

Corrosion rate measurements of steel in concrete – Evaluation of a new algorithm for analysis of galvanostatic potential transients

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ABSTRACT:

For concrete structures where reinforcement corrosion is the main degradation mechanism, reliable non-destructive techniques for assessment of the corrosion state of reinforcement are required. Instruments for field measurements of the corrosion rate in reinforced concrete have been used during the last decades. However, both laboratory and field studies have shown that the measured corrosion rates are strongly affected by the measurement parameters used. Several studies have shown that the reason for this is related to the equivalent system used in the instruments for describing the electrical behaviour of the steel-concrete system. To mitigate this, a new equivalent system and corresponding algorithm for analysis of galvanostatic potential transients has been developed. The algorithm has been adopted in a new instrument for on-site corrosion rate measurements. This paper describes the initial steps and first results of the evaluation of the newly developed algorithm.

1 INTRODUCTION

For concrete structures where reinforcement corrosion is the main degradation mechanism periodical condition assessments are essential to optimize the maintenance. In this connection, reliable non-destructive techniques for assessment of the corrosion state and rate of the reinforcement are required. Technical recommendations for corrosion rate measurements have been published, Andrade et al. (2004); but no standards describing the procedure to be followed or guidelines for interpretation of measurements exist. The corrosion rate, often expressed as the corrosion current density, i_{corr} , is determined by measuring the polarization resistance, R_p , and using the empirical Stern-Geary relationship, Stern & Geary (1957):

$$i_{corr} = \frac{B}{R_p \times A} \quad (1)$$

Where B is a proportionality factor that depends on the anodic and cathodic Tafel slopes and A is the polarized surface area on the reinforcement.

Several steady and non-steady (transient) electrochemical techniques for determining the polarization resistance, R_p , of steel in concrete exist: linear polarization resistance (LPR), Gonzales et al. (1980), Millard et al. (1992), electrochemical impedance, John et al. (1981) and galvanostatic pulse measurements, Elsener et al. (1997), Elsener (2005). Of these, only few have been adopted in instruments for on-site measurements. In the instrument GalvaPulse, which has been one of the two most widely used commercial instruments for on-site measurements during

the last decade, Nygaard et al. (2009), the galvanostatic potential transient technique is used for determining the polarisation resistance, R_p , and thus the corrosion current density, i_{corr} . This technique assumes that a simple Randles circuit, Gabrielli et al. (1979) describes the potential response, E_t , of a steel-concrete system as function of time when a galvanostatic current, I_{CE} , is applied. Under this assumption the potential response, E_t , as a function of the polarization time, t_p , can be expressed by, Elsener et al. (1997), Elsener (2005), Gabrielli (1979):

$$E_t = I_{CE} \left(R_{\Omega} + R_p \left(1 - e^{-t_p / R_p C_{dl}} \right) \right) \quad (2)$$

where R_{Ω} is the Ohmic system resistance (IR drop) and C_{dl} the double layer capacitance. Two methods are typically used for obtaining R_p from Equation 2 when analyzing a measured potential transient: a linearization, Elsener et al. (1997), Elsener (2005) and a curve-fitting procedure, Elsener et al. (1997), Elsener (2005), Luping (2002). It has been shown that over a wide range of polarization resistances, R_p , very similar values are obtained with both procedures, Elsener et al. (1997). Due to the lower computational power required the linearization procedure is used in the GalvaPulse instrument.

In addition to the electrochemical technique for determining the polarization resistance, R_p , the GalvaPulse as well as most other commercially available instruments used during the last decade makes use of a so-called confinement technique. The confinement technique should in principle control the current distribution from the electrode placed on the concrete surface to the embedded reinforcement and thus determine the polarized steel surface area, A . The various confinement techniques used in the different commercially available corrosion rate instruments are described in detail in many publications, including e.g. Nygaard et al. (2008), Luping (2002) and Nygaard (2009).

Both on-site investigation and laboratory studies have shown that significantly different corrosion rates are obtained when different commercially instruments are used Nygaard (2008), Gepraegs & Hansson (2004), Flis et al. (1993), Flis et al. (1995). For both the galvanostatic potential transient technique as used in the GalvaPulse instrument and other techniques used in instruments for on-site use it has been shown that the measured corrosion rates are highly affected by the chosen measurement time and current. To mitigate this, a new algorithm based on a modified Randles system incorporating a Constant Phase Element (CPE) describing the non-ideal capacitive behaviour of the steel-concrete system has been developed and implemented in a new hand-held instrument for on-site use – the CorroMap. No current confinement is used in the instrument. A detailed description of the modified equivalent circuit on which the algorithm is based can be found in Feliu et al. (1998) and Feliu (2004). Based on results from earlier unpublished studies the newly developed algorithm should significantly decrease the effect of measurement time and current on the measured polarisation resistance, R_p , and thus corrosion current density, i_{corr} .

This paper presents the initial steps and first results of the evaluation of the newly developed algorithm. The performance of the algorithm and thus the hand-held instrument are as a first step evaluated through series of comparative measurements on concrete slabs with passively and actively corroding segmented reinforcement bars. Surface measurements of the corrosion rate of the embedded bars are made with the new and a first-generation corrosion rate instrument (the GalvaPulse) and compared with macro-cell current measurements assumed to provide information on the actual corrosion state and rate of the embedded bars.

2 EXPERIMENTAL

The test specimens with segmented reinforcement bars used in the study were fabricated in 2006 for a study on the effect of confinement techniques Nygaard (2008). Since then, the specimens have been stored under normal laboratory conditions and prior to the investigations presented in this paper, re-conditioned at 20 °C and 90 % relative humidity and kept under these conditions during the tests.

2.1 *Preparation of test specimens*

For the investigations three concrete specimens with varying amount of admixed chloride were used: Specimen 1: 0 %, Specimen 2: 1.5 % and Specimen 3: 4 % chloride by mass of cement. Each test specimen consisted of a rectangular concrete slab ($1.5 \times 0.12 \times 0.5 \text{ m}^3$) with two segmented reinforcement bars and three embedded MnO_2 reference electrodes as shown in figure 1. White Portland cement (CEM I 52.5) and a w/c ratio of 0.45 was used for the concrete. Details of the mix design, cement composition and the fresh and hardened properties of the concrete mixes are given in Nygaard (2008). After casting, the specimens were kept in the moulds for one day, demoulded and stored at 20 °C and 95 % relative humidity for almost one year. Following this, the specimens were stored five years at normal indoor laboratory conditions after which the specimens were reconditioned at 20 °C and 95 % for three months before the testing was started.

In each concrete specimen two 12 mm diameter segmented reinforcement bars were embedded; an upper bar simulating passive (0 % chloride) and intense localised corrosion (1.5 and 4 % chloride), and a lower bar simulating passive (0 % chloride) and general corrosion (1.5 and 4 % chloride), both with cover depths of 30 and 75 mm. The segmented reinforcement bars were prepared by mounting a combination of carbon and stainless steel segments, i.e. circular steel rings on a non-conducting fibreglass bar. This bar contained a slot for the connecting wires; one 0.05 mm^2 wire was soldered to the inside of each segment, allowing for external connection. Silicone washers with a thickness of 1 mm were placed between the steel segments, electrically isolating the segments and sealing the reinforcement bar system. A detailed description of the segmented reinforcement bars can be found in Nygaard (2008).

2.2 *Experimental approach*

Immediately after placing the test specimens in the climate chamber at 20 °C and 90 % relative humidity for re-conditioning, all segments on each reinforcement bar were connected to a switchboard. Apart from connecting the segments making each bar act as an electrical continuous reinforcement bar, the switchboard allowed the macro-cell current running to or from the individual segments to be measured without disconnecting the segments at any time. For the initial evaluation of the newly developed algorithm and with this the CorroMap instrument, the full series of measurements described in the following were repeated twice over a period of two months.

2.2.1 Surface measurements of the corrosion rate

Measurements were made with the GalvaPulse (with and without current confinement) and the CorroMap instruments along and directly above each segmented reinforcement bar (upper and lower bar in Specimens 1 to 3) on both sides of the specimens (cover of 30 mm and 75 mm, respectively). Along each bar the measurements were made with a spacing of 50 mm, resulting in a total of 29 measurement points per segmented reinforcement bar per side. With the GalvaPulse instrument measurements were made with and without current confinement as

mentioned above. For all measurements a polarisation time of 10 seconds was used. On Specimen 1 with 0 % chloride and thus passive reinforcement a polarisation current of $10\ \mu\text{A}$ was used, whereas a current of $20\ \mu\text{A}$ was used for the measurements on Specimens 2 and 3 with 1.5 and 4 % chloride, respectively. For all measurements, i.e. with and without current confinement the polarised steel area was set to $26.4\ \text{cm}^2$, corresponding to the assumed confinement length of 70 mm and the reinforcement diameter of 12 mm.

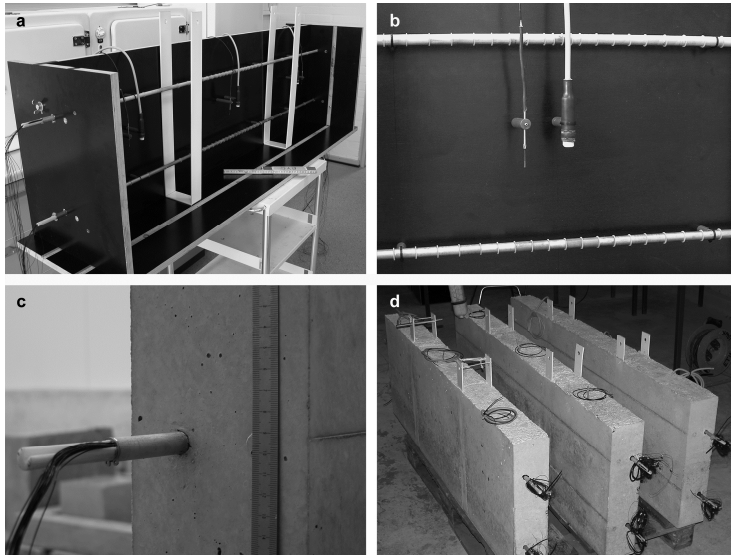


Figure 1: Manufacture of the test specimens. a: Segmented reinforcement bars, reference electrodes and lifting frames are mounted in the mould (side removed for better view). b: All reinforcement segments are isolated with silicone rings. c: The fibreglass bar with the connecting wires and a part of the end of the 160 mm end-segment protruding from the specimen. d: The three test specimens after production. From Nygaard (2008).

For the CorroMap measurements the same polarisation currents and time were used as with the GalvaPulse instrument. It should be noted that the polarisation time of 10 seconds is fixed in the CorroMap instrument and cannot be changed by the user. The polarised steel area was set to $22.6\ \text{cm}^2$, corresponding to the diameter of the counter-electrode (default setting), and a reinforcement diameter of 12 mm.

After each measurement with one of the instruments, the segmented reinforcement bar was allowed to depolarise to the initial equilibration potential, E_{corr} , before a new measurement was initiated. This was checked by measuring the potential of the reinforcement bar versus the embedded MnO_2 reference electrodes.

2.2.2 Macro-cell current measurements

Before and after all surface measurements were made on a segmented reinforcement bar, the macro-cell currents running from or to each segment on the bar were measured. The measurements were made by inserting a zero-resistance ammeter in the connection on the switch board to the individual segments. Insertion of the zero-resistance ammeter was done without electrically disconnecting the segments at any time so as not to disturb the electrochemical system. The zero-resistance ammeter used had a current range of $\pm 1\ \text{mA}$ and a resolution of $0.1\ \mu\text{A}$. From the measured absolute macro-cell current the macro-cell current density, i_{corr} , was calculated for each segment using the length and diameter of the considered segment.

3 RESULTS AND DISCUSSION

Below selected results are presented. To illustrate the performance of the algorithm and thus the hand-held instrument when measuring on passive reinforcement, the results from the lower reinforcement bar in Specimen 1 are included. In addition to this, the performance when measuring on reinforcement with active localised corrosion (pitting) and active general corrosion is illustrated by the results from the measurements on the upper and lower reinforcement bars, respectively, in Specimen 3. For both Specimen 1 and 3 only the results from the surface measurements on the side with 30 mm cover are included. The presented results are consistent with the full series of measurements on both sides of the specimens (30 and 75 mm concrete cover) repeated twice over a period over two months.

3.1 *Passive reinforcement*

3.1.1 Half-cell potential measurements

The half-cell potential, E_{corr} , corrosion current density, i_{corr} , and ohmic resistance, R_{Ω} , values obtained with the GalvaPulse and CorroMap on the lower segmented bar in Specimen 1 with 0 % chloride and thus passive reinforcement are shown in figure 2. No macro-cell currents running between the segments could be measured due to their passivity. With the GalvaPulse half-cell potentials, E_{corr} , in the range from -25 to -75 mV versus Ag/AgCl were measured along the passive reinforcement bar, whereas slightly more negative values ranging from -50 to -100 mV versus Ag/AgCl were measured with the CorroMap, see figure 2, top graph. This indicates that there may have been a slight potential difference between the reference electrodes in the two instruments at the time of measurement although both are Ag/AgCl electrodes. Considering the interpretation guidelines in ASTM C 866 (1977) the majority of the measured half-cell potentials indicate 90 % probability of no corrosion (more positive than -83 mV versus Ag/AgCl).

3.1.2 Ohmic resistance

When it comes to evaluation of the algorithm used in the CorroMap instrument, the ohmic resistances, R_{Ω} , and corrosion current densities, i_{corr} , are the most interesting parameters as both are output parameters from analysis of the measured potential transient (in contrast to the E_{corr} values). Although, a significant scatter was observed in the determined ohmic resistances, R_{Ω} , along the reinforcement bar, it is evident that lower R_{Ω} values were generally obtained with the CorroMap and the GalvaPulse when no current confinement was used, than with the GalvaPulse when using current confinement, see figure 2, middle and bottom graphs. The reason for the differences can be explained by the current confinement technique used in the GalvaPulse. As described in Nygaard (2008) the GalvaPulse instrument applies a guard-ring current of same size as the counter-electrode current (in addition to this) when current confinement is used with the intent of controlling the polarised area of the reinforcement. Thus, when current confinement is used the total current applied is double as when no confinement is used. However, in the analysis of the potential transient only the counter-electrode current is considered. As the ohmic resistance, R_{Ω} , is calculated from Ohms law using the potential response, E_{Ω} , measured immediately after initiating the counter-electrode current pulse (and potentially the confinement current pulse), it is clear that the use of current confinement has a significant effect on the determined ohmic resistance, R_{Ω} .

3.1.3 Corrosion current density

A clear effect of the current confinement was also observed from the measured corrosion current densities, i_{corr} . When measuring with the GalvaPulse without current confinement and the CorroMap corrosion current densities, i_{corr} , in the range from approximately 0.2 to 1.4 $\mu\text{A}/\text{cm}^2$ and 0.3 to 1.0 $\mu\text{A}/\text{cm}^2$, respectively, were obtained. Much lower values (approximately half) in the range from 0.07 to 0.6 $\mu\text{A}/\text{cm}^2$ were obtained with the GalvaPulse when using current confinement, see figure 2, bottom graph. As for the ohmic resistance, R_Ω , the differences in measured corrosion current densities, i_{corr} , is a result of the current confinement.

It is interesting to see that very similar corrosion current densities, i_{corr} , were obtained with the CorroMap and GalvaPulse without current confinement. This basically shows that with the polarisation time and current used (10 sec and 10 μA , respectively for both instruments) very similar corrosion current densities, i_{corr} , are measured with the two instruments. However, this may not - and with the newly developed algorithm should not - be the case over a wide range of polarisation times and currents. As shown in Nygaard (2008) the polarisation resistance, R_p , and thus the corrosion current density, i_{corr} , determined with Equation 2 based on the simple Randles system is strongly affected by the polarisation time and current used. For passive reinforcement the measured corrosion rate is often seen to decrease with a factor 10 or more when increasing the polarisation time from e.g. 10 to 60 seconds. As described earlier, the effect of time and current on the newly developed algorithm based on a modified, i.e. more complex version of the simple Randles system is expected to mitigate these effects. This will be investigated thoroughly by a parameter study in a future project.

When considering the corrosion current densities, i_{corr} , measured without current confinement, i.e. with the CorroMap and GalvaPulse without confinement (see figure 2, bottom graph) it is evident that the values measured are significantly higher than those normally reported in the literature for passive steel in concrete (approximately 0.1 $\mu\text{A}/\text{cm}^2$ or less), Gowers et al. (1994). This is because the current applied from a small counter-electrode placed on the concrete surface spreads laterally over a large length of the passive reinforcement due to the high polarisation resistance, R_p , i.e. low corrosion current density, i_{corr} , of the embedded steel, Gepraegs & Hansson (2004). As a result of the lateral current spread only a fraction of the applied counter-electrode current enters the assumed polarisation area on the steel and thus a much lower polarisation, i.e. charging is obtained (in that area). In the analysis of the measured potential transient, the actual, i.e. lower current entering the assumed polarisation area or the actual polarised area being much larger than the assumed area cannot be taken into consideration and thus too high corrosion current densities, i_{corr} , are obtained. The different current confinement techniques used in earlier instruments, i.e. 1st generation instruments like the Galvapulse were aimed at solving this problem. However, as shown in numerous publications their functionality and efficiency are questionable, Nygaard (2008) and literature cited herein. Thus, when measuring on real-size structures an overestimation of the corrosion rate of the passive steel is inevitable.

3.2 Actively corroding reinforcement

3.2.1 Half-Cell Potential and Ohmic Resistance Measurements

The half-cell potential, E_{corr} , corrosion current density, i_{corr} , and ohmic resistance, R_Ω , values obtained with the GalvaPulse and CorroMap on the upper and lower segmented bar in Specimen 3 with 4 % chloride are shown in figure 3, left and right graph, respectively. In both graphs, i.e. for both reinforcement bars the anodes found to be anodic (actively corroding) from the macro-cell current measurements are shown with black bold lines on the first axis and the anodic

current density given. The red bold lines indicate the position and extent of the individual anodic segments.

For the upper segmented reinforcement bar (figure 3, left graph) the half-cell potentials, E_{corr} , measured along the bar were in the range from -255 to -296 mV versus Ag/AgCl, and no significant differences were observed between the values measured with the GalvaPulse and the CorroMap. Along the lower segmented reinforcement bar (figure 3, right graph) more negative half-cell potentials were measured in the range from -300 to -366 mV versus Ag/AgCl. Also here, no significant potential variations were measured along the bar. According to the guidelines in ASTM C 876 (1977) all half-cell potential values on the upper as well as the lower segmented reinforcement bar indicated 90 % probability of corrosion (more negative than -233 mV versus Ag/AgCl).

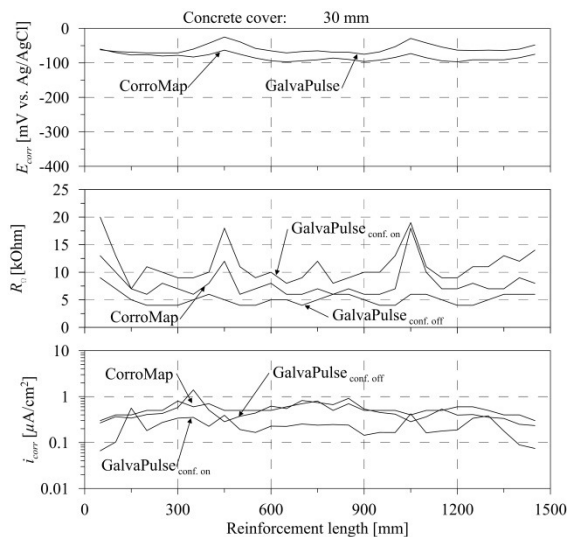


Figure 2: Specimen 1 with 0 % chloride, lower reinforcement bar: Half-cell potentials, E_{corr} , (top), ohmic resistances, R_{ohm} , (middle) and corrosion current densities, i_{corr} , (bottom) measured on the surface with 30 mm concrete cover with the GalvaPulse and CorroMap.

For both the upper and the lower reinforcement bar it was seen that the measured half-cell potential values were more or less constant along the bars without any local variations near or around the actively corroding anodes. This must be a result of the low concrete resistivity (due to the mixed-in chloride) making the anodes able to polarise all cathodic segments on the bars. The low concrete resistivity was evident from the measured ohmic resistances, R_{ohm} : With both instruments the ohmic resistance values were in the range from 0.5 to 2 kOhm, see figure 3, middle graphs.

3.2.2 Corrosion current density

On the upper bar (figure 3, left graph) with a single 10 mm long centrally placed actively corroding segment corrosion current densities, i_{corr} , varying with a factor of approximately 2 were measured along the bar with both the CorroMap and the GalvaPulse with and without confinement: With the CorroMap and GalvaPulse without confinement i_{corr} values of 1.5 and 1.1 $\mu\text{A}/\text{cm}^2$, respectively, were measured directly above the corroding anode, whereas values from approximately 0.5 to 0.8 $\mu\text{A}/\text{cm}^2$ were measured at the ends of the reinforcement bar (over passive reinforcement). With current confinement the measured i_{corr} values were approximately

half of the values obtained without confinement, i.e. approximately $0.5 \mu\text{A}/\text{cm}^2$ directly above the corroding anode and $0.25 \mu\text{A}/\text{cm}^2$ at the ends of the reinforcement bar. The same trends were observed for the lower bar, however, higher corrosion current densities, i_{corr} , were in general measured due to the higher corrosion activity (number of corroding segments and corrosion rate).

For both the upper and the lower reinforcement bar the use of current confinement did not change the pattern of the measured corrosion current density, i_{corr} , along the bars making localization of the individual anodes easier. The only effect of the current confinement was observed as a shift in the measured corrosion current density, i_{corr} , with a factor of approximately 0.5. This is in agreement with observations in earlier studies, Nygaard (2008), Nygaard et al. (2009). The relatively small variation in the measured corrosion current density, i_{corr} , along the segmented reinforcement bars with (discrete) actively corroding anodes is most likely a result of the phenomenon often referred to as self-confinement. Self-confinement basically occurs as the current applied from a small counter-electrode on the concrete surface follows the path of lowest resistance to the embedded steel reinforcement: On reinforcement with discrete actively corroding areas the current from the counter-electrode therefore flows laterally through the concrete and into the active areas due to their low polarisation resistance, R_p , Gepreags & Hansson (2004), Nygaard (2009). As a result of the self-confinement an exact calculation of the polarisation resistance, R_p , and thus the corrosion current density, i_{corr} , requiring knowledge of the polarised area, A , cannot be achieved.

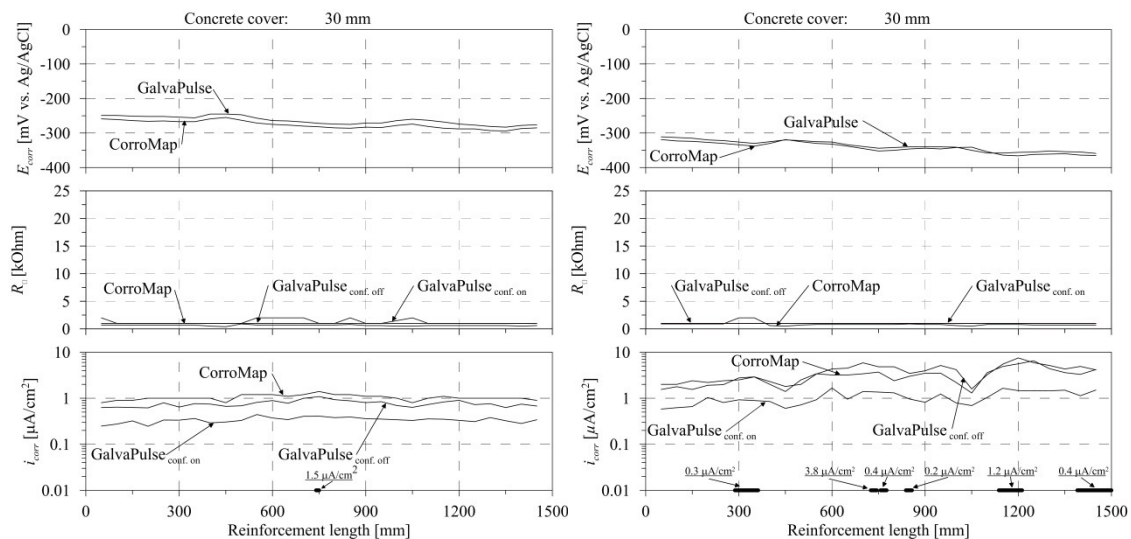


Figure 3: Specimen 3 with 4 % chloride, upper (left) and lower (right) segmented reinforcement bar: Half-cell potentials, E_{corr} (top), ohmic resistances, R_{ohm} (middle) and corrosion current densities, i_{corr} (bottom) measured on the surface with 30 mm concrete cover with the GalvaPulse and CorroMap. The black bold lines/dots on the first axis indicate the position and extent of the anodic areas on the segmented reinforcement bars. The current density given for the anodes were determined from the macro-cell current measurements.

In spite of this a good indication of the corrosion state and rate of the embedded segmented reinforcement bars was obtained with the CorroMap and GalvaPulse when comparing the measurements on the upper and lower segmented bar in Specimen 3: on the lower bar markedly higher corrosion current densities were obtained reflecting the larger extent and higher corrosion rates of the segments. As mentioned earlier for the passive reinforcement bar in Specimen 1

very similar corrosion current densities, i_{corr} , were obtained with the CorroMap and GalvaPulse without current confinement on both the upper and lower segmented reinforcement bars. Again, this basically shows that with the polarisation time and current used (10 sec and 20 μ A, respectively for both instruments) very similar corrosion current densities, i_{corr} , are measured with the two instruments. The similar i_{corr} values obtained with the CorroMap and GalvaPulse on both passive and actively corroding reinforcement, with varying corrosion extent and rate could indicate that no significant difference exist between the old algorithm based on the simple Randles circuit and the newly developed algorithm based on the modified Randles circuit. However, for all measurements a polarisation time of 10 seconds has been used as this has been recommended in earlier studies on galvanostatic potential transient measurements, Nygaard (2008), Nygaard (2009), Luping (2002).

Based on the consistent results obtained with the newly developed algorithm and hand-held instrument a detailed study on the effect of polarisation time and current on the measured corrosion current density, i_{corr} , will be initiated in order to investigate the reliability, possibilities and limitations of the newly developed algorithm.

4 CONCLUSIONS

A new algorithm based on a modified Randles system incorporating a Constant Phase Element (CPE) has been developed and implemented in a new hand-held instrument for on-site corrosion rate measurements. The performance of the new algorithm and thus the new instrument has been evaluated through series of comparative measurements on concrete slabs with passive and actively corroding segmented reinforcement bars. The comparative studies comprising measurements of half-cell potential, ohmic resistance and corrosion current density were performed with the newly developed instrument – the CorroMap and a first-generation instrument – the GalvaPulse - with and without current confinement.

Lower values of ohmic resistance were measured by means of the new CorroMap instrument without current confinement than by means of the old GalvaPulse instrument using current confinement. The reason for this difference is the additional current applied from the guard ring in order to confine the counter-electrode current.

Similar corrosion current densities were measured by the new CorroMap and the old GalvaPulse instrument without current confinement on both passive and actively corroding reinforcement. When measurements were performed with the old GalvaPulse instrument with current confinement much lower (approximately half) corrosion current densities were obtained on both passive and actively corroding reinforcement. As in case of the ohmic resistance the reason for the difference is the current applied from the guard ring (in addition to the counter-electrode current).

Based on the consistent results obtained with the newly developed algorithm and hand-held instrument a detailed study on the effect of polarisation time and current on the measured corrosion current density, i_{corr} , will be initiated in order to investigate the reliability, possibility and limitation of the newly developed algorithm.

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