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Corrosion behaviour of low-nickel austenitic stainless steels reinforcements: A comparative study in simulated pore solutions

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Abstract

The use of austenitic stainless steel reinforcements is one of the most reliable methods to assure the durability of reinforced concrete structures exposed to aggressive environments, but the initial cost of the material often limits its use in practice. Nickel is one of the alloying elements that raise most the cost of the stainless steels reinforcements. In this work the corrosion resistance of low-Ni 204Cu reinforcements is compared with that of more traditional austenitic stainless steels such as 304, 304L, 316, 316L and 316Ti. Polarization tests were carried out in simulated carbonated and non-carbonated pore solutions with different chloride concentrations. Results prove the very good corrosion behaviour of 204Cu stainless steel in carbonated media and in non-carbonated, chloride contaminated media. The corrosion resistance of low-Ni type 204Cu is only meaningfully lower than that of more traditional austenitic stainless steels in very highly aggressive solutions.

Keywords: Corrosion; Carbonation; Chloride; Reinforcement; Stainless steels

1. Introduction

Conventional steel reinforcements embedded in concrete are passive due to a thin protective oxide layer (about 10 nm) [1] that is formed on its surface in high alkaline media such as that contained in the pores of the concrete (pH about 12.6). However, these reinforcements suffer severe corrosion problems when the reinforced concrete structure is exposed to chloride contaminated environments and/or when the concrete cover is carbonated. Chlorides favour localized pitting corrosion of the steel, and carbonation a generalized attack. Independently of the corrosion mechanism, the presence of the oxides produced by corrosion on the surface of the reinforcements weakens the adherence to the concrete, and, since the specific volume of the corrosion products is higher than that of the non-corroded metal, these oxides generate stresses in the concrete. The concrete cover cracks easily because of its low tensile strength. Corrosion rates of the steel that can be moderated in other media (about 1 μA/cm^2) are inadmissible in concrete, because they cause cracking of the cover in less than 10 years [2]. Cracks in the concrete and the eventual detachment of part of the cover expose the steel to the atmosphere and accelerate the corrosion attack that can reduce the section of the reinforcements and lead to the failure of the structure.

The high cost of repairing corroded reinforced concrete structures calls for protective strategies that will be exposed to an aggressive environment. Galvanizing of the reinforcements prevents the corrosion of the steel in chloride contaminated concrete only for a short time [3,4]. Epoxy coating of the reinforcements weakens adhesion between metal and concrete, and does not protect the steel in highly chloride-contaminated environments [5,6]. Other options, such as the application of waterproof coatings on the concrete [7], the addition of corrosion inhibitors [8] or the realkalisation of structures that have lost their passivity [9], have also proved to have drawbacks. Cathodic protection and the use of stainless steel reinforcements seem to be the only methods whose protective efficiency is not questioned. Long term cathodic protection becomes an expensive method, because it needs continuous monitoring by specialized personal, it is therefore very interesting to know in depth the advantages and limitations of the use of stainless steel reinforcements.

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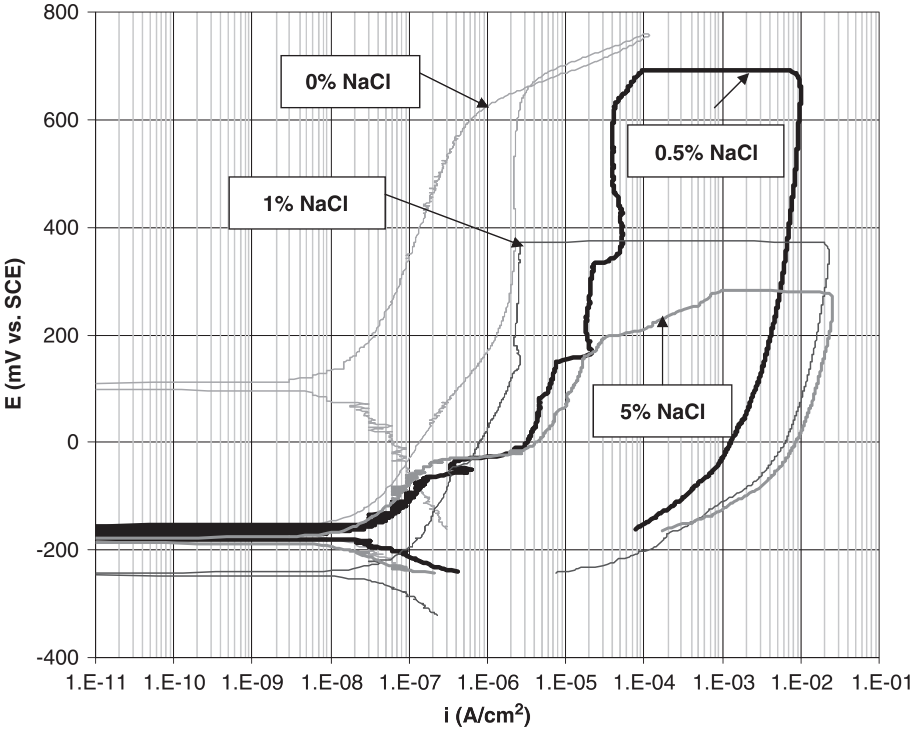

Fig. 1. Influence of the chloride concentration on the polarization curves of corrugated 204Cu in saturated, non-carbonated $Ca(OH)_2$ solutions.

Up to now, most of the published literature has focused on the most commercial austenitic stainless steel types, such as AISI 304 and 316 [10–17]. The results obtained have shown the very good corrosion resistance of these materials in chloride-bearing concrete or simulated pore solutions. Corrosion problems have been detected in these steels exposed to very aggressive media when they are welded [10]. The causes of the decrease of the corrosion resistance due to welding have been analysed by several authors [18,19]. Heat tints formed on the surface during the welding process are usually

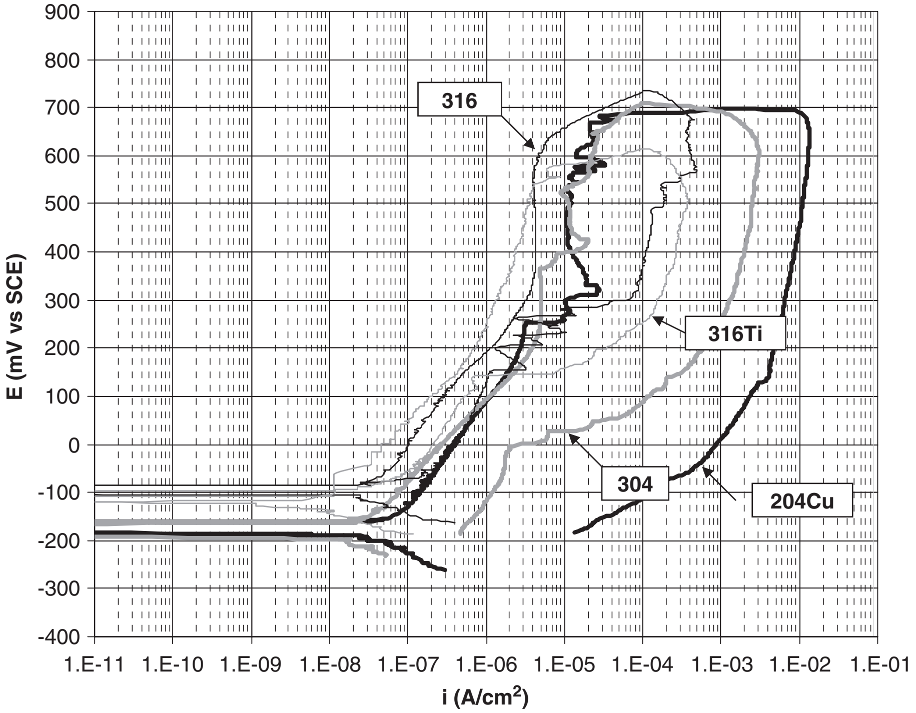

Fig. 2. Influence of the composition of the austenitic stainless steel on the polarization curves obtained in saturated, non-carbonated $Ca(OH)_2$ solutions with a 0.5% NaCl.

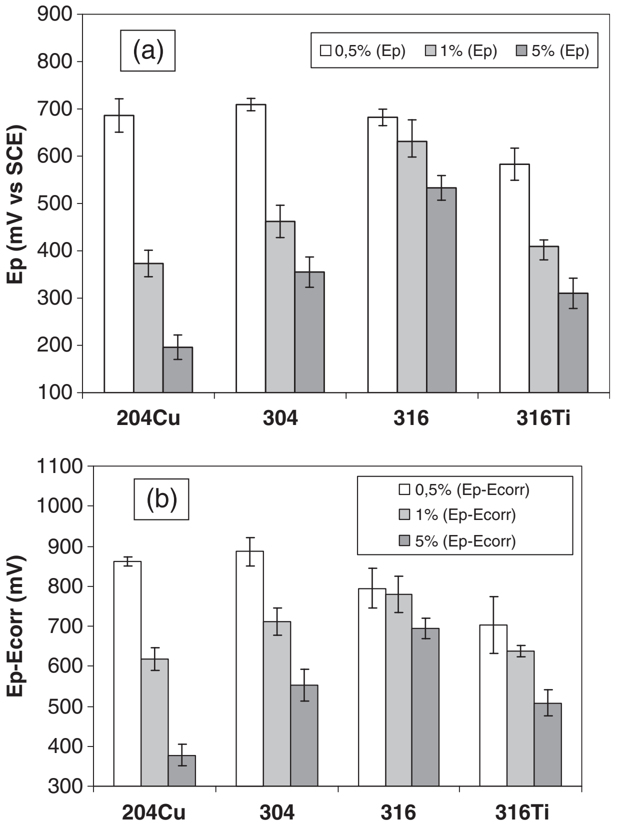

Fig. 3. Results obtained from the anodic polarization curves for corrugated stainless steels in saturated, non-carbonated $Ca(OH)_2$ solutions with different chloride concentrations: (a) pitting potentials and (b) distance between the pitting potential and the corrosion potential.

blamed for the increase of the susceptibility to the corrosive attack. Mechanical steel couplers tend to be used to avoid the problems caused by welding.

Duplex stainless steels reinforcements are coming into use. Recent works have demonstrated that their corrosion resistance in these media is similar to that of the traditional austenitic types [18] or even better [10,20,21], depending on the composition of the studied duplex steel. However, ferritic types (the cheapest traditional stainless steels) have been shown to be susceptible to pitting corrosion in severe marine environments [10,18,22].

The price of stainless steels makes the use of this material suitable only to reinforce the most critical areas of structures (such as the tidal areas in partly submerged structures or the outer reinforcements in bridges). It is estimated that the use of a 5–10% of stainless steel reinforcements (in volume) is enough to guarantee the durability of structures in highly aggressive environments [23], but even this limited use of stainless steel can increase the initial cost of the structure by 5–15% [10]. There are studies that prove that the use of stainless steel reinforcements in highly aggressive environments is cheaper in the long term than cathodic protection [14,24], but the high initial cost often limits their use.

Nickel is one of the alloying elements that increases significantly the cost of such austenitic and duplex stainless steels; above that of other majority alloying elements such as chromium. Its cost has risen a 60% in the last 4 years, reaching 13 000 €/t in 2005.

High-manganese, low-nickel austenitic stainless steels have proved to have mechanical properties comparable to those of traditional austenitic steels. Their corrosion resistance in acid and in neutral solutions with chlorides is always better than ferritic stainless steel types and sometimes they are very close to 304 austenitic type [25,26]. This work studies the corrosion behaviour of a low-nickel stainless steel, type 204Cu, in media that simulate the solution contained in the pores of the concrete. The use of 204Cu could mean a saving of about 35–40% in the price of the stainless steel reinforcements as compared to type 304.

2. Materials and methods

Cold-worked corrugated bars of austenitic stainless steel of different compositions (204Cu, 304, 304L, 316, 316L and 316Ti) were used in this study. The chemical composition of the stainless steels is shown in Table 1. The diameter and mechanical properties of the reinforcements are shown in Table 2.

The corrosion behaviour of the materials was studied in $Ca(OH)_2$ saturated solutions. Fresh, non-carbonated solutions were used to simulate non-carbonated concrete, and solutions in which the pH had been reduced to about 9 by bubbling CO_2-enriched air were used to simulate carbonated concrete. In both types of solution, different amounts of NaCl were added (in quantities that range from 0% to 5% (w/w)) to test the effect of chloride contamination.

The susceptibility to pitting corrosion of the corrugated stainless steels was characterized by cyclic polarization curves. The procedure is based on the standard ASTM

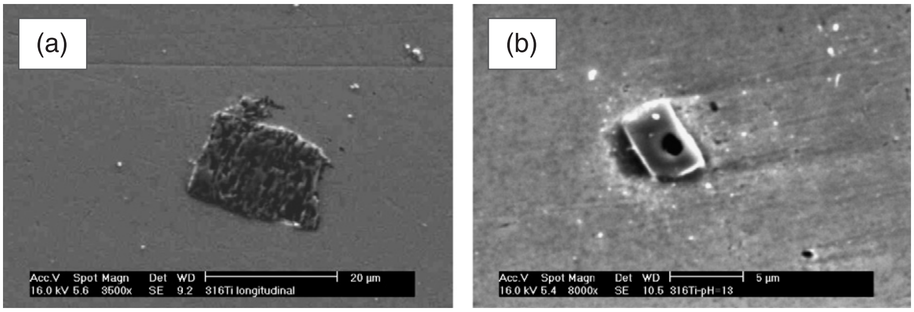

Fig. 4. Morphology of the TiN precipitates in the 316Ti corrugated steel: (a) before any corrosion test; (b) after a potentiostatic experiment.

G61 test, modified for the study of reinforcements in simulated pore solutions. The electrochemical measurements were carried out after 20 h of exposure of the reinforcement to the test solution, to try to reach a stabilization of the corrosion potential (E_{corr}). Regions of the reinforcements of 2 cm length and with the polished cross section have acted as the working electrode. Saturated calomel electrode (SCE) and graphite bars were used as reference electrode and counter-electrodes, respectively. The sweeping rate of the potential was 0.17 mV/s. Pitting potential (E_{p}) was determined as the potential value at which the current sharply rises when the working electrode are anodically polarized. The current limit for reversing the sign of the potential sweep was 10^{-4} A/cm^2. It has been considered necessary to repeat each test 4–6 times to assure reliable results, due to the scattering on the corrosion results caused by the heterogeneities on the surface of the corrugated materials. The reliability of the parameters obtained from the curves is calculated using the standard deviations of the data.

Potentiostatic tests in non-carbonated, saturated $Ca(OH)_2$ solution with 1% NaCl were carried out occasionally, to study the morphology of the attack. In this test, the cell configuration was the same as that for the polarization curves. Potentials slightly above the pitting potential of the working electrode in this medium were imposed on the stainless steel for 30 min.

The presence of precipitates in the materials and the morphology of the attack after the corrosion test were studied by optical microscopy and by scanning electronic microscopy (SEM).

Galvanic currents have been measured between pure TiN pieces and 316 type stainless steel to study possible galvanic effects in 316Ti reinforcements. For these measurements, identical areas of both materials (7.07 cm^2) have been exposed for 45 min to a non-carbonated, saturated $Ca(OH)_2$ solution with 1% of NaCl.

3. Results and discussion

The results obtained in all the testing media proved that low-carbon stainless steels (304L and 316L) exhibit identical corrosion behaviour to that of stainless steel reinforcements of similar composition with higher carbon content (304 and 316). The different deformation levels

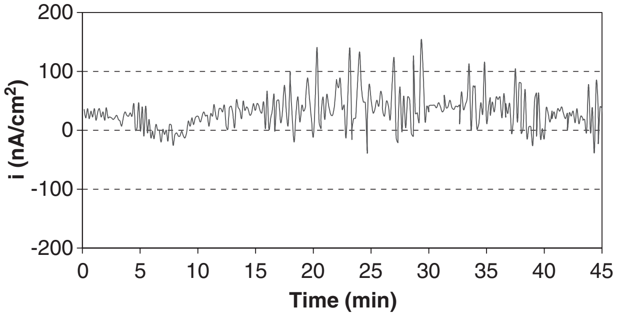

Fig. 5. Galvanic current measured between pure TiN and 316 type stainless steels in non-carbonated, saturated $Ca(OH)_2$ solution with 1% NaCl.

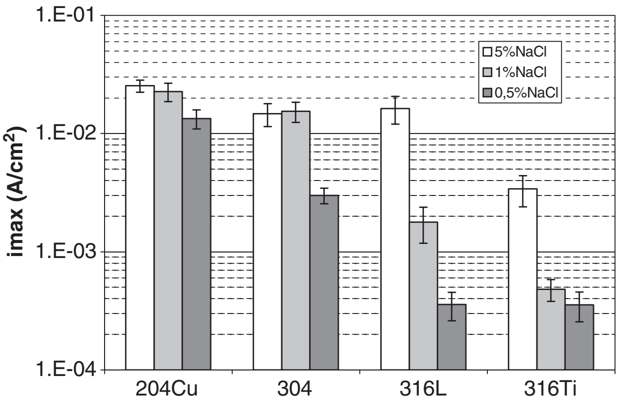

Fig. 6. Maximum current densities registered in the polarization curves after reversing the potential sweep al 10^{-4} A/cm^2.

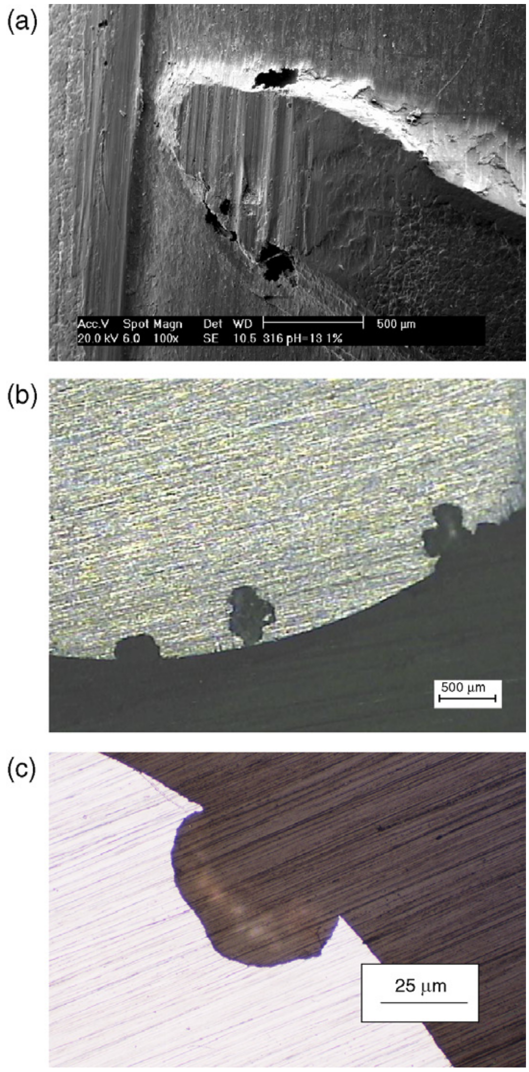

Fig. 7. Morphology of the attack on the different stainless steels after polarization tests in non-carbonated, saturated $Ca(OH)_2$ solutions with chlorides: (a) plain view of pit on 316 on 5% NaCl; (b) cross-sectional view of pits on 304 in 5% NaCl; (c) cross-sectional view of a pit on 204Cu in 0.5% NaCl.

between 304 and 304L types, which strongly affect the mechanical properties of the reinforcements (Table 2), do not modify meaningfully their corrosion resistance. Results of 304 and 316 in all the figures shown in this work are also representative of the corrosion behaviour of 304L and 316L types respectively.

3.1. Non-carbonated media

The influence of the chloride content on the pitting resistance of the corrugated low-nickel 204Cu stainless steel at pH≅12.6 is shown in Fig. 1. In the absence of chlorides, no sign of attack can be detected by this technique. The sudden increase of the current density that can be observed for this material at potentials above 650 mV vs. SCE is caused by the oxidation of hydroxyl ions of the solution:

$$4OH^- \rightarrow O_2 + 2H_2O + 4e^-$$

When NaCl is present in the solution, the onset of the pitting attack can be clearly detected in the curves when sufficiently high anodic polarizations are imposed. As expected, the E_p decreases and comes closer to the E_{corr} when the NaCl concentration in the test solution increases. No repassivation of the pits is observed when the potential sweep is reversed in any of the studied chloride concentrations.

Fig. 2 compares the pitting behaviour of the 204Cu with those of other austenitic stainless steel in the same media (0.5% NaCl). It can be observed that the E_p of the low-nickel steel is very close to the pitting potential of the more traditional steels in saturated $Ca(OH)_2$ solutions with 0.5% NaCl additions. This suggests the very low pitting corrosion susceptibility of the 204Cu steel reinforcement in non-carbonated chloride-contaminated concrete.

The closeness between the pitting corrosion resistance of 204Cu and other austenitic stainless steels at highly alkaline pHs, are corroborated by the results obtained in solutions with high chloride concentrations (Fig. 3a). As expected, the E_p decreases for all the materials as the chloride content in the testing solution increases. Moreover, the increase of the aggressivity of the environment enhances the different pitting corrosion behaviour of the reinforcements due to their chemical composition. In solutions with higher chloride content, the differences in E_p begin to be significant between 204Cu steel and high-Ni austenitic steels. The difference $E_p - E_{corr}$ in the 204Cu type ranges from about 380 to about 860 mV, depending on the chloride concentration of the test solution (Fig. 3b). 204Cu steel shows slightly higher susceptibility to pitting corrosion than other studied stainless steels in high chloride concentration solutions.

It should be borne in mind that there are differences between the development of the corrosion process in simulated pore solutions and those in real reinforced structures. In concrete, the availability of the elements that control the corrosion rate (water and oxygen) is different to that in an aqueous medium. Moreover, the probability of the onset of the attack is also very different. In practice, the typical heterogeneities of the concrete/reinforcement interface favour the formation of local corrosion cells with potential differences much higher that those caused by the cells that can appear in a solution test. The

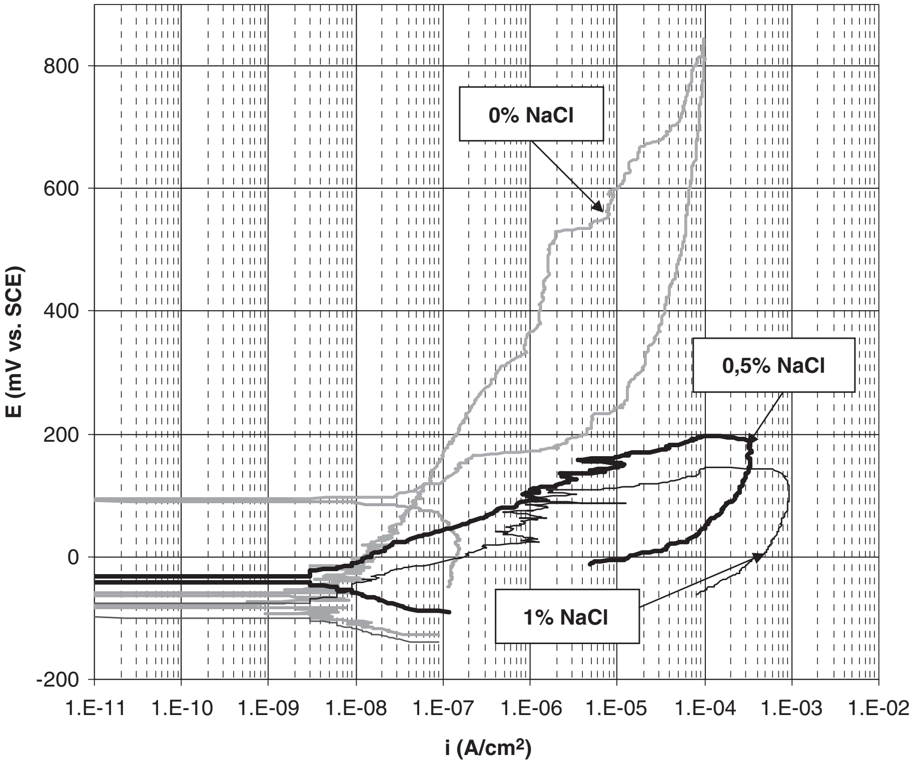

Fig. 8. Influence of the chloride concentration on the polarization curves of corrugated 204Cu in carbonated, saturated $Ca(OH)_2$ solutions.

real force of the corrosion cells in concrete depends on the composition on the concrete and the aggressivity of the environment where the structure is exposed, but the susceptibility to pitting corrosion will always be higher in real conditions than in solution tests. On the other hand, long-term studies of high-Ni austenitic stainless in chloride contaminated concrete have proved the immunity of these material to corrosion [10–12,18,27]. As E_p and $E_p - E_{corr}$ differences for 204Cu steel are not much lower than these measured in the 304 and 316 types, this seems to indicate that this corrugated steel could be able to withstand the potential differences existing in concrete in most of the existing marine environments and it could be assumed that the low-Ni austenitic stainless steel would have similar behaviour to the other more traditional types. Only in extremely aggressive conditions, as those that suffer some structures exposed in Persian Gulf, these reinforcements could possibly present corrosion problems. The much lower price of 204Cu stainless steel than that of Ni-rich stainless steel could stimulate the use of these stainless steel reinforcements and prevent many corrosion problems. Before drawing definitive conclusions it is advisable to check these results with long-term test carried out in mortar.

Fig. 3 also shows the beneficial effect of Mo, especially in more aggressive solutions: 316 and 316L steels present a less pronounced decrease on E_p value when chloride content increases. The intrinsic instability of E_{corr} in corrugated materials makes $E_p - E_{corr}$ difference results a less reproducible parameter than E_p for comparative evaluation of the corrosion performance of different stainless steels. For example, 316 tested in 0.5% NaCl presents a $E_p - E_{corr}$ distance lower than that expected, mainly due to the fact that most of polarization curves of this material in this environment showed E_{corr} values higher than usual.

Another interesting behaviour that can be deduced from Figs. 2 and 3 is the slightly lower pitting resistance of 316Ti stainless steel as compared to that of other 316 or 316L types. In 316Ti steel, the Ti content favours the formation of numerous TiN precipitates in the microstructure (Fig. 4a). The presence of these precipitates could favour the onset of localized attack. As can be seen in Fig. 4b, image obtained after a potentiostatic test, TiN precipitates tend to dissolve (the surface roughness changes, a hole is formed in the middle of the precipitate, and a crevice appears between the precipitate and the stainless steel matrix). The galvanic current measured between a 316 stainless steel and a TiN specimen is shown in Fig. 5. The electrodes have been connected in such a way that if TiN were to exhibit cathodic behaviour against the stainless steel, a positive current would have been detected. As the measured current fluctuated slightly around zero, it can be concluded that corrosion of the matrix is not based on a galvanic corrosion phenomenon. The formation of crevices between the particulates and the matrix are due essentially to the chemical dissolution of the TiN. The geometrical heterogeneities that would cause the partial dissolution of the TiN on the surface of 316Ti stainless steel would favour the formation of corrosion cells and the eventual nucleation of pits. This might explain why the pits nucleate at lower potentials on 316Ti than on a conventional 316, in which these precipitates do not exist.

Another beneficial effect of Mo additions can also be deduced from the obtained corrosion results. In all the polarization curves, the sign of the potential sweep was reversed when the current density reached 10^{-4} A/cm^2. However, the current continued to increase for a while, even though decreasing potentials were being imposed (Figs. 1 and 2). This reveals the strong tendency of the attack to continue increasing once the pits have emerged. The maximum current density registered depends on the chloride content of the dissolution and on the composition of the reinforcement (Fig. 6). When the chloride concentration in the media is lower, the growth rate of the pits has a clear tendency to stabilize after reversal of the potential sweep. The influence of the metal base composition on the maximum current density has proved to be very important, more important than the chloride concentration in the range studied. 316L and 316Ti reinforcements showed their ability

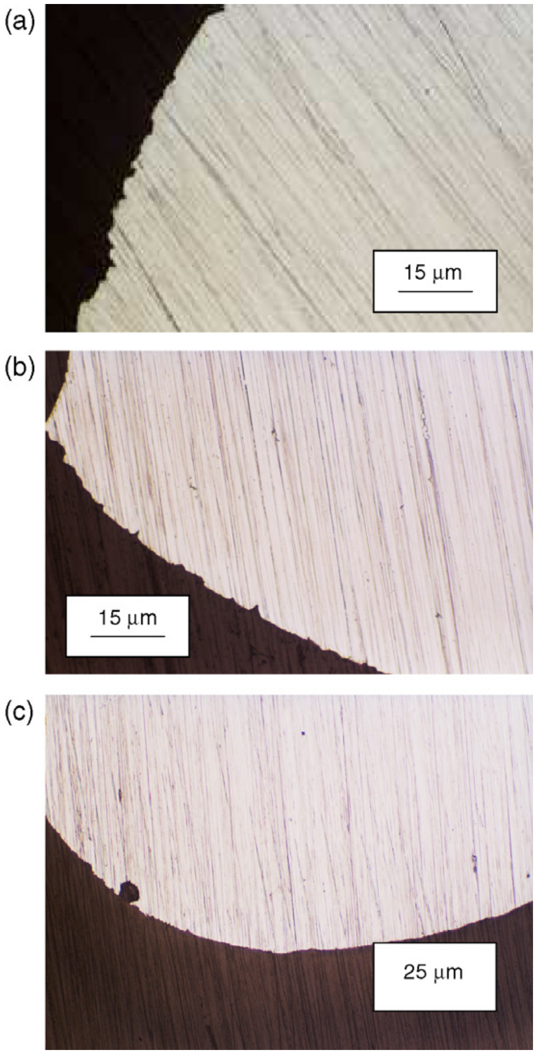

Fig. 9. Morphology of the corrosion on the 204Cu stainless steel after polarization tests in carbonated, saturated $Ca(OH)_2$ solutions with chlorides: (a) generalized attack in medium without chlorides; (b) generalized attack on 204Cu in 0.5% NaCl; (c) pit on 204Cu in 0.5% NaCl.

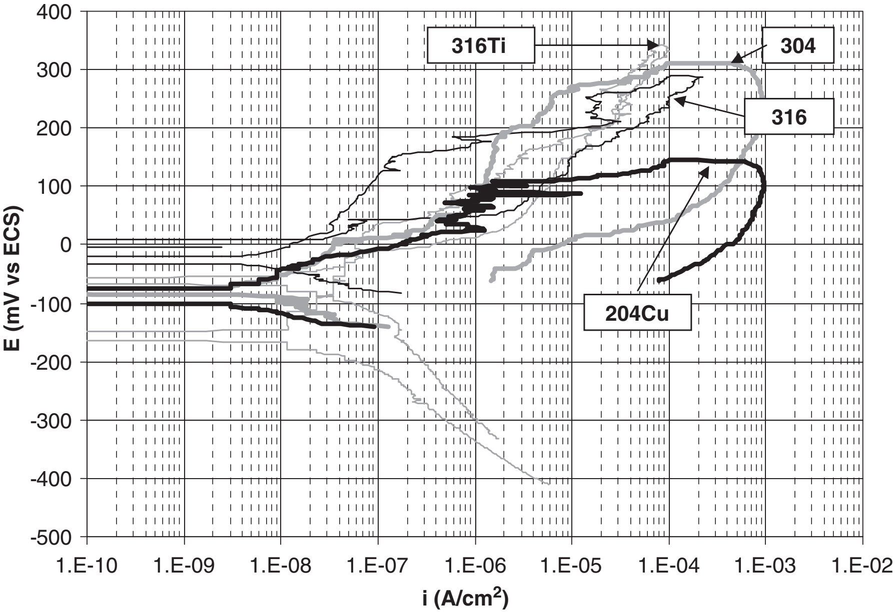

Fig. 10. Influence of the composition of the austenitic stainless steel on the polarization curves obtained in carbonated, saturated $Ca(OH)_2$ solutions with 1% NaCl.

to stabilize the corrosive attack better than 304, probably due to the presence of Mo in the former stainless steels. 204Cu has maximum current densities only slightly higher than those of 304 in the same media. The adverse effect caused by the reduced Ni-content of the type 204Cu on the pitting corrosion behaviour is almost balanced by the beneficial effect of other additives such as Cu or N, present in higher concentration than in the 304 type.

The study of the morphology of the pits carried out in the specimens after the polarization tests have allowed us to conclude that big pits are formed in the austenitic stainless steels studied (Fig. 7). Though there are pits all over the surface of the tested reinforcements, they tend to nucleate preferentially on the edges of the corrugation (Fig. 7a). Two reasons can explain this behaviour: geometrical irregularities that favour the formation of corrosion cells, and also corrugations are the more deformed areas and so, the more prone to corrosion. The pits formed always have globular shape, or they exhibit a very open and shallow shape (in relation with the material loss that suppose those big pits), as can be seen in Fig. 7b and c.

3.2. Carbonated media

Fig. 8 shows the polarization curves for 204Cu reinforcement on a carbonated, saturated $Ca(OH)_2$ solution, depending on the chloride concentration. In the absence of chlorides, 204Cu stainless steel maintains its passivity over a wide range of potentials (more than 600 mV). This passivity range seems to be enough to guarantee the passivity of the reinforcements embedded in carbonated concrete without chlorides. Only at high overpotentials is passivity destroyed and a significant increase of the corrosion rate is detected before the potential of the hydroxyl oxidation reaction is reached. The morphology of attack is very different in carbonated solutions than that of non-carbonated solutions. No pits are macroscopically visible after the test. The increase of the anodic intensity detected from 530 mV vs. SCE is due to a generalized attack (Fig. 9a) that affects great areas of the surface of the reinforcements, especially in the region of the corrugation.

In presence of chlorides, 204Cu steel is not able to keep a stable passivity in carbonated media, and it corrodes with an

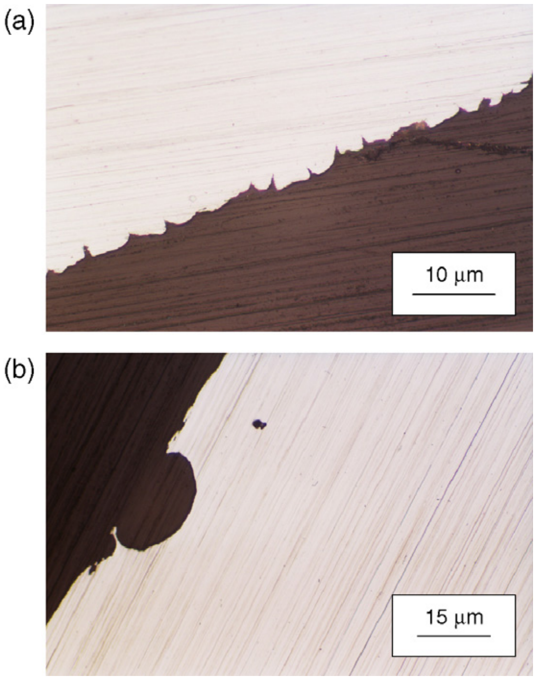

Fig. 11. Morphology of the corrosion on high-Ni austenitic stainless steels after polarization tests in carbonated, saturated $Ca(OH)_2$ solutions with chlorides: (a) generalized attack on 304 in 1% NaCl; (b) pit on 304L in 1% NaCl.

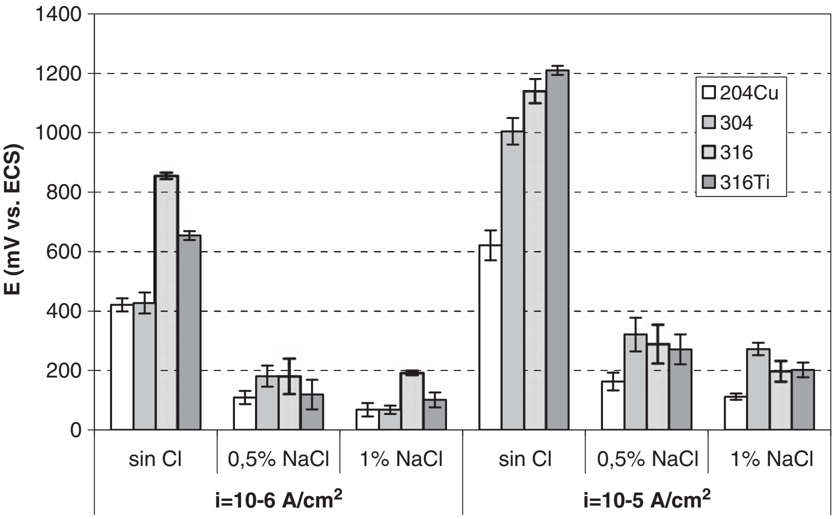

Fig. 12. Potentials at which determinated values of current density are reached for stainless steels in carbonated, saturated $Ca(OH)_2$ solutions with different chloride concentrations.

appreciable intensity when it is submitted to not very high anodic overpotentials (Fig. 8). As occurred in non-chloride carbonated media, the attack proceeds in a generalized way (Fig. 9b), affecting large areas of the surface and being more intense on the corrugation. Occasionally, the presence of chlorides promotes few isolated pits on 204Cu reinforcements (Fig. 9c). These pits are less numerous and smaller than those that appear in non-carbonated environments (Fig. 7).

The high-Ni austenitic reinforcements in carbonated media have a polarization curve with a shape similar to that of 204Cu, but higher overpotentials are required to reach high anodic currents (Fig. 10). It is well known that Mn and Ni promote austenitic microstructures which are more corrosion resistant in chloride media than ferritic microstructures. However, the presence of Ni also has other beneficial effects. It has been shown that the presence of Ni in the stainless steels decreases the thickness of the passive oxide layer and if the Ni content of the alloy is high this element may be found in the external region of the oxide layer and may also increase the corrosion resistance of the stainless steel [28].

Visual observation of numerous cross sections of high-Ni austenitic reinforcements has allowed us to conclude that the corrosive attack also tends to proceed in a generalized way at pH≅9 (Fig. 11a). As occurred with 204Cu, generalized attack tends to centre on areas with greater mechanical deformation. The number of areas, which, apparently, had not been attacked, was greater in high nickel steels than in 204Cu. 316, 316L and 316Ti types demonstrated to have more corrosion free zones than other tested stainless steels. This agrees with the greater ability to control the attack once it has started (in Fig. 10 it can be seen that curves for 316 and 316Ti come back almost on the same path once we reverse the potential of the anodic sweep, while 304 and 204Cu tend to describe a loop with higher intensities during the reversal sweep). Moreover, as occurred with 204Cu, the study of the morphology of the attack on high nickel steels has detected small isolated pits after testing the steels in chloride media (Fig. 11b).

In order to evaluate the general corrosion susceptibility of 204Cu steel in carbonated media and compare it with those of other austenitic stainless steels, Fig. 12 shows the potential values needed to reach the determined anodic intensities.

Table 1
Chemical composition of the studied corrugated stainless steels

AISI type	204Cu	304L	304	316L	316	316Ti
C	0.0490	0.0230	0.0630	0.0180	0.0520	0.0290
P	0.0280	0.0310	0.0310	0.0300	0.0400	0.0290
S	0.0020	0.0010	0.0025	0.0020	0.0010	0.0010
Si	0.2340	0.3610	0.3120	0.3650	0.2850	0.4500
Mn	8.2570	1.4480	1.4220	1.4410	1.4800	1.2130
Cr	16.1240	18.2980	18.3300	16.9740	17.0340	16.6820
Ni	1.8890	8.6750	8.1180	11.2090	10.3860	11.2540
Mo	0.0150	0.2750	0.2970	2.0810	2.2060	2.2320
Pb	–	0.0007	0.0007	–	–	
Ti	0.0030	0.0040	0.0040	0.0040	0.0040	0.2510
N	0.1308	0.0507	0.0447	0.0476	0.0406	0.0205
Cu	2.6510	0.4870	0.3160	0.4970	0.3680	0.4130
Sn	0.0110	0.0130	0.0170	0.0120	0.0110	0.0110
Co	0.0040	0.1900	0.1280	0.1310	0.1360	0.1070
B	0.0006	0.0005	0.0002	0.0052	0.0034	0.0012
Nb	0.0080	0.0140	0.0090	0.0100	0.0150	0.0060
Al	0.0050	0.0070	0.0080	0.0001	0.0001	0.0880
V	0.1070	0.0920	0.0910	0.0870	0.0980	0.0880
As	0.0020	0.0100	0.0070	–	–	

Table 2
Physical and mechanical properties (tensile strength, yield strength at 0.2% and elongation 5XD) of the studied corrugated stainless steels

AISI type	204Cu	304L	304	316L	316	316Ti
∅ (mm)	5	10	8	6	6	12
Tensile strength (MPa)	918	747	1035	897	874	860
Yield strength (MPa)	765	580	923	710	693	726
Elongation (%)	32	38	21	27	27	22

Previous research [29] have demonstrated that corrosion intensities higher than 10^{-6} A/cm^2 should be considered highly dangerous, because they would consume a structure's service life in less than 10 years. In the figure, it can be seen that the strength of the corrosion cells necessary to reach this current intensity is quite similar for 204Cu as for other tested steels. Only Mo-types seem to need higher polarizations to reach a corrosion rate of 1 μA/cm^2, especially in media without chlorides. It is necessary to consider very high corrosion intensities, higher than 10^{-5} A/cm^2, to detect big differences between 204Cu reinforcement and other austenitic types. In very aggressive media (carbonated and highly chloride contaminated) it is possible that the behaviour of 204Cu would not be as good as, for example, the 304 type. Further long-term tests with carbonated mortar or concrete specimens in chloride environments should be undertaken before recommending this type of low-Ni reinforcements for such applications.

4. Conclusions

The most important conclusions to be drawn from the results presented in this work are:

- The low-Ni type 204Cu stainless steels have corrosion behaviour very similar to that of traditional austenitic stainless steels in chloride contaminated, non-carbonated $Ca(OH)_2$ solutions. If further experimental results obtained in mortar or concrete confirm these data, the use of this type instead of the 304 type for structures exposed in these conditions would mean a reduction of about 35–40% of the cost of the stainless steel reinforcements.
- The low-Ni type 204Cu stainless steel also maintains a wide passivity range in carbonated solutions without chlorides. These results suggest a good durability in carbonated concrete.
- The corrosion resistance of low-Ni type 204Cu is only meaningfully lower than that of more traditional austenitic stainless steels in highly aggressive solutions, and further long-term tests in carbonated mortar or concrete would be necessary before recommending this type of reinforcement for structures to be exposed in highly contaminated environments.
- The corrosion behaviour of 316Ti stainless steel reinforcements is influenced by the presence of TiN precipitates that decrease its corrosion resistance.

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