic-type 183 disturbance. The measurement was taken with an Autolab PGSTAT 100 Potentiostat. The measuring cell arrangement was a 3-electrode one: the working electrode on which corrosion 184 185 was measured was each rebar; the reference electrode was a saturated calomel electrode 186 (SCE) partly immersed in solution; the carbon fibre mesh of each analysed specimen was used 187 as the counter electrode. The obtained result was also normalised by the rebar surface (15.71  $cm^2$ ) to obtain microcell current intensity ( $i_{CORR,MICRO}$ ). To take this measurement, bars must 188 189 be electrically disconnected from carbon fibre mesh. After taking measurements, rebars (C) 190 were reconnected to mesh to continue participating in macrocell processes until the next 191 measurement was taken.

During the monitoring period, the water level was always 1.5 cm below the upper specimen side thanks to a level sensor being used. Every 2 weeks, the chloride content in solution was controlled to ensure that the chloride concentration remained constant. To do so, water samples were taken and analysed by Mohr's method (Standard UNE-ISO 9297:2013) (2013).

196 Specimens were subjected to rebar inspection at the end of the corrosion monitoring period. 197 For this purpose, rebars were removed from specimens and cleaned for a 10-minute cycle in 198  $0.1M H_3PO_4$ , which was repeated twice. Afterwards, a gravimetric comparison between the 199 estimated mass loss was made according to the corrosion monitoring results and the real mass 200 loss.

## 201 **3.- Results and Discussion**

# 202 <u>3.1.- Chloride diffusion test</u>

203 <u>Figure 5</u> shows the chloride content (% per cement weight) obtained for each analysed sample
 204 in relation to the depth at which samples were taken.



206

Figure 5- Chloride content per cement weight (%) depending on depth

207 With these data, and in line with European Standard EN 12390-11 (AENOR, 2019), the  $D_{app}$ 208 coefficient value was calculated by Fick's second law, Eq. 3:

$$C(x,t) = C_0 + (C_s - C_0) \cdot \left[ 1 - \operatorname{erf}\left(\frac{x}{2\sqrt{D_{app} \cdot t}}\right) \right]$$
 (Eq. 3)

209 where  $D_{app}$  is the non-steady state diffusion coefficient, expressed as m<sup>2</sup>/s,  $C_s$  is the chloride concentration on the concrete surface,  $C_0$  is the initial chloride concentration (chloride content 210 211 in % mass prior to immersion in NaCl solution) and C(x, t) is chloride content in the % of 212 cement weight obtained at a given depth (x) and for a given exposure time (t). The initial 213 chloride content (chloride contributed by raw materials) was obtained from the samples not 214 contaminated by chloride solution, with 0.03% in UH-150, 0.04% in C90, and 0.09% in C30 and 215 C50. The surface chloride concentration ( $C_s$ ) and the  $D_{app}$  coefficient were obtained by fitting Eq. 3 to the experimental data on the chloride concentration at different depths (Figure 5) by a 216 217 non-linear least squares' regression analysis. Figure 6 shows the curve fitting with the 218 experimental data obtained for each concrete type.



219 220

Figure 6- Experimental values and curve fitting depending on the concrete type



	C30	C50	C90	UH-150
$D_{app}$ (x10 <sup>-11</sup> m/s <sup>2</sup> )	6.806	4.326	0.111	0.031
222				

223 <u>Table 3</u> shows the average  $D_{app}$  coefficient value obtained for each concrete type. This 224 parameter is a good indirect indicator of materials' durability because it represents resistance 225 that materials confer chloride diffusion. The results demonstrated a huge difference between 226 the different concretes. Concrete UH-150 had the lowest value thanks to a very dense 227 microstructure, as previous works have shown (Valcuende et al., 2021a; 2021b). The  $D_{app}$ 228 value obtained with concrete C90 was one order of magnitude higher than that for UH-150. 229 Using higher size aggregates increased the heterogeneity of porous structure, which facilitated 230 greater chloride diffusion. The mixture of both concretes C90 and UH-150 included silica fume, 231 which segmented the capillary network and favoured tortuosity (Dotto et al., 2004; Kupwade-232 Patil et al., 2018; Bentz et al., 2000). This characteristic distinguished these concretes from C30 233 and C50 by having a much higher  $D_{avp}$  value (about two orders of magnitude higher than UH-234 150).

The interest in this value lies in the possibility of estimating the required period of time for chlorides to reach rebars and corrosion onset. This time is calculated by Eq. 4, which was obtained from Eq. 3:

$$t = \frac{x^2}{4 \cdot D_{app} \cdot \left[ erf^{-1} \left( \frac{C_s - C_{crit}}{C_s - C_0} \right) \right]^2}$$
 (Eq. 4)

238 where  $erf^{-1}$  is the inverse Gaussian error function, x is the rebar cover depth, and  $C_{crit}$  is the 239 critical chloride concentration threshold that alters rebar passive layer stability and leads to 240 corrosion onset. The C<sub>crit</sub> parameter is not a well-defined value in the scientific literature. The 241 higher the chloride concentration on the steel surface, the more likely corrosion onset is. A 242 wide range of values has been suggested (Angst et al., 2009; Angst, 2011, 2019; Gao et al., 243 2019). By taking the 0.6% cement weight as a value that some recommendations propose 244 (Ministerio de Fomento, 2008), Figure 7 represents the corrosion onset curves according to 245 concrete cover.



246

247

Figure 7- Corrosion onset depending on concrete type and concrete cover

According to Figure 7, it is possible to calculate the time needed for corrosion onset for the concrete types and covers used in the specimens described in Section 2.2.2. <u>Table 4</u> offers the calculated values. These are approximate values and cannot be taken as certain values. However, they allow comparisons to be made between different concretes.

252

253 <u>Table 4</u>- Time period (years) estimated for rebar depassivation according to concrete type and cover

	C30	C50	C90	UH-150
Cover: 5 mm (+5)	0.01	0.03	1.08	5.62
Cover: 10 mm (+10)	0.05	0.12	4.32	22.47
Cover: 20 mm (+20)	0.20	0.47	17.26	89.89
Cover: 30 mm (+30)	0.44	1.05	38.83	202.36

# 255 <u>3.2.- Microcell corrosion current (i<sub>CORR,MICRO</sub>)</u>

256 Figure 8 depicts the average microcell corrosion current value ( $i_{CORR,MICRO}$ ) of the rebars 257 embedded in specimens. In each graph of Figure 8, the limit value is marked at 0.1  $\mu$ A/cm<sup>2</sup>, 258 which is the threshold indicated by the Rilem TC-154 EMC recommendation (2004) below 259 which the corrosion rate can be considered negligible. This document also points out that the 260 0.1-0.2  $\mu$ A/cm<sup>2</sup> range of values can be taken as a transition zone between the passive and 261 active corrosion states. The graphics also point out the threshold of 1  $\mu$ A/cm<sup>2</sup>, which is the 262 limit from which the corrosion rate is high.



![](_page_10_Figure_6.jpeg)

Figure 8- Microcell corrosion current depending on concrete type and concrete cover

### 265 *3.2.1. Effect of concrete type and concrete cover*

266 Corrosion started in all the rebars embedded in the C30 specimens. The  $i_{CORR,MICRO}$  values 267 over 0.1  $\mu$ A/cm<sup>2</sup> were recorded on the first days for the rebars with less cover (5 mm). The time needed for the bars with a bigger cover to undergo these increases was longer and closely related to the data obtained in <u>Table 4</u>. Thus, for example, for the rebars with the 30 mm cover, depassivation was estimated to take place at the age of 0.44 years (160 days), which coincides with the age at which the NC rebars actually started to present values above 0.1-0.2  $\mu$ A/cm<sup>2</sup> (≈151 days).

This same trend was noted in specimens C50. With the 20 mm cover, the corrosion levels of 273 274 the rebars with the NC configuration exceeded 0.1  $\mu$ A/cm<sup>2</sup> after about 150 days. This result 275 matches the estimation of 0.47 years ( $\simeq$  170 days) set by the calculations done with the 276 obtained  $D_{avv}$  (Table 4). For the specimens C50 with a 30 mm cover, the  $i_{CORR,MICRO}$  values 277 were below 0.1 µA/cm<sup>2</sup>, which indicates that rebars had still not depassivated. The forecasted 278 time before the surface of these rebars reached the critical chloride content took longer than 1 279 year ( $\simeq$  390 days) (Table 4). This information agrees with the fact that no corrosion signs were 280 detected after the 320-day follow-up (see Section 3.5).

The very high resistance of concretes C90 and UH-150 to chloride diffusion, as shown by the diffusion test and the estimated data found in <u>Table 4</u>, as well as their high electrical resistivity (Valcuende et al., 2021a), justify that no corrosion was recorded in these concretes (<u>Figure 8</u>). All the rebars had values below 0.1  $\mu$ A/cm<sup>2</sup>. For the most unfavourable case (5 mm cover), the corrosion current at the end of the monitoring period was 0.028  $\mu$ A/cm<sup>2</sup> in concrete C90 and 0.015  $\mu$ A/cm<sup>2</sup> in concrete UH-150.

287 3.2.2. Effect of connection between rebars and carbon fibre mesh on microcell corrosion 288 currents

289 As observed in the graphs of Figure 8, especially for concretes C30 and C50, the rebars that 290 were connected to carbon fibre mesh (C rebars) (to simulate the connection to other passive 291 reinforcements) tended to depassivate at earlier ages and reached higher corrosion rates than 292 the isolated rebars (NC rebars). Thus, for example, with specimens C30 and a 30 mm cover, the 293 corrosion onset in the NC rebars occurred at 151 days, while the corrosion onset in the C 294 rebars took place at 30 days. Furthermore, the  $i_{CORR,MICRO}$  reached a value of 0.38  $\mu$ A/cm<sup>2</sup> in 295 the NC rebars and 5.1  $\mu$ A/cm<sup>2</sup> in the C rebars, which is more than one order of magnitude 296 difference between them. This tend was also observed in concrete C50 for covers up to 20 297 mm.

298 What these data reveal is that a connection to passive reinforcements accelerates corrosion 299 onset and increases microcell corrosion kinetics. This is because chloride ions (negative 300 electrical charge) are attracted by steel, which has a positive electrical charge as a result of the 301 current flow between the rebar and carbon fibre mesh. Thus, when a macrocell current is 302 present (as in the C rebars), chloride diffusion is intensified by a migration process. This fact 303 was demonstrated by the total chlorides analysis performed in the surroundings of the C and 304 NC rebars embedded in several specimens (Figure 9) at the end of the follow-up period. The 305 chloride content around the rebars subjected to macrocell currents (connected rebars) was 306 between 80% and 600% higher than in the NC rebars.

![](_page_12_Figure_0.jpeg)

308

312

Figure 9- Chloride content (% per cement weight) around rebars

![](_page_12_Figure_4.jpeg)

310 Figure 10 depicts the average macrocell corrosion current value ( $i_{CORR,MACRO}$ ) of the rebars

311 connected (C) to carbon fibre mesh.

![](_page_12_Figure_7.jpeg)

313 *Figure 10- Macrocell corrosion current depending on concrete type and concrete cover configuration* 

The macrocell current evolution displayed a similar trend to corrosion  $i_{CORR,MICRO}$ . Significant increases in  $i_{CORR,MACRO}$  took place on the first days for the rebars embedded in concrete C30 (at 10 days in the rebars with a 5 mm cover, at 30 days in the rebars with covers of 10 mm and 317 20 mm, and at 60 days for the rebars with a 30 mm cover). In fact, the  $i_{CORR,MACRO}$  values 318 were higher than 10  $\mu$ A/cm<sup>2</sup> in all cases, which denotes very high corrosion levels. This 319 behaviour was also noted for the rebars embedded in concrete C50 for the 5 mm and 10 mm 320 covers. This trend was repeated for the rebars with a 20 mm cover whose values were 1 321  $\mu$ A/cm<sup>2</sup> after 225 days.

In concretes C90 and UH-150, macrocell current intensity was below  $0.1 \ \mu$ A/cm<sup>2</sup> throughout the study period. These concretes' high resistance to chloride diffusion (Section 3.1.) hindered chloride ions reaching rebars during their exposure period. Moreover, limited permeability to gas (Valcuende et al., 2021a; 2021b) prevents oxygen availability at rebars' depths, which does not allow the onset of corrosion processes. The high electrical resistivity of both these concrete types (Valcuende et al., 2021a; 2021b) also limits ionic mobility, which lowers the possibility of electrochemical processes like corrosion taking place.

# 329 <u>3.4.- Total corrosion current (*i*<sub>CORR</sub>) and mass loss</u>

330 In order to properly know the rebar corrosion state and the influence of macrocell processes, it

is necessary to jointly consider macrocell and microcell corrosions (Eq. 1) to calculate the

![](_page_13_Figure_5.jpeg)

![](_page_13_Figure_6.jpeg)

![](_page_13_Figure_7.jpeg)

Figure 11- Corrosion current depending on concrete type and concrete cover configuration

In the rebars where corrosion onset was clear (concretes C30, and C50 with 5, 10 and 20 mm
 covers), a large difference was observed between the rebars that had undergone macrocell
 processes (C rebars) and those that had undergone only local corrosion processes (NC rebars).

The  $i_{CORR}$  values showed differences of almost two orders of magnitude in concrete type C30 and of one order of magnitude in concrete type C50.

The results obtained in concretes C90 and UH-150 were below  $0.15 \,\mu\text{A/cm}^2$ , even with a 5 mm cover. These data indicate that the repercussion of macrocell currents on the rebars in passive state was minimum.

By integrating the corrosion intensity-time curve, the mass loss ( $\Delta m, calc$ ) that accumulated during the exposure period was calculated in accordance with Faraday's law (Eq. 5), as shown by:

$$\Delta m, calc = \frac{M}{n F} \cdot \int_{t_0}^t i_{CORR} \cdot dt \qquad (Eq. 5)$$

where *M* is the steel atomic mass (55.845 g/mol), *t* is the time in seconds, *n* is the number of electrons released or acquired during the corrosion process (2 for this case) and *F* is Faraday's constant (96845 C/mol). Mass loss ( $\Delta m$ , calc) was obtained in g/cm<sup>2</sup> and normalised by rebars' working area (15.71 cm<sup>2</sup>).

350 The obtained results are presented in Figure 12 and Table 5. Figures 12(a) and 12(b) show 351 mass loss after 320 days under the seawater exposure conditions for NC (not connected to 352 macrocell) and C (participating in macrocell processes), respectively. By comparing both 353 graphics, once again mass loss was higher in those specimens that participated in macrocell 354 processes (C). Thus, for example, the average mass loss obtained for C30 (5 mm cover) was 355 130 mg in specimens NC and was 6600 mg in specimens C; that is, 50-fold more. The data also 356 showed that the influence of macrocell currents was greater the lower the concrete quality 357 was, and the smaller concrete cover was.

358 It is also noteworthy that mass loss was insignificant due to macrocell currents when a rebar 359 was not depassivated; e.g. for UH-150 (30 mm cover), mass loss was 1.92 mg and 2.81 mg in 360 specimens NC and C, respectively.

![](_page_15_Figure_0.jpeg)

362 <u>Figure 12</u>- Mass loss of rebars considering (a) only corrosion per microcell and (b) total corrosion

#### 363 <u>Table 5</u>- Accumulated mass loss (mg)

361

Concrete type	C	30	C50		C90		UH-150	
Electric configuration	(C)	(NC)	(C)	(NC)	(C)	(NC)	(C)	(NC)
Cover: 5 mm (+5)	6,676.9	128.98	3,442.6	52.57	22.23	2.24	7.09	1.96
	(9.58)	(7.29)	(11.13)	(13.96)	(3.97)	(3.31)	(2.61)	(1.68)
Cover: 10 mm (+10)	5,393.7	80.05	2,269.9	45.54	5.91	2.17	4.58	1.75
	(13.78)	(10.40)	(9.15)	(4.58)	(1.98)	(1.12)	(2.62)	(1.18)
Cover 20 mm (+20)	4,468.1	101.12	627.02	21.25	2.30	2.94	3.10	2.62
Cover: 20 mm (+20)	(12.23)	(5.41)	(6.91)	(7.05)	(2.72)	(2.94)	(2.37)	(1.65)
Cover: 30 mm (+30)	3,679.9	28.64	9.62	2.45	4.03	2.29	1.10	2.02
	(9.67)	(2.48)	(4.13)	(2.14)	(2.18)	(1.81)	(3.44)	(1.84)

364 Numbers in brackets are the coefficients of variation

# 365 <u>3.5.- Visual inspection</u>

In order to confirm the obtained results, all the concrete specimens were visually inspected(Figure 13).

![](_page_16_Figure_0.jpeg)

369 <u>Figure 13</u>- The state that specimens were in after being immersed in 0.5M NaCl solution for more than
 370 320 days

The damage noted on concretes C30 and C50 was sometimes relevant. For covers 5 mm and 10 mm, specimens presented large rust stains, or even cracking. This damage concentrated on lateral bars; that is, on those connected to carbon fibre mesh (C bars) and, therefore, on those that participated in macrocell processes. For bigger covers (20 mm or 30 mm), much less damage was observed on concrete C30 and damage was almost inexistent on the concrete C50 specimens, which showed only small rust stains and very few small cracks. All these results correspond well to the corrosion rates recorded on these bars (Figure 11).

The visual inspection revealed no damage on specimens C90, and microscopic rust stains on the surface of the UH-150 specimens owing to the corrosion of more superficial steel fibres. Thus oxidation affected only fibres closer to the surface and not inner fibres or rebars (Figure

381 <u>14</u>).

![](_page_17_Picture_3.jpeg)

382

383 <u>Figure 14</u>- Details of: (1) superficial fibre corrosion; (2) rusty superficial fibres; (3) inner fibres with no
 384 corrosion signs

385 Finally, a visual inspection of rebars was made after removing them from the test specimens. 386 This examination confirmed the strong influence of macrocell processes on total rebar 387 corrosion (Figure 15). For instance, with concrete C30 and 5 mm or 10 mm covers, the rebars 388 that participated in macrocell processes (C rebars) displayed very severe corrosion attack, with 389 marked section loss. This means that the large electrons demanded by carbon fibre mesh 390 brought about greater steel oxidation. Conversely, the rebars that were not electrically 391 connected to mesh (NC rebars) did not display appreciable damage. For bigger covers or other 392 better-quality concretes, this effect was not visually noticeable because the corrosion that 393 occurred during the testing period was very slight or even negligible in many cases.

![](_page_18_Figure_0.jpeg)

394 395

Figure 15- State that the different rebars were in

# 396 <u>3.6.- Gravimetric analysis</u>

The gravimetric test is the reference for the correct calibration of the corrosion rate measurement following the RILEM Recommendation of TC 154-EMC (2005). After being exposed to a marine environment for more than 320 days, the rebars that had been weighed before specimens were manufactured were weighed again. In Figure 16 the real mass loss of rebars is represented (gravimetric test) vs. the theoretical loss obtained by Eq. (5) in the two following cases: a) considering only microcell processes ( $i_{CORR,MICRO}$ ); b) considering the total corrosion current ( $i_{CORR}$ ) obtained from the sum of ( $i_{CORR,MICRO}$ ) and ( $i_{CORR,MACRO}$ ).

![](_page_19_Figure_0.jpeg)

405

Figure 16- Gravimetric comparison

406 When only considering microcell processes, the theoretical values tended to underestimate 407 real mass loss due to corrosion by 90%. Conversely, when the analysis included macrocell 408 processes, a good fit appeared between the theoretical and real values, which deviated by 5%. 409 The big difference between both cases was caused by using a large cathodic surface because it 410 increased the importance of macrocell processes by recreating conditions that would take 411 place in densely reinforced areas. This fact demonstrates that monitoring rebars by measuring 412 only the *i*<sub>CORR,MICRO</sub> can lead to mistakes and uncertain estimations of structures' service life, 413 which would underestimate damage to zones affected by chlorides. Therefore, both microcell 414 and macrocell currents must be considered when designing embedded corrosion monitoring 415 systems to accurately control reinforced concrete structures located in marine environments.

## 416 **5.- Conclusions**

The following conclusions can be drawn from this study on the influence that macrocell currents have on monitoring reinforced concrete structures exposed to marine environments for almost 1 year. This research was carried out using a large cathodic surface that simulates the conditions that would take place in densely reinforced areas:

- 421 -the estimation done from Fick's second law of the time needed for chlorides to reach a rebar422 and for corrosion onset provided good matches to reality;
- +the existence of macrocell currents generates greater anodic polarisation in active rebar
  zones, which favours chloride migration (negative electrical charge) towards the steel surface.
  As a result of this, corrosion onset occurs earlier, the concentration of the chlorides around the
  rebar is higher and the corrosion rate increases;
- -when a rebar is not depassivated, steel mass loss is negligible due to macrocell currents. However, upon corrosion onset, the influence of these currents is greater the worse concrete quality is and the smaller the concrete cover is. In concrete C30, considering macrocell currents, or not, leads to differences in the  $i_{CORR}$  values of almost two orders of magnitude, and in concrete C50 of one order of magnitude. As a result of this, for concrete C30 and the 5 mm cover, mass loss was 50-fold higher than in the rebars not affected by macrocell currents;

-if, as is normally the case of many monitoring systems, the corrosion rate is determined only
by measuring microcell currents (local corrosion), the obtained values tend to underestimate
the real mass loss due to corrosion by, in this case, 90%. Conversely, when the analysis
includes macrocell processes, a good match between theoretical and real values is obtained.
Therefore, both microcell and macrocell currents must be considered when designing
corrosion monitoring systems to accurately control reinforced concrete structures.

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