Effect of environmental exposure conditions on the corrosion rate of carbon steel bars in carbonated concrete

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Abstract

In urban environments the propagation phase of corrosion can represent a significant part of the entire service life of a reinforced concrete structure. To properly evaluate its duration, the knowledge of the corrosion rate is essential. This paper reports the corrosion rate and corrosion potential of carbon steel bars embedded at different depths (i.e. 10, 25 and 40 mm) in concretes with different binders and water/binder ratio of 0.61, exposed both outdoor in Milan in unshel-tered conditions and in laboratory conditions characterized by different temperatures and relative humidity levels. Concrete resistivity at different depths is also reported. Relative humidity strong-ly influenced the corrosion potential, corrosion rate and resistivity, whilst temperature played an important role only at a high relative humidity level. Correlations between these parameters were analysed and discussed.

1 Introduction

The service life of reinforced concrete (RC) structures is limited by the corrosion of steel reinforcement that, when exposed in urban environment, is mainly due to concrete carbonation [1]. Usually attention is focused on the initiation phase, but, in these exposure conditions, the propagation phase can represent a significant part of the service life and, hence, it has to be properly designed. A correct design of the propagation phase starts from the knowledge of the corrosion current density (that represents the rate of occurance of the corrosion process, i.e. the corrosion rate), which can vary even significantly changing the environmental exposure conditions and the type of concrete.

As far as the impact of the exposure conditions on the corrosion current density, in outdoor exposure conditions, unsheltered from rain, highly variable values, between 1 to 30 mA/m², were determined [2]-[3]. Laboratory tests aimed at reproducing the unsheltered exposure conditions, i.e. keeping the specimens partially or fully immersed for long time or exposing the specimens to wet/dry cycles, confirmed results obtained outdoor. In laboratory, also other exposure conditions were simulated, by varying the relative humidity from 0 to 100%. In the 95-100% R.H. range, corrosion current density between 2 and 30 mA/m² was detected by several Authors [4]-[7]. The decrease of the relative humidity led to a decrease of the corrosion current density, that approached values between 2 and 10 mA/m² in the 80-90% R.H. range [8],[9]. A further decrease, to values lower to 1 mA/m², was observed in the 60-75% R.H. range [2],[10]. As it can be observed, usually the corrosion behaviour of steel is investigated by means of laboratory conditions rather than through real outdoor exposure conditions.

As far as the effect of the type of concrete on the corrosion current density, it is hard to be defined, since tests on concrete made with Portland cement were usually carried out. However due to the growing interest in supplementary cementitious materials, SCMs, to improve the sustainability of reinforced concrete structures, the influence of the type of binder on the corrosion current density in different exposure conditions needs to be investigated [11].

This paper reports an experimental study aimed at evaluating the corrosion behaviour of carbon steel bars embedded in concretes made with water/binder ratio of 0.61 and Portland cement and different SCMs (limestone, fly ash, natural pozzolan and ground granulated blast furnace slag). Concretes were exposed both in unsheltered outdoor exposure conditions and in controlled laboratory conditions, varying the relative humidity from 80 to 100% and the temperature from 20 to 40°C. In particular the corrosion potential, the corrosion current density and the concrete electrical resistivity at different depths were monitored in time.

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2 Experimental procedure

Portland cement, CEM I 52.5R (OPC), and five blended cements, obtained by replacing part of the cement with SCMs, in particular with 15% and 30% of ground limestone (15LI and 30LI), 30% of fly ash, 30% of natural pozzolan (PZ) and 70% of ground granulated blast-furnace slag (GGBS) were used to produce six concretes with water/binder ratio of 0.61 and binder dosage of 300 kg/m³. 1857 kg/m³ of crushed limestone aggregate, with maximum size of 16 mm, were used and an acrylic superplasticizer was added to the mix to achieve a class of consistence *S4* according to EN 206 standard.

After mixing, concretes were cast in reinforced prismatic specimens, covered with a plastic sheet and stored in laboratory at 20°C, demoulded after 24 hours and cured, for other 6 days, at 20°C and 95% relative humidity. At the end of the curing, after maintaining the specimens in laboratory conditions for two weeks, they were exposed in a carbonation chamber, with 100% of CO₂, until they were fully carbonated (cores were periodically taken from the specimens to verify, by means of phenolphthalein tests, the carbonation).

The prismatic specimens, $60 \text{ mm} \times 250 \text{ mm} \times 150 \text{ mm}$, were reinforced with three ribbed carbon steel bars with a diameter of 10 mm and a cover depth of 10, 25 and 40 mm. Each specimen was equipped with stainless steel wires to be used as auxiliary electrodes in the electrochemical measurements (Fig. 1). Two specimens for each concrete were made. After masking the lateral surfaces of the specimens and the external parts of the rebars with an epoxy coating, one specimen for each concrete was exposed, for about two years, outdoor in unsheltered natural conditions (on the roof of the Department of Chemistry, Materials and Chemical Engineering of Politecnico di Milano), where average daily temperature varyed between -5 and 30°C, average daily relative humidity between 30 and 100% and the rainfall reached values up to 38 mm. The other specimen was exposed to controlled cycles of temperature and relative humidity. In particular, the following exposure conditions of temperature (T) and relative humidity (R.H.) were imposed for 2-4 weeks until stable conditions were reached: T = 20°C/ R.H. = 80%, T = 20°C/ R.H. = 90%, T = 20°C/ R.H. = 95\%, T = 20°C/submerged; T = 40°C/ R.H. = 80%, T = 40°C/ R.H. = 90%.

During the exposure period, electrochemical measurements of half-cell potential of steel (E_{corr}) versus a saturated calomel electrode (SCE), placed on the specimen surface in the central part of each bar, and linear polarization resistance measurements (R_p) were carried out to monitor the corrosion behavior of steel. From R_p measurements, corrosion current density, icorr, was determined as: icorr = B/(R_p), where B was assumed equal to 26 mV. Concrete electrical resistivity was also measured between the two wires placed at the different depths, through measurements of condictivity.



Fig. 1 Geometry of reinforced concrete specimens (dimension in mm).

3 Results and discussion

Figs. 2 and 3 show, as examples, the trend of corrosion potential and corrosion current density (left) of the three rebars embedded at different depths in concrete made with 30LI exposed outdoor in unsheltered conditions for approximately 2 years (Fig. 2) and to controlled cycles of temperature and relative humidity (Fig. 3). Moreover the electrical resistivity of concrete at the bars depths is also shown (right). For the concrete exposed outside, the three parameters fluctuated in time, due to the variation of the climate conditions. For instance, E_{corr} varied from around -450 mV to +200 mV vs SCE, i_{corr} from 0.01 to 13 mA/m² and ρ from 130 to 14000 Ω ·m. Different peaks can be observed with low values of corrosion current density (i.e. lower than 0.1 mA/m²), high values of corrosion potential (higher than 0 mV

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vs SCE) and high values of concrete electrical resistivity. These peaks corresponded to the drier periods, characterized by high temperature (around 30°C), low relative humidity (lower than 60%) and absence of rainfall and suggested that when concrete was dry corrosion slowly propagated. Conversely, peaks with low values of E_{corr} and ρ and quite high values of i_{corr} were detected in the more rainy periods, clearly suggesting that bars were heavily corroding.



Fig. 2 Trend in time of the corrosion potential (diamond symbols) and corrosion current density (triangular symbols) (left) measured on the bars embedded at different depths and of concrete electrical resistivity (right) measured at the bar depth in concrete made with 30LI exposed in outdoor unsheltered conditions (black symbols: upper depth; grey symbols: middle depth; white symbols: lower depth).



Fig. 3 Trend in time of the corrosion potential (diamond symbols) and corrosion current density (triangular symbols) (left) measured on the bars embedded at different depths and of concrete electrical resistivity (right) measured at the bar depth in concrete made with 30LI exposed to different environmental conditions: $T = 20^{\circ}C/R.H. = 95\%$, a; $T = 20^{\circ}C/H =$ submerged, b; $T = 20^{\circ}C/R.H. = 90\%$, c; $T = 40^{\circ}C/R.H. = 90$, d; $T = 20^{\circ}C/R.H. = 80\%$, e; $T = 40^{\circ}C/R.H. = 80\%$, f (black symbols: upper depth; grey symbol: middle depth; white symbols: lower depth).

At the beginning of the exposure to controlled cycles (Fig. 3), i.e. at the end of the exposure in the accelerated carbonation chamber (T = 20°C, R.H. = 65%), high electrical resistivity (higher than 10000 $\Omega \cdot m$), quite low corrosion current density (of the order of 0.1 mA/m²) and quite high corrosion potential (around 0 mV vs SCE) were detected. The exposure to an environment with high relative humidity, i.e. Federica Lollini1, Elena Redaelli1 3 95%, led to a decrease of the electrical resistivity, which reached values of the order of 2000 Ω -m, an increase of the corrosion current density which, however, remained lower than 2 mA/m² and a decrease of the corrosion potential. Variations of the corrosion conditions occurred after the exposure to the other controlled environments. The higher corrosion current density, and the lower electrical resistivity, were determined in submerged condition, where values of i_{corr} also higher than 10 mA/m² were measured. In all the environmental conditions, comparable values of electrical resistivity, corrosion current density and corrosion potential were determined at the different depths, suggesting that similar humidity and corrosion conditions were present.

The effect of the exposure conditions and concrete composition on the corrosion potential, corrosion current density and concrete electrical resistivity was investigated by evaluating, for each concrete and condition, the average values and the range of variability. Average values of the three parameters were evaluated considering values obtained on all the three rebars and the variability was evaluated considering the maximum and minimum values reached in each exposure condition (for controlled conditions, the transient period was neglected).

Fig. 4 shows for all concretes the average values and the range of variation of E_{corr} , i_{corr} and ρ as a function of relative humidity and temperature; for comparison, values obtained outdoor are also shown. The exposure conditions and, in particular, the relative humidity strongly affected the three parameters. For the corrosion potential a decrease was observed when the relative humidity increased to 95% and in submerged conditions, where it approached values between -750 and -650 mV vs SCE. At 80% R.H. the corrosion potential was higher than -250 mV vs SCE (Fig. 4a). As far as the corrosion current density is concerned, values lower than 1-2 mA/m² were detected for relative humidity lower than 90%; an increase of the relative humidity led to an increase of i_{corr} up to values between 3 and 10 mA/m² measured when the specimens were submerged (Fig. 4b). Temperature affected the corrosion potential and the corrosion current density when relative humidity was 90%. For instance, at 90% R.H. corrosion current density increased from values between 0.4 and 1 mA/m² to values between 1.4 and 3.9 mA/m². Conversely when the relative humidity was 80%, corrosion current density lower than 0.5 mA/m² was detected for temperatures of both 20 and 40°C. As far as the concrete electrical resistivity is concerned, as expected, the lowest values were measured when concrete was submerged, whilst the highest values when the concretes were exposed to 80% R.H. Average values between 4000 and 6800 Ω ·m were detected for relative humidity of 90% that decreased to average values between 130 and 480 Ω ·m when the specimens were submerged (Fig. 4c). Values in between were observed in the other environmental exposure conditions. As for corrosion current density a significant effect of temperature was detected when the relative humidity was 90%; for instance, the electrical resistivity decreased from values between 1700 and 3600 Ω ·m to values between 215 to 900 Ω ·m when temperature increased from 20 to 40°C. In outdoor unsheltered conditions the values of E_{corr} , i_{corr} and ρ were intermediate to those detected in the different controlled laboratory conditions, indicating that the typical exposure conditions that can occur, at least in the temperate climate of Milan, were simulated through controlled cycles of temperature and relative humidity. For instance, the highest values of corrosion current density measured in outdoor conditions during the rainiest periods were similar to the highest values measured in submerged conditions. The flat exposure of the specimens in outdoor conditions fostered their wetting during rain events, leading the concrete to conditions similar to saturation. Conversely the lowest values were comparable with those measured with R.H. of 80%. The sole effect of temperature on specimens exposed outdoor was not easy to be detected; it is reasonable to assume that in hot periods, high temperatures were reached and kept inside the concrete, due to the low concrete thermal conductivity and solar irradiation, that could facilitate the water evaporation. Comparable values of concrete electrical resistivity can be observed between the outdoor exposure and the simulated laboratory conditions.

Fig. 4 also allows the evaluation of the role of the type of binder on the corrosion behaviour of carbon steel bars and on the concrete electrical resistivity. The lowest corrosion current densities were detected in all the exposure environments, including the outdoor conditions, on concrete made with Portland cement: for instance, in submerged conditions, average values of the order of 3 mA/m^2 were determined, whilst on the concretes made with blended cements values even double were obtained (Fig. 4b). On concrete made with OPC cement also the highest values of concrete electrical resistivity were measured. This might be due to the interruption of concrete curing after 7 days and the subsequent exposure to accelerated carbonation, that might have interrupted the hydration of pozzolanic and hydraulic binders, without resulting in the well-known pore refinement that usually leads to higher concrete electrical resistivity of these binders in comparison to OPC. The FA concrete showed, in the

different controlled exposure environments, higher resistivity and lower corrosion current density in comparison to 15LI, 30LI, PZ and GGBS concretes. The possible effect of concrete porosity on resistivity as well as corrosion behaviour of steel bars needs to be better investigated by considering concrete made with different water/binder ratio.





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For instance, in submerged conditions, corrosion current densities of 7, 8, 10, 9 and 9 mA/m² were respectively determined on FA, 15LI, 30LI, PZ and GGBS concretes.

Results previously presented showed that the type of binder has a significant impact on corrosion current density and this means that the duration of the propagation period can be significantly different using concretes made with different binders. To quantify the effect of the type of binder on the duration of the propagation period, a simplified probabilistic approach was applied. For structures exposed to carbonation induced corrosion, the propagation period can be defined as the time required to consume the steel bars of an amount that leads to the cracking of the concrete cover. Then, the probability of failure was evaluated through the following limit state equation:

$$p_f = P\{g < 0\} = P\{P_{lim} - v_{corr} \cdot t_n < 0\}$$
(1)

where P_{lim} in μm is the limit penetration of corrosion, v_{corr} in $\mu m/year$ is the corrosion rate and t_p the propagation time.

The corrosion rate was described through a beta distribution function, with mean values, *m*, determinated from the corrosion current density ($v_{corr} = 1.16 \cdot i_{corr}$) in outdoor unsheltered exposure conditions and a standard deviation of 0.25m (upper and lower limits equals for all the types of binder of 0.1 and 15 µm/year, in agreement with the minimum and maximum values detected outdoor, were considered). P_{lim} was described by means of a normal distribution with a mean value and a standard deviation respectively equal to 100 and 30 µm.

Fig. 5 shows the probability of failure as a function of the propagation time evaluated solving the limit state equation (1) through a Montecarlo method. The probability of failure, i.e. the probability of reaching the limit state, increased increasing the time. Assuming a target probability, P_0 , equal to 10% (red horizontal line in Fig. 5), the propagation time varied from about 18-20 years for GGBS, 30LI and PZ, to 24-26 years for 15LI and FA to 60 years for OPC. Hence, the use of a blended cement could halve or reduce even more the propagation time in comparison to the Portland cement in unsheltered exposure conditions. However it should be noted that, in the evaluation of the service life, the initiation time cannot be neglected and that in the evaluation of the propagation period a simplified limit state equation was used.



Fig. 5 Probability of failure as a function of the propagation times for concretes made with w/b ratio of 0.61 and different types of binder and indication of the target probability of failure, P₀ (red line).

As a matter of fact, the equation did not take into account the tensile strength of the different types of concrete that might affect the limit penetration of corrosion, i.e. the thickness of steel bar that has to be consumed to lead to the concrete cracking. In less resistant and more porous concretes, the amount of corrosion products needed to crack the concrete cover might be higher than those required in more resistant concrete, with the consequence of an increase of the propagation time, but a higher reduction of the cross section. In absence of values of corrosion current density to be used in a probabilistic approach for the evaluation of the propagation period, correlation with other parameters would be useful. At this regard, the correlations between i_{corr} and ρ and between i_{corr} were investigated and shown in Fig. 6. Fig. 6a reports the i_{corr} values obtained on the three rebars embedded at the different

depths and the ρ measured at the same bars depths. A linear correlation, in a logarithmic scale, can be observed between corrosion current density and concrete resistivity, suggesting that the rate of occurrence of the corrosion process is determined by the electrical resistivity of the concrete measured at the bar depth. This correlation seemed to be independent from the type of binder. Decreasing the electrical resistivity, the corrosion current density increased. For instance, for resistivity higher than 1000 Ω ·m the corrosion current density was lower than 1-2 mA/m² regardless the type of binder, whilst for resistivity lower than 200 Ω ·m the corrosion current density increased up to values higher than 8-10 mA/m². In Fig. 6b the relationship between the corrosion current density and the corrosion potential can be observed. A dispersion of data can be observed, however a correlation between the two parameters can be detected, which, again, seemed to be independent from the type of binder. Decreasing the corrosion potential, the corrosion current density increased. For instance, quite high values of corrosion potential of the order of -100/-200 mV vs SCE corresponded to corrosion current densities significantly lower than 1 mA/m². Corrosion current density of the order of 10 mA/m² was measured when the corrosion potential was lower than -600 mV vs SCE. The availability of such correlations, independent from the type of binder, would allow the determination of the corrosion current density from the measurement of corrosion potential or concrete resistivity that are easier to be determined, for instance in field, in comparison to corrosion current density.





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4 Conclusions

From the evaluation of the corrosion conditions of carbon steel bars embedded in 7-day cured carbonated concrete made with Portland cement and different SCMs and exposed both to unsheltered outdoor conditions and laboratory conditions with different temperatures and relative humidity levels, the following conclusions can be drawn:

- the exposure conditions and, in particular, the relative humidity, strongly influenced the corrosion potential, corrosion current density and concrete electrical resistivity. At 20°C negligible corrosion current density was measured up to relative humidity of 90%. Temperature played a role only in environments with a high relative humidity (i.e. 90%);

- corrosion parameters were affected by the type of binder and, at equal environmental exposure conditions, concrete made with Portland cement had the highest resistivity and the lowest corrosion current density. As a result the duration of the propagation time was, at least, halved in concrete made with Portland cement in comparison to concretes made with blended cements;

- corrosion current density can be estimated from concrete electrical resistivity evaluated at the same depth of the bars and from corrosion potential and the relationships were not significantly dependent on the type of binder.

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