

## Valorization of Cork Waste to Improve the Anti-Corrosion Properties of Concrete Reinforcements

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Corrosion of steel reinforcement is the most important mode of concrete structures damages. It strongly depends on the composition and physicochemical properties of the cementitious medium. The use of waste materials as lightweight aggregates in concrete is environmentally recommended in polluted environments such as marine and/or industrial atmospheres in order to reduce its porosity and ensure the requested protection of reinforcing steel. The present study investigated the effect of waste cork addition on corrosion resistance of steel rebar in mortar specimen prepared in the laboratory. The main objective of this study was to improve the corrosion resistance of reinforcing steel. Another objective of this study was to valorize this ecological product and preserve the environment. Results obtained from various electrochemical tests indicated that the presence of a fine cork powder substantially improved the corrosion resistance of steel in the mortar contaminated by chloride ions. This improvement was reflected by a notable decrease in corrosion current density and a shift of corrosion potential of the steel towards more noble values. Moreover, the presence of a fine cork powder in the mortar had no adverse effect on its mechanical properties.

**Keywords:** Cork, Corrosion, Steel, Concrete, Chlorides

### 1. Introduction

In sound concrete, reinforcing steel is naturally protected against corrosion thanks to the high alkalinity offered by the concrete pore solution ( $\text{pH} > 12$ ). However, because of chlorides ions penetration and/or atmospheric  $\text{CO}_2$  diffusion, the thermodynamic conditions providing this chemical protection are modified, which leads to an active corrosion of steel rebar [1-4]. As a result, corrosion decreases the cross-sectional area of steel bars, deteriorates the mechanical properties, induces the cracking or the spalling of concrete cover and alters the bond performance between steel and concrete [5-8]. Therefore, corrosion is one of the most predominant deterioration in reinforcement concrete (RC) structures, it's considered to be one of the worst and most frequent pathologies of civil building engineering and, as well as being a technical-economical-social problem, it still represents a great waste of natural resources [9,10].

Besides the chemical protection offered by concrete pore solution, concrete cover provides a physical protection for rebar steel. However, this protection is intimately related to the cementitious medium proprieties, as well as, permeability, compactness, absorption, etc. Permeability is one of the most important proprieties, because aggressive ions, such as chlorides ions, diffuse and reach easily the rebar steel surface in porous concrete which causes a premature degradation of reinforced structures. Thus, in practice, we interest to reduce the concrete permeability as much as possible, that means obtaining a dense and less porous concrete which ensure, not only a requested compressive strength, but also prevent the diffusion and penetration mechanism of aggressive agents from the external environment. The concrete mixture is the most practical way to minimize steel reinforcement corrosion in contaminated concrete [11-15]. The use of waste materials as lightweight aggregates in concrete is highly recommended in seismic risk areas and environmentally recommended. Thus, we often try to modify the concrete composition by adding

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different kinds of adjuvants (synthetic or natural) under different forms (solid, aqueous, etc.). Different agro-concretes containing flax shives [16], hemp shives [17], sunflower [18], miscanthus [19], rice and wheat husk [20], date palm [21], straw [22], diss and doum [23] and corn stalk [24] were developed in order to improve the quality concrete.

Cork is known worldwide as a sealant in bottles, but it is also a material suited for various demands of the construction sector: its combined characteristics of lightness, elasticity and resilience, impermeability, insulation, wear-resistance, fire retardant qualities, hypoallergenic properties and durability differentiate it from wood or stone [25,26]. The increasing market pressure towards natural and sustainable materials contributes to the natural appealing of cork. The use of this ecological product in the building sector has become an increasingly interesting alternative for enhancement of concrete properties, namely, lightness, elasticity, good thermal and acoustical insulation capacity and positive carbon balance [27-32]. Recently, Borges *et al.* [33] studied the properties of cement-based renders containing different content of fly ash and expanded cork or/and clay. Authors highlighted that increase of cork amount engendered a drop in both thermal conductivity and mechanical properties of the studied mortars, qualifying them as thermal mortars. Novais *et al.* [34] attempted to produce inorganic polymer composites by incorporation of expanded cork as lightweight aggregates. By incorporating virgin cork aggregates, Tedjditi *et al.* [35] developed a novel lightweight concrete. Liu *et al.* [36] studied the influence of temperature and moisture on the thermo-physical properties of mortars incorporating several cork percentages. Increasing the temperature from 20°C to 70°C increased thermal conductivity and specific heat by 7.5% and 27.3%, respectively, while, raising relative humidity from 0% to 100%, increased these latter by 60.7% and 26.7%, respectively.

The present study is a contribution in the innovative researches on use of cork waste in cementitious materials, in particular cork powder, as replacement for fines of sand in mixtures. Thought this experimental program we studied the effect of waste cork addition on the corrosion resistance of steel rebar in mortar specimen

prepared in laboratory. The main objective is to improve the anti-corrosion properties of concrete, valorize one of the most used wastes for the treatment of water contaminated by heavy metals and therefore preserve the environment. To achieve our goal, electrochemical tests were carried out on steel rebar in mortar modified by adding of waste cork powder with different grain sizes and different concentration. Parallel to this electrochemical characterization, mechanical tests were carried out to evaluate the cork effect on the compressive strength of mortar specimens.

## 2. Experimental

### 2.1 Material and sampling

In this work the tested material was a low-carbon steel. Samples were cut from deformed steel bar, 6 mm in diameter, their top and bottom ends were masked with adhesive tape and epoxy resin giving an exposed (active) lateral surface of 3.76 cm<sup>2</sup>, Fig. 1. All of the samples were mechanically polished using a series of silicon carbide emery paper.

### 2.2 Electrolytic solution

The electrolytic medium used in this study is a mortar (sand + cement + mixing water). NaCl concentration of 3 % was added in the mixing water in order to simulate the internal environment of a contaminated mortar.

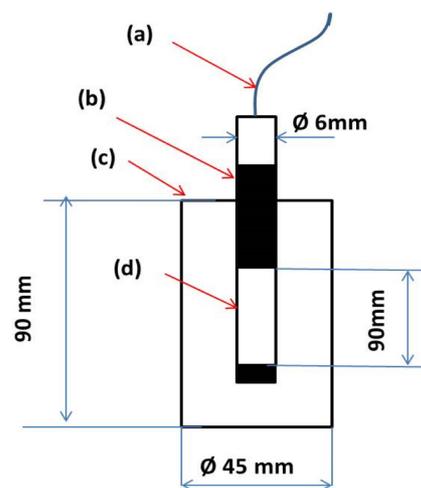
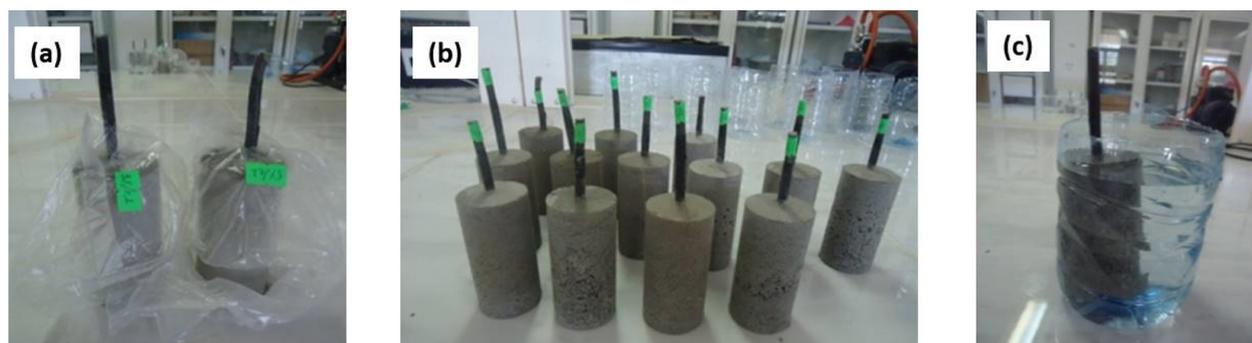


Fig. 1. Shape and size specimens: (a) electrical wire, (b) steel rebar, (c) mortar, (d) active surface of steel rebar



**Fig. 2. Preparation of mortar specimens: (a) PVC molds after pouring the mortar, (b) mortar specimens after demolding, (c) curing of mortar specimens (14 days)**

**Table 1. Different concentrations and granulometry of cork powder**

Cork concentration (%)	1		3		5	
Cork granulometry (mm)	0.08	0.2	0.08	0.2	0.08	0.2

### 2.2.1. Mortar mixture, and curing procedure

The mortar was mixed manually using the following dosage: 223.5 g sand, 74.5 g cement, 37.3 mL distilled water. After mixing, the mortar paste was poured into PVC molds 45 mm in diameter and 90 mm in height. The steel bars, previously degreased with acetone, were embedded in the center of the molds. Once the pouring and compaction of the mortar were achieved, all of the specimens were covered by plastic covers to prevent evaporation of the mixing water, Fig. 2a.

After 48 hours the specimens were demolded and immersed completely in distilled water for a cure period of 14 days, Fig. 2b, c.

Before adding to the mortar, the cork powder has been sifted, to obtain the desired particle size, washed several times (6 to 8 times) with distilled water and dried in the oven. Once prepared, the cork was added at the time of mixing mortar. The sand is partially replaced by cork powder with two different particles size. The used concentrations and particles size are shown in Table 1.

### 2.3 Electrochemical tests

The evaluation of steel corrosion behavior in mortar, with and without waste cork addition, was carried out by means of various electrochemical techniques: free potential measurement of steel embedded in mortar using a saturated calomel electrode (SCE) and a high impedance millivoltmeter, Fig. 2a.

Potentiodynamic measurements carried out using a Radio-Meter Analytical potentiostat (PGP201 model) and a cell measuring system with three electrodes: saturated calomel electrode (SCE) as reference electrode, platinum wire as counter electrode and reinforcement steel as working electrode. Before each test, we measured the free potential after a hold time of 30 minutes; then the polarization curves were plotted in a potential range of  $-800$  to  $+800$  mV/SCE with a scan rate of  $0.25$  mV.s<sup>-1</sup>.

Accelerated corrosion test by application of a constant potential on an electrolytic cell composed of rebar steel embedded in the mortar (anode) a pre-cleaned and degreased steel bar (cathode) immersed in chlorinated distilled water, Fig. 3b. (schematic diagram of corrosion cell added)

### 2.4 Mechanical tests

Parallel to the electrochemical analysis, mechanical tests were conducted on cylindrical specimens, with 50 mm in the diameter and 100 mm in the height, in order to evaluate the effect of cork addition (rate and particle size) on the compressive strength of the hardened mortar. This mechanical testing is considered as the most popular test performed on concrete in construction as it gives a general idea on the all the characteristics of concrete. Based on this test, one can either accept or reject a concrete work.

Compressive strength tests were performed using a CONTROLAB model hydraulic press with a crushing speed of  $6$  MPa.s<sup>-1</sup>, Fig. 4.

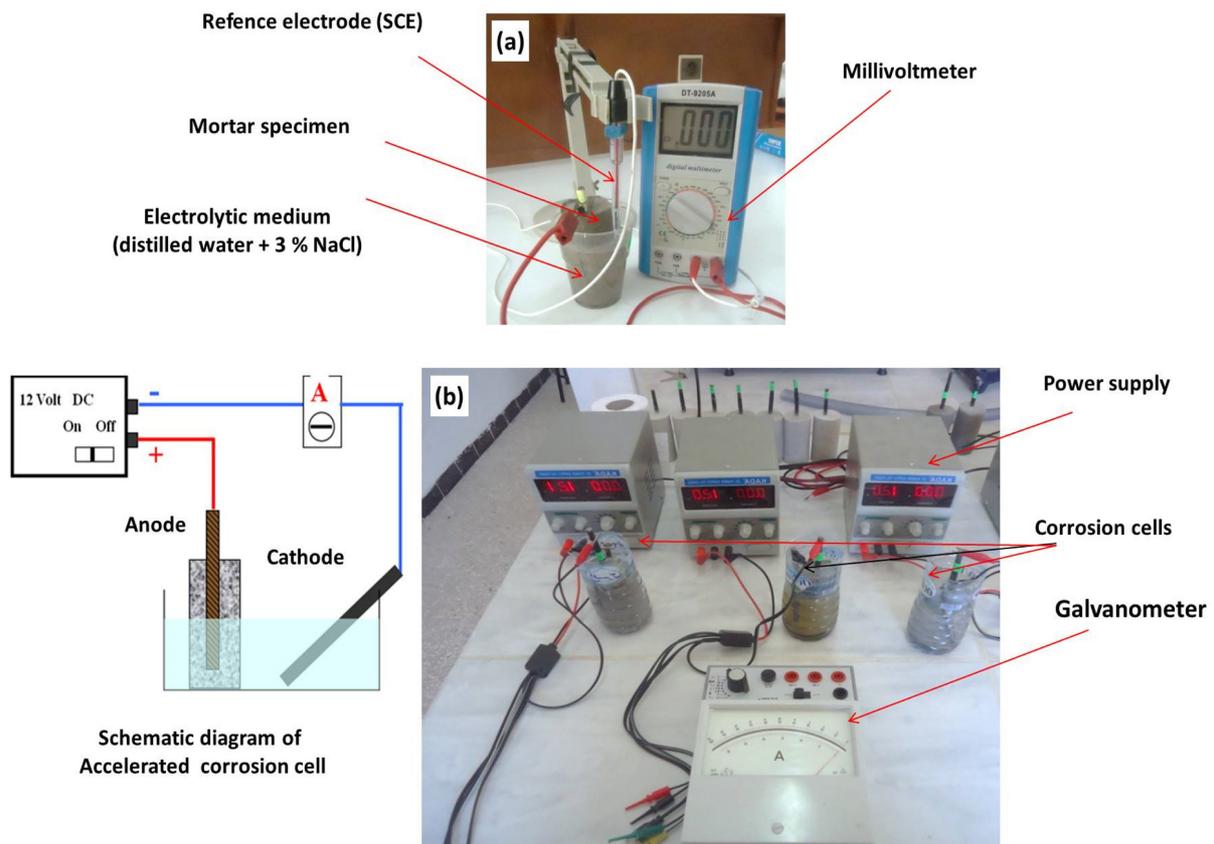


Fig. 3. Corrosion potential measurement and accelerated corrosion test

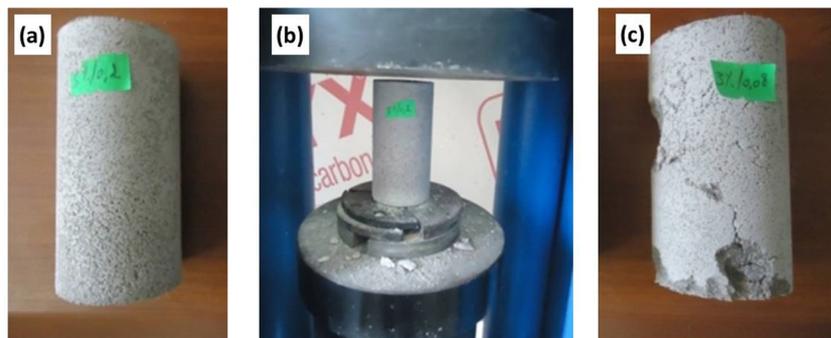


Fig. 4. Specimen condition before and after compressive strength test : (a) Mortar specimen before compressive strength test, (b) specimen under compressive strength device, (c) specimen after compressive strength test

### 3. Results

#### 3.1 Evolution of open circuit potential (OCP)

##### A - Effect of cork concentration:

The effect of the cork concentration on the evolution of open circuit potential of steel rebar in mortar is expressed by the curves of Fig. 5. Two cork granulometry were used: 0.08 mm and 0.2 mm for three different concentrations (1, 3 and 5%).

From the obtained curves we can see clearly the effect of cork concentration on the free potential evolution of steel. Thus, we find that the addition of 1% of cork powder has a positive influence on the free potential for the used particles sizes (0.08 and 0.2 mm). However, the increase in the cork content causes an evolution of the OCP potential in the negative direction which reflecting an electrochemical activity of the steel.

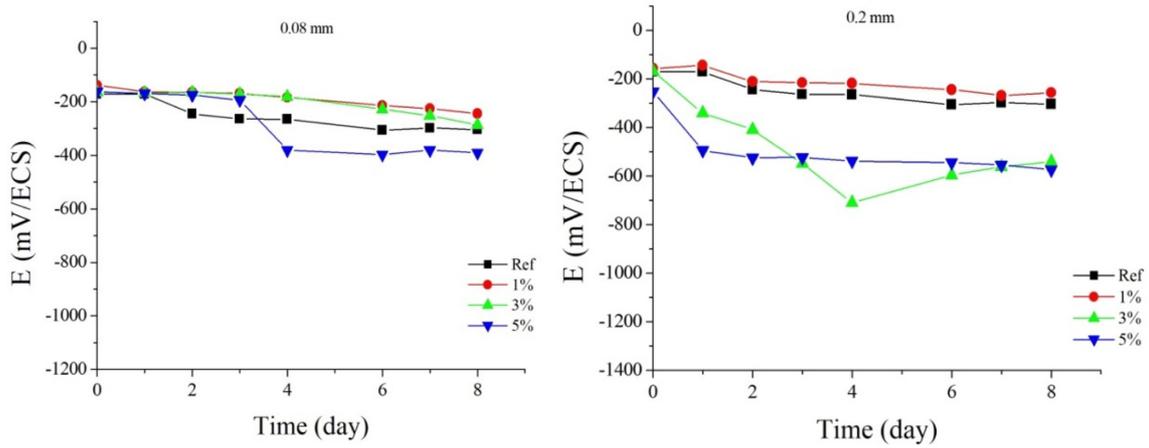


Fig. 5. Effect of cork content on the free potential evolution of steel

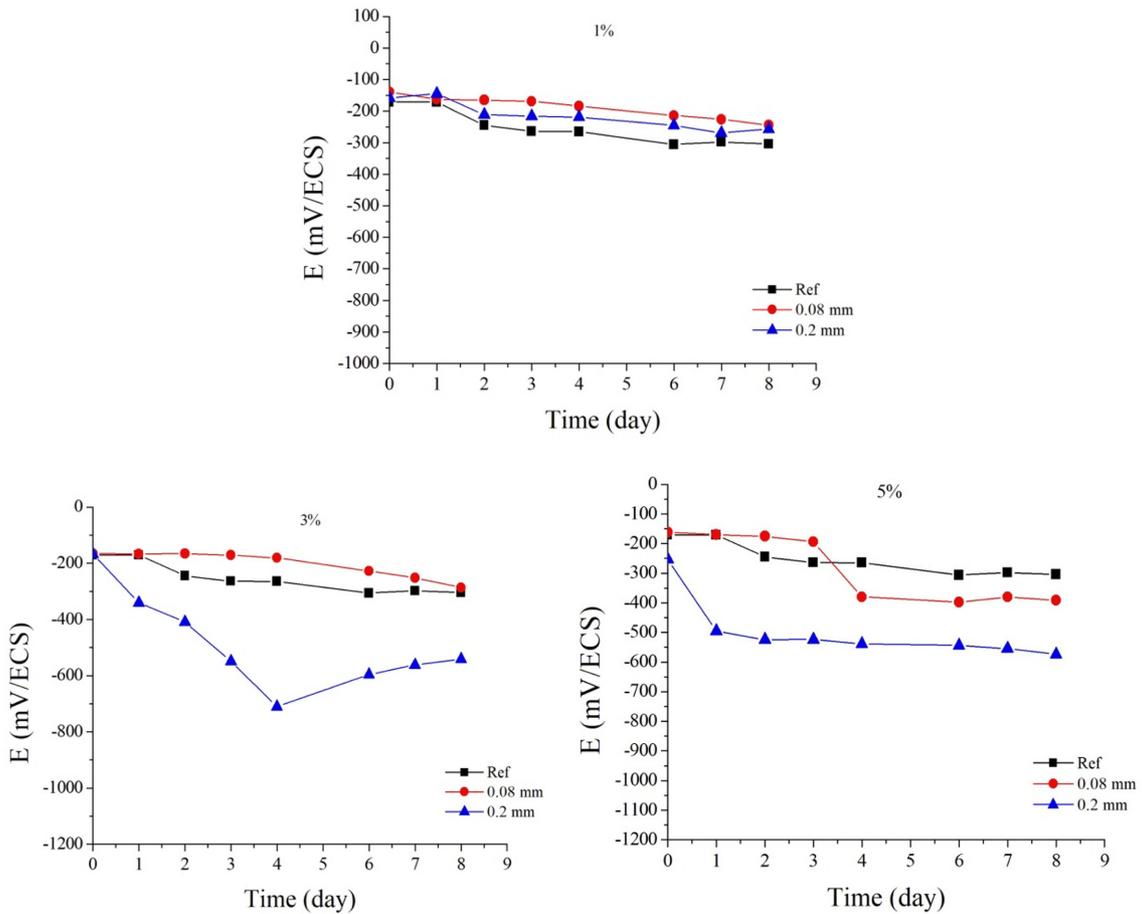


Fig. 6. Effect of cork granulometry on the corrosion potential evolution of steel

*B - Effect of cork granulometry:*

The curves reflecting the effect of particle size of cork on the free potential evolution are shown in Figure 6. As can be seen the granulometry of cork powder has a

remarkable influence on the evolution of rebar steel potential in contaminated mortar specimens. Thus, we find that, for the used concentrations (1%, 3% and 5%), the smaller the grain size of the cork the higher the potential

values of steel. For example, after 8 days of immersion time, the corrosion potential of steel embedded in mortar

with a cork content of 3%, passes from -220 mV/SCE to -580 mV/SCE for the particle size equal to 0.2 mm.

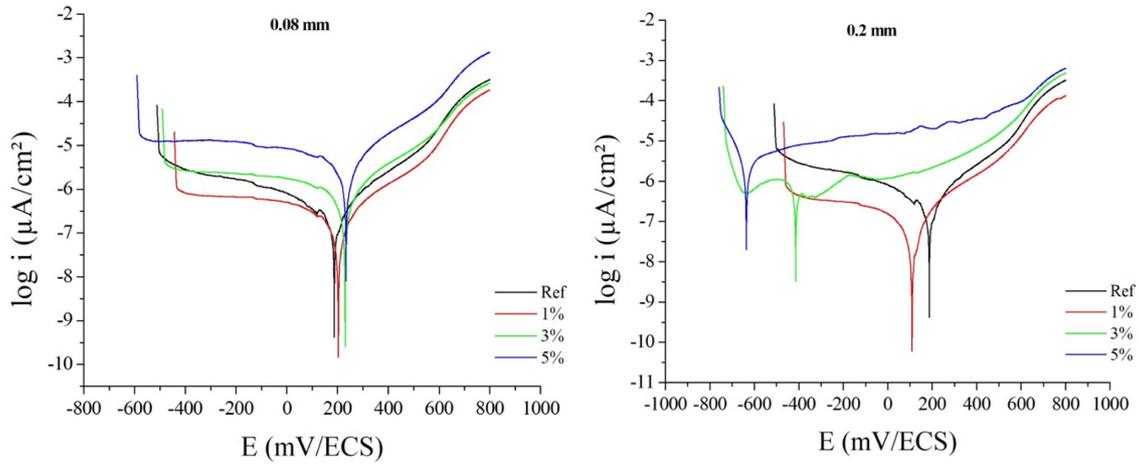


Fig. 7. Effect of cork concentration on the electrochemical behavior of steel

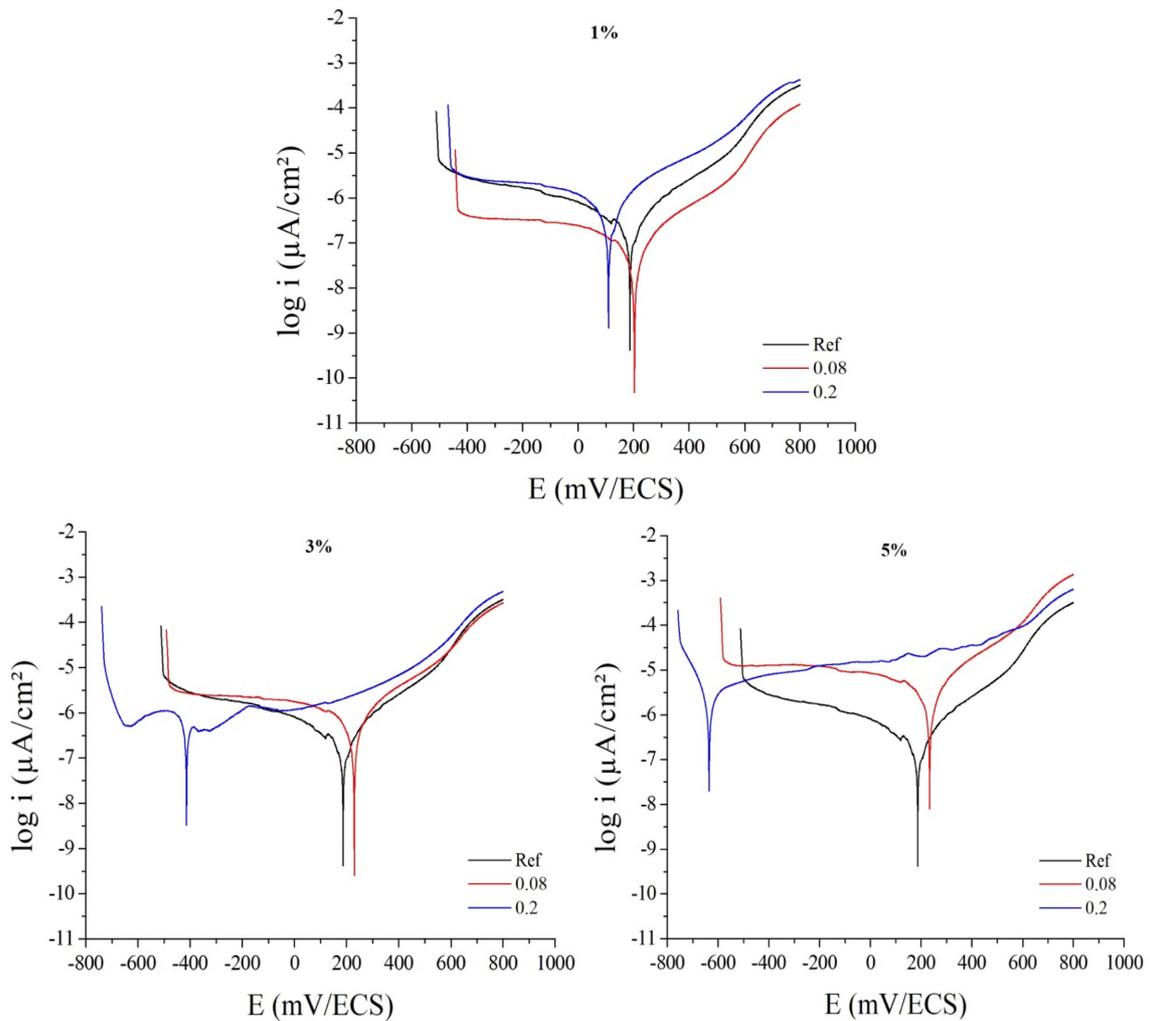


Fig. 8. Effect of cork granulometry on the electrochemical behavior of steel

### 3.2 Potentiodynamic curves

#### A - Cork concentration effect:

The polarization curves reflecting the effect of cork concentration (1%, 3% and 5%) on the corrosion behavior of steel in the mortar are shown in Fig. 7.

The obtained curves show clearly the cork addition effect on the electrochemical behavior of steel in mortar. Thus, we note that for both granulometry (0.08 and 0.2 mm), the cork addition for a concentration greater than or equal to 3% significantly affects the anti-corrosion properties of this porous medium. So, compared to the reference sample (0% cork), steel in the specimens containing 3% and 5% of cork presents a higher current density and a less noble values of corrosion potential, especially for the 0.2mm granulometry. On the other hand, we can notice that, for the two granulometry used, the addition of cork with a concentration of 0.1% does not affect the electrochemical behavior of the steel.

#### B - Cork granulometry effect:

The potentiodynamic curves reflecting the effect of cork particle size (0.08 and 0.2 mm) on the steel corrosion behavior in contaminated mortar are shown in Figure 8. The curves were obtained for three different concentrations (1%, 3% and 5%).

The curves show clearly the effect of cork fineness on the anticorrosion properties of mortar. So, from these results it can be seen that the smaller is the grain size of the cork, the more steel is protected, especially for the 1% and 3% concentrations. In consequence, compared to the reference specimen, the electrochemical data ( $i_{\text{corr}}$  and  $E_{\text{corr}}$ ) evolve in the direction of a clear improvement of corrosion steel resistance.

### 3.4 Accelerated corrosion test

After a test period limited by the beginning of mortar cover damage (cracking and bursting), all of the specimens were broken for a macroscopic analysis. The obtained macrographs are shown in Fig. 9a and b.

According to the obtained macrographs, the surface conditions of steel embedded in the different mortar specimens confirm the results obtained by free potential and potentiodynamic measurements. Thus, we note that the steel embedded in mortar containing the cork with a particle size equal to 0.08 mm, for the different

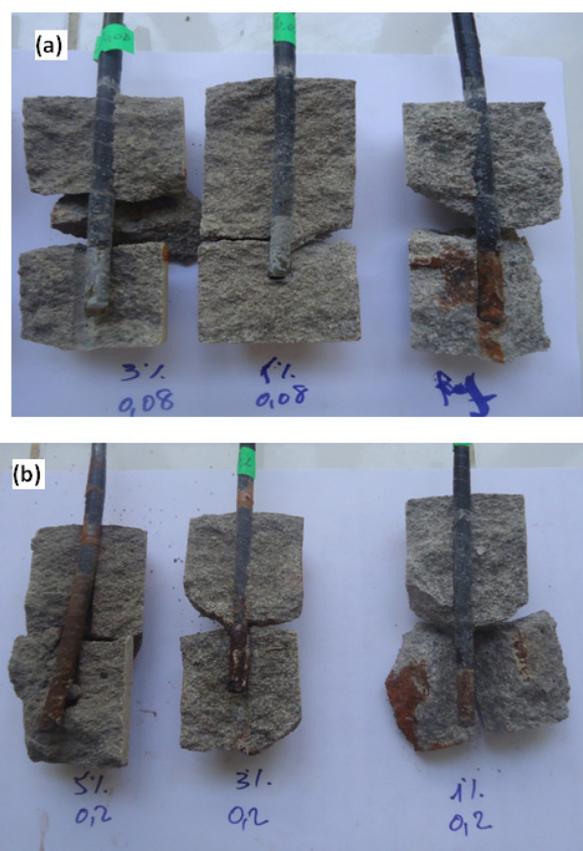


Fig. 9. Macrographs showing the steel surface condition after accelerated corrosion test: (a) granulometry = 0.08 mm, (b) granulometry = 0.2 mm

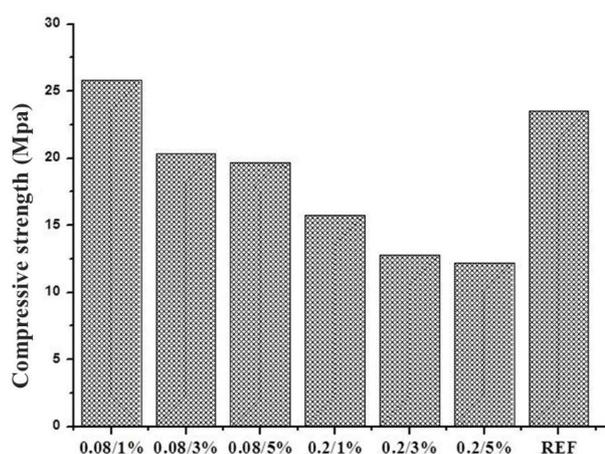


Fig. 10. Effect of granulometry and cork content on the compressive strength of the mortar

concentrations, present an intact surface condition, which reflects the significant improvement of the anticorrosion properties of the modified mortar. On the other hand, the

surface of steel in the mortar specimens containing cork of 0.2 mm is severely attacked.

### 3.5 Compressive strength test

The results obtained by the compressive strength tests, performed on the mortar samples with and without cork addition, are shown by the histograms of Fig. 10. According to these histograms, we find that the addition of the fine cork (grain size equal to 0.08 mm) for a concentration equal to 1%, does not affect the compressive strength of the hardened mortar. On the other hand, the use of a coarser cork significantly affects the compressive strength of the mortar especially for high concentrations (3% and 5%).

## 4. Discussion

According to the results obtained by different electrochemical techniques (measurement of the open circuit potential, potentiodynamic polarization and accelerated corrosion cell) we have seen that the addition of cork powder has a remarkable effect on the anticorrosion properties of the hardened mortar. However, this effect depends both on the grain size and on the rate of added cork. Indeed, the substitution of 1% of the mass of the sand by a fine cork powder (particle size = 0.08 mm) ensures a perfect protection for the reinforcements. This protection, reflected by a decrease in the corrosion current ( $i_{\text{corr}}$ ) and a positive shift in corrosion potential ( $E_{\text{corr}}$ ) of steel as well as an intact surface condition of the samples, is ensured for a cork content around 3% by mass of sand. On the other hand, the addition of a coarser cork powder (particle size = 0.2 mm) does not provide any protection against corrosion of steel rebar especially at high concentrations (5% of the mass of the sand). The electrochemical analysis, in this case, indicates a remarkable electrochemical activity of steel. The surface observations also show an accentuated degradation of the samples.

The protection provided by the cork fines is a physical protection because the presence of this compound causes a modification of the internal structure of the mortar (shape, size and number of pores). Indeed the presence of a sufficient content of fine cork grains reduces the size of the pores and can even cause the complete closure of

the pores with a small size which significantly affects the transport mechanisms within the cementitious medium. The fine particles occupying the pores thus play the role of a physical barrier against the diffusion of aggressive agents, such as chloride ions, towards the metal surface which allows maintaining the thermodynamic conditions necessary for the stability of the passivation layer of the metal offered by the high alkalinity of the interstitial solution of the fresh cement medium. However, it should be noted that the cork used in this work has been washed for several times with distilled water, so it is pure and free from all contamination, which qualifies it as a chemically inert element and does not in any way influence on the chemistry of the interstitial solution. On the other hand, the occupation of the pores by the cork particles promotes the trapping of aggressive ions by absorption phenomenon which reduces their concentration in the interstitial solution.

Regarding the evolution of the mechanical properties, the compression tests show that the presence of cork fines has a significant influence on the compressive strength of the hardened mortar. This influence depends on the fineness of cork as well as its content in the mortar. We have seen that the use of a sufficiently fine powder (particle size = 0.08 mm) causes a slight increase in the resistance of the mortar. This can be explained by the fact that the presence of fine cork substituted the air that occupies the pores of the cementitious medium which caused the improvement of the compressive strength. These results are in agreement with those of Ana Mafalda *et al.* [37], they affirm that it is possible to use abundant waste from cork industry, cork powder, as part of the fine material of self-consolidating concrete, achieving a good level of strength and appropriate durability for common applications. However, the use of a coarse cork powder at relatively high rates causes a remarkable drop in the compressive strength; in particular for cork contents of the order of 5%, this is mainly due to the difference between the resistance of sand and that of cork. The substitution of a significant amount of sand by cork causes a reduction in the resistance of the mortar. These results are in agreement with the results presented by Branco *et al.* [38] regarding the effect of cork on the mechanical properties of concrete, they state that the use of expanded cork leads to further reductions in terms of strength.

González *et al.* [39] stipulate in turn that the drop in the mechanical resistance of the mortar in the presence of the cork can be attributed to the large quantity of water absorbed by the cork. So it is imperative that the cork used in cementitious environments must be fine and added in small quantities in order to not alter the resistance properties of the latter.

## 5. Conclusion

Using waste in concrete is a contribution to sustainability in construction. In this work, effect of waste cork addition of on the corrosion resistance of steel rebar in mortar specimen was evaluated. The following conclusions can be drawn:

1. The cork addition has a significant effect on the corrosion behaviour of steel rebar in contaminated mortar.
2. The use of a fine cork powder provides a good protection of rebar steel in the contaminated mortar against pitting corrosion induced by chlorides ions attack.
3. The cork concentration added to the mortar has a big importance, it's strongly depending on the particle size of the cork powder.
4. The addition of fine cork, particle size equal to 0.08 mm, does not affect the compressive strength of the mortar, in particular for moderate concentrations (< 3%).
5. The beneficial effect of small particles size of cork powder on the corrosion behaviour of steel embedded in contaminated mortar can be explained by the fact that cork particles occupy the pores of the mortar which decreases the permeability and prevents the Cl<sup>-</sup> ions diffusion towards steel surface.
6. In the presence of cork particles in the pore of hardened mortar chloride ions are trapped by absorption phenomenon which reduces their concentration in the pore solution.

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