

# Alkyd based water-reducible anticorrosive paints and their antifungal potential

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## ABSTRACT

Concerns about ozone layer, contamination, fossils resources and health, has banned or restricted the use of traditionally organic solvents in paints. As a consequence, new resins that employed water as main solvent are being studied. Among these resins, alkyds are the most studied one as they can also be synthesised from natural vegetable sources, making them more acceptable.

The aim of this paper was to study the anticorrosive behaviour of alkyd / polysaccharide paints containing zinc phosphate or zinc aluminum phosphate as main pigment and their performance against fungal growth. Two different PVC (pigment volume concentration) were studied. Painted panels were studied by exposition to salt spray and humidity chambers, where rusting and blistering degrees were evaluated, and by electrochemical tests (corrosion potential measurements, ionic resistances determination, linear polarization curves and polarization resistances determination).

Results showed that these type of paints can be used to protect steel and that the type of pigment and the PVC are important to the protection. Besides, good selection of both also prevents fungi growth in a high humid environment.

## 1. Introduction

Paints are one of the most useful methods to protect metals from corrosion [1–4]. Their main compounds are resin (film forming material), pigments (anticorrosive pigment, prime, fillers) and solvents [5]. Alkyd resins are world-wide used as film forming material due to their ease of application and the good protection they provide to metals exposed to mildly aggressive environments [1,5,6]. Besides, alkyd resins are polyesters prepared by the condensation of polyols, polybasic acids, fatty acids or triglyceride oils [7], natural sources can be used as raw materials [8,9]. However, alkyd resins use white spirit as solvent and, due to the concerns about the ozone layer, contamination, fossils resources and health, organic solvents traditionally used are being banned or restricted. These regulations enhanced the research and development of new water soluble resins (resins that are soluble in water) or water-reducible resins (resins having hydrophilic groups in most or all molecules) [7,10,11]. Modifications of resins to incorporate water as solvent can be done in the manufacturing stage by selecting

water-compatible raw materials [12,13] or incorporating additives such as polysaccharides [14] and silanes [15] during paint preparation.

An important problem in water-borne coatings and paints is that they are prone to microbiological degradation. This degradation is due to the fact that microorganisms are able to use paint components (resin, thickeners) as carbon sources [16–18]. Biodegradation is not only important in top coats. Fungi's hyphae can penetrate the different paint layers, degrading also the inner ones [19]. In order to avoid degradation, biocides must be added also to anticorrosive paints [16].

Water-based-modified alkyd resins had been used to prepare anticorrosive coatings with good results. Dhoke prepared coatings with highly substituted methoxymethyl melamine resin as cross-linking for a water-based-alkyd resin with anticorrosive resistance properties [20]. Zhong *et al.* tested fluorinated acrylic-silicone modified alkyd hybrid resin (FASAR) as anticorrosive coating for steel, obtaining better results when FASAR was cross-linked with trimethylolpropane-tris-( $\beta$ -N-aziridiny) propionate [21].

Phosphates, exchanged zeolites, polyaniline and ferrites are used as

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**Table 1**  
Characteristics of the resins.

Characteristics	Alkyd resin	Polysaccharide resin
Viscosity (cp)	6000 ± 1500	1200 ± 400
Solid content (% by weight)	60 ± 1	44 ± 3
pH	Not determined	7 ± 1
Density (g/cm <sup>3</sup> )	0.92 ± 0.01	1.19 ± 0.05

**Table 2**  
Paints formulations.

Compounds (% by volume)	20ZP	32ZP	20ZAP	32ZAP
Alkyd resin	55.8	46.0	55.8	46.0
Polysaccharide resin	1.1	3.7	1.1	3.7
TiO <sub>2</sub>	4.1	7.0	4.1	7.0
Zinc Phosphate (ZP)	3.3	5.6	—	—
Zinc Aluminium Phosphate (ZAP)	—	—	3.3	5.6
Thickener	1.1	1.1	1.1	1.1
Disperser	0.3	0.3	0.3	0.3
Butyl glycol	5.7	6.3	5.7	6.3
Antiflash rusting	1.2	1.4	1.2	1.4
Co-drier	0.2	0.2	0.2	0.2
Ca-drier	1.2	1.0	1.2	1.2
Antiskinning	0.3	0.3	0.3	0.3
Water	13.4	16.7	14.1	17.5
White spirit	12.5	10.4	11.7	9.4
Total	100.0	100.0	100.0	100.0
Pigment Volume Concentration	20.0	32.0	20.0	32.0

anticorrosive pigments in paints [22–26]. Phosphate-based pigments such as zinc phosphate (ZP) and zinc aluminum phosphate (ZAP) are claimed to protect steel from corrosion by phosphating and shielding the surface [27–29] being ZAP better than ZP in damage areas [30].

The aim of this research work was to study paints formulated with a water reducible mixture of synthetic alkyd and polysaccharide resins and two anticorrosive pigments: zinc phosphate (ZP) [31,32], a traditional one; and zinc aluminum phosphate (ZAP), a third generation phosphate-based pigment [28,33,34]. Paints were formulated with two PVC (Pigment Volume Concentration) and without biocide. Results showed that with a good selection of pigments and PVC in paint formulation anticorrosive and antifungal paint can be reached.

## 2. Materials and methods

### 2.1. Raw material characterization

The selected film forming materials were two mixtures of a medium oil alkyd (Alkopol 457/60, 60:40 resin:white spirit, Diransa, Argentina) and polysaccharide (LPR76 Lorama, 44:66 resin:water, Brazil) resins. Table 1 shows some characteristics of the commercial resins. The resins employed and the resins mixtures were characterized by FTIR spectra. The ratios of alkyd / polysaccharide resins used in the FTIR analysis were those used in the formulated paints, 55.8 / 1.1 and 46.0 / 3.7, by volume (Table 2).

### 2.2. Paint formulation, preparation and application

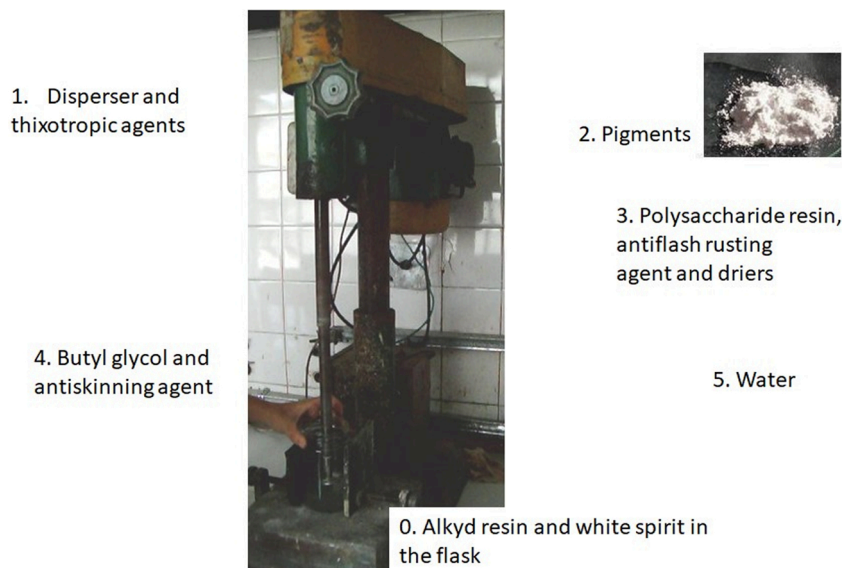
Besides the resins, ZP and ZAP (both provided by SNCZ, France) were used as anticorrosive pigments. The paint formula was completed with titanium dioxide as prime pigment, Co- and Ca-octoates (Casal del Rey, Argentina) as driers, thixotropic (Bentone SD-1, Elementis, Argentina), disperser and humectant (Tego 651, TEGO, Argentina) agents, butylglycol (Química Martín, Argentina) as coalescent, sodium benzoate (Anedra, Argentina) as flash rusting inhibitor and methyl ethyl ketoxime (Casal del Rey, Argentina) as antiskinning additive.

Paints were formulated considered two pigment volume concentrations (PVC) 20 and 32 % being PVC defined as:

$$PVC = \frac{\text{Volume of pigments}}{\text{Volume of pigments} + \text{Volume of resins}} \quad (1)$$

The ratio alkyd / polysaccharide resins was a studied variable but titanium dioxide / anticorrosive pigment ratio was constant and the amount of anticorrosive pigment was 44.5 %, by volume, of the pigment formula. Paints formulations can be seen in Table 2. Taking into account the PVC and the anticorrosive pigment used, the paints were labelled as: 20ZP, 32ZP, 20ZAP and 32ZAP.

Paints were prepared in a high-speed disperser (Corradi, MTA-90 L/4, Vórtex, Argentina) in a 500 mL flask and employing a 3 cm blade. In a first step, the alkyd resin and white spirit were mixed at low speed (700 rpm) for 3 min. Then, the disperser and thixotropic agents were added, at the same speed and dispersed for 3 min. In a third step, the pigments were incorporated at low speed, one by one, and dispersed for 10 min at high speed (1500 rpm) before adding the other one. Afterward the speed was lowered to 700 rpm and the polysaccharide resin, sodium benzoate



**Fig. 1.** Paint preparation.

**Table 3**  
Characteristics of the paints.

Characterization	20ZP	32ZP	20ZAP	32ZAP
Solid content	54.5	59.7	53.6	57.7
Viscosity (UK)	59.0	62.4	62.5	61.6
Grain size (Hegmann units)	5	5	5	5
Hiding power ( $\mu\text{m}$ )	75 + 75 + 75	75 + 75 + 75	75 + 75 + 75	75 + 75 + 75
Set-to-touch time (min)	75	75	75	75
Dry-through time (h)	5	5	5	5

(dissolved in distilled water) and driers were added and dispersed for 5 min. Finally, water, butyl glycol and antiskinning agent were incorporated and dispersed for 5 min (Fig. 1).

The use of alkyd resin together with the polysaccharide resin allowed to diminish the amount of white spirit by half volume [35,36].

The solids content was determined adding, by weight, all the solids (resins, pigments, thixotropic and anti flash rusting agents) in 100 g of paint, Table 3.

The Stormer viscosity of the paints was determined by the Gardner KU-2 viscometer (USA), and adjusted to 58–63 UK in order to apply the paints by spray gun (Mota Premium, P800, Argentina).

The fineness of dispersion was evaluated employing the Hegman gage. This method uses a calibrated tapered groove varying in depth from 100 to zero  $\mu\text{m}$  (zero to 8 Hegman units, respectively). The liquid paint is placed in the deeper end of the channel and the excess is drawn to the shallow end with a scraper. The depth at which continuous groupings of particles or agglomerates protrude through the surface of the wet film is taken as the fineness of dispersion value [37].

The hiding power of the paints was evaluated extending the paint uniformly (employing an applicator with 75  $\mu\text{m}$  gap), onto a combination black-and-white board. If, after drying, the black-and-white squares are still seen, another application is done, until reaching the point of complete hiding. Results are given by the wet thickness of the films applied to hide the black-and-white board [38].

The set-to-touch time was determined lightly touching the film after awhile of application with the tip of a clean finger. Immediately, the fingertip is placed against a piece of clean, clear glass to determine when the paint does not adhere to the finger or transfer to the glass. Is considered dry-through time the time elapsed until the paint is not distorted or detached when the thumb is applied to it and rotated through a 90° angle [39].

SAE 1010 steel panels were sandblasted up to Sa 2 1/2 degree (SIS 05 59 00), degreased with isopropyl alcohol and painted with a spray gun. The final dried thickness was  $85 \pm 7 \mu\text{m}$ , determined by a Schwyz, SC117-02 (Switzerland) thickness gauge. The painted panels were left at  $20 \pm 2 \text{ }^\circ\text{C}$  and 65 % RH conditions for 15 days before testing.

## 2.3. Assays on painted panels

### 2.3.1. Dried adhesion

The dried adhesion of the paints to the substrate was evaluated according to ASTM D 3359 [40]. A pattern with six parallel cuts in each direction was made in the film (up to the substrate), with a cutting steel tool. Afterwards, a tape was applied over the cuts and then removed. Paint adhesion was then evaluated by comparison with illustrations and according to the removed painted area.

### 2.3.2. Chambers accelerated tests

Blistering and rusting degrees (ASTM D714 [41] and ASTM D610 [42], respectively) were evaluated as time elapsed on panels exposed to the salt spray chamber (ASTM B117 [43]). In this chamber, painted panels are exposed to an atomized neutral (pH 6.5–7.2) 5 % by weight sodium chloride solution. Blistering and rusting degrees are evaluated by comparison with illustrations in the ASTM standards.

In some of the panels exposed, a cut through the paint was done by a cutting steel tool in order to expose the metal directly to the salt and the creepage from the scribe was measured with a ruler ( $\pm 0.1 \text{ cm}$ ) and evaluated employing ASTM D1654 standard [44].

Other panels were placed in the humidity chamber (ASTM D2247 [45]), with 100 % relative humidity and 40  $^\circ\text{C}$ , so that condensation forms on the panels. Rusting and blistering degrees were evaluated. As no biocide was added to the paints, fungi growth was observed during exposition. These fungi were observed by stereoscopic microscope (LEICA S8APO, Germany), isolated and identified. This growth is due to the fact that fungi can use as carbon source chemical compounds from the paint and some components encourage microbial growth on the surface. Moreover, some abiotic factors influence this growth, for example, humidity and temperature.

Samples of the fungi were taken by scraping the paints and serial dilutions (1:10) were made using physiological solution [46]. Then 0.1 mL of solution were spreaded in Petri dishes with Agar malt extract (MEA), supplemented with streptomycin (30 mg / 100 mL) to inhibit bacterial growth and Rose Bengal as a fungistatic (5 mg / 100 mL). Petri dishes were incubated at 28  $^\circ\text{C}$  for at least 48 h to proceed with the isolation of the fungi by conventional microbiological techniques. The isolates were identified to the lowest possible taxon using standard taxonomic keys and bibliographic material [47,48]. Characteristics structures of the fungi were observed by Leitz wetzlab microscope (Germany).

### 2.3.3. Electrochemical tests

The cell to carry out the electrochemical tests was design by delimiting 3  $\text{cm}^2$  area (A) employing a cylinder (8 cm high) glued to the painted surfaces. The cylinder was filled with 30 mL of NaCl 0.5 M. Some paint was removed from one end of the painted panels in order to get good metallic connexion.

Conductivity (Ci) measurements were done on these same cells employing a Pt ring and an ATI Orion 170 (USA) conductivity meter with high entrance impedance. The ionic resistance (Ri) was calculated as

$$R_i = C_i^{-1} \cdot A \quad (2)$$

being A, the working painted area.

The polarization resistance (Rp) was determined from polarization curves, employing a Gamry Interface 1000 potentiostat (USA). The sweep amplitude was  $\pm 20 \text{ mV}$  from the corrosion potential and the scan rate 0.5 mV/s. Evaluation of Rp began when Ri values were lower than  $10^6 \Omega \cdot \text{cm}^2$ , when the electrolyte has penetrated the barrier coating [49, 50].

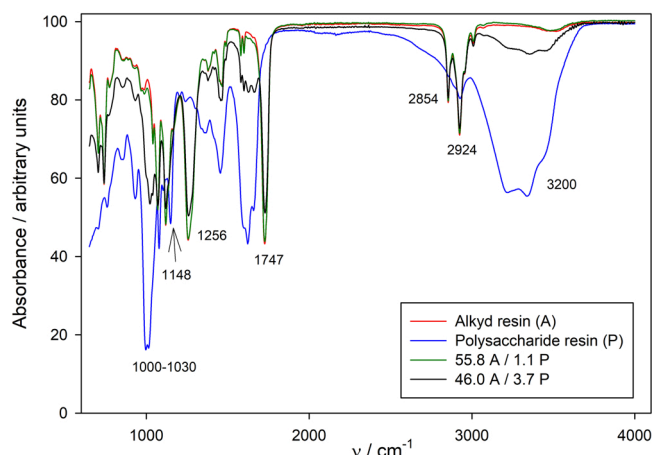


Fig. 2. FTIR spectra of the resins and their mixtures.

**Table 4**  
Corrosion and blistering degrees of the panels exposed to the salt spray chamber.

Sample	18 days		33 days			
	Rusting*	Blistering <sup>§</sup>	Rusting*	Blistering <sup>§</sup>		
20ZP	10	10	10	6F		
32ZP	10	8F	7G	4MD		
20ZAP	10	10	10	8F		
32ZAP	10	10	10	4F		
<b>*Rusting degree</b>	10	9	8	7	6	5
rusted area / %	< 0.01	0.01–0.03	0.03–0.1	0.1–0.3	0.3–1	1–3
<b>§Blistering degree</b>						
Frequency	Dense, D	Medium dense, MD	Medium, M	Few, F		
Size	10	8	6, 4	2		
Comments	No blistering	Smaller size blister easily seen by unaided eye	Progressively larger sizes			

S: spot rusting – the bulk of rusting is concentrated in a few localized areas. G: general rusting – various size spots are randomly distributed across the surface.

**Table 5**  
Creepage of corrosion and blistering from the scribe.

Sample	8 days	18 days
20ZP	8	0
32ZP	7	0
20ZAP	9	7
32ZAP	8	2

### 3. Results and discussion

#### 3.1. Raw material characterization

The FTIR spectra of the resins and their mixtures can be seen in Fig. 2. In the case of the alkyd resin, the main bands of soyabean oil can be seen: 3009, 2924 and 2854  $\text{cm}^{-1}$  due to CH— stretching vibrations; 1747 and 1256  $\text{cm}^{-1}$  due to CO— double and single bonds stretching and at 1659  $\text{cm}^{-1}$  due to CC— double bond stretching [51–53]. In the

case of the polysaccharides resin (P), the main bands appeared at 3200  $\text{cm}^{-1}$  due to OH stretching (broad band), at 2923  $\text{cm}^{-1}$  due to CH stretching. The bands at 1148  $\text{cm}^{-1}$ , assigned to secondary OH groups, and the broad one at 1000–1030  $\text{cm}^{-1}$ , indicate the presence of pyranose ring that is common in polysaccharides [54].

Regarding the mixtures of the resins, alkyd resin (A) bands generally prevail in the spectra. It is difficult to say if the pyranose ring is still present in the mixtures as the characteristics bands of the ring are not clearly seen, due to the low amount of resin used [54]. When the mixture corresponds to 46.0A/3.7 P (with higher amount of P resin) the broad band at 3200  $\text{cm}^{-1}$ , due to OH groups of the polysaccharide, can be seen.

#### 3.2. Assays on painted panels

##### 3.2.1. Dried adhesion

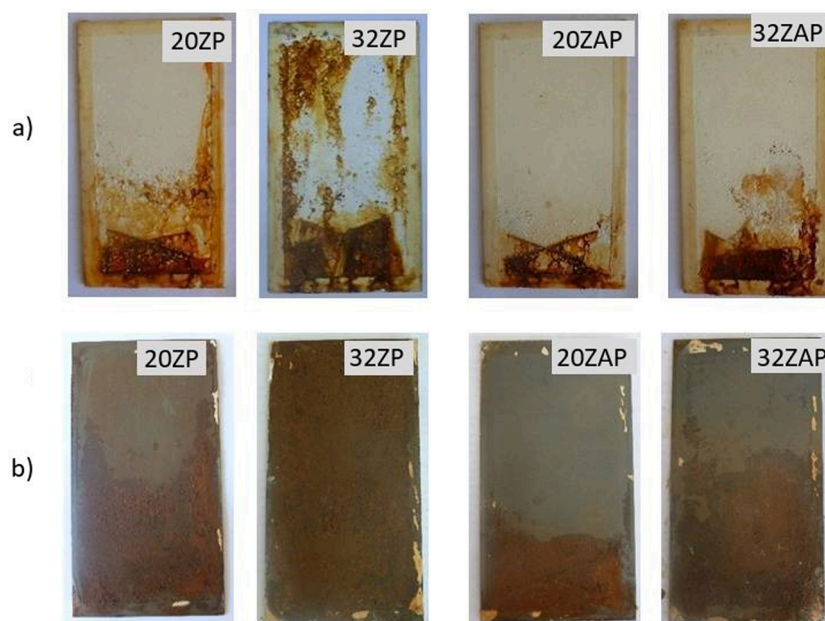
The dried adhesion of the paints to the substrate was qualified as 5B (none of the paint was removed) in the cases of panels 20ZP, 32ZP and 20ZAP. In the other case, less than 3 % of the paint was removed after the test, and the adhesion was qualified as 4B.

##### 3.2.2. Chambers accelerated tests

**3.2.2.1. Salt spray chamber.** Rusting started to be important after 33 days, 32ZP panels exposed to the salt spray chamber. The other panels were qualified with 10. Blisters appeared also earlier (18 days) in the case of panels 32ZP, the size of the blisters was small and the frequency few (Table 4). These results are better than those obtained in some cases with solvent alkyd paints [1,36,55] and comparable to others [56], taking into account the differences in film thickness.

The evaluation of the creepage of the corrosion from the scribe can be seen in Table 5. After 18 days in the chamber, the corrosion crept from the scribe up to more than 20 mm in the case of the panels 20ZP and 32ZP, and 2 mm and 12 mm in the cases of 20ZAP and 32ZAP, respectively. However, the scribe area was very damage as blisters and delamination appeared up to 30–60 mm from the scribe. In Fig. 3a the painted panels with the scribe, exposed to the salt spray chamber for 18 days, can be seen.

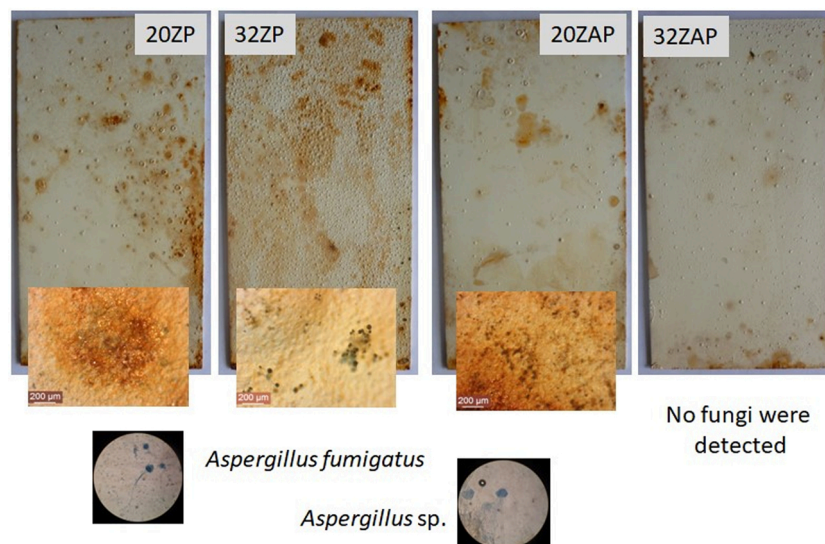
After 18 days, these panels were removed and unpainted by immersion in NaOH 10 % at 60 °C, in order to evaluate the corrosion underneath the paint. The evaluation of these panels, Fig. 3b, showed that



**Fig. 3.** Panels exposed in the salt spray chamber for 18 days: a) painted panels, b) unpainted panels.

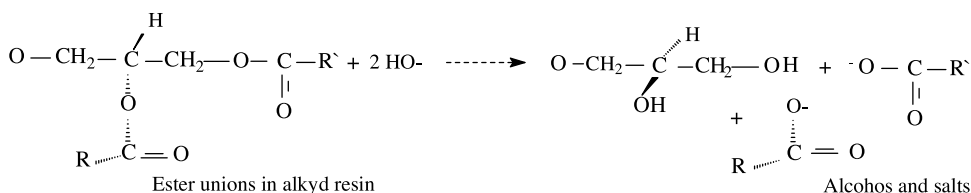
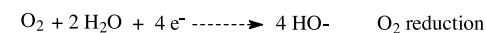
**Table 6**  
Corrosion and blistering degree of the painted panels exposed to the humidity chamber.

Samples	8 days		18 days		33 days	
	Corrosion	Blistering	Corrosion	Blistering	Corrosion	Blistering
20ZP	10	6M	10	4M	5G	2M
32ZP	10	6M	6G	6M	5G	6M
20ZAP	10	4M	8S	4M	7G	2M
32ZAP	10	4M	10	4MD	6G	4D



**Fig. 4.** Panels exposed to the humidity chamber after 33 days. Detail: fungi growth on the samples (80X), and stereomicroscopy photographs of the fungi.

those with ZP as anticorrosive pigment presented a large corroded area around the scribe mark, especially in the case of those protected with the paint with PVC 32 %. Panels painted with the paints containing ZAP, were also corroded around the scribe, but in a lesser degree. These results agree with those obtained by Mousavifard et al., being ZP less resistant than ZAP to aggressive environment in damage painted areas [57]. The important degradation around this cathodic area is due to the increased in pH as a consequence of the oxygen reduction reaction (cathodic delamination) [58]. The main reactions that occur are the  $O_2$  reduction and the hydrolysis of the ester unions in the alkyd resin,



producing the degradation of the resin:

The lower degradation in the case of panels protected with paints containing ZAP indicates that the pigment is able to form a passive film on the substrate [57]. Besides, as the degradation depends on the presence of  $HO^-$ , ZAP may protect the substrate by its buffer properties

[59]. It must be also taken into account that alkyd resins are susceptible of  $HO^-$  degradation [60]. The unpainted area far from the scribe was only corroded in the case of panels 32ZP.

**3.2.2.2. Humidity chamber.** In the humidity chamber, Table 6, blisters appeared very early due to the sensibility of the alkyd resin to this environment [60]. The blistering degree was important in all the cases but higher in 20ZP and 20ZAP panels, probably due to the presence of a greater number of carboxylic groups that are sensitive to water. Similar results were obtained by other authors employing alkyd solvent resins

[1,56]. Corrosion appeared after 18 days, being important in the case of panels 32ZP. In the case of 20ZAP, only few and small corrosion spots can be seen. After 33 days of exposure, corrosion was general and covered between 1 and 3 % of the surface of panels 20ZP and 32ZP. 20ZAP and 32ZAP panels presented less corrosion degradation.

Through stereoscopic microscopy, fungal growth was observed on the panels exposed in the humidity chamber for 33 days except in the

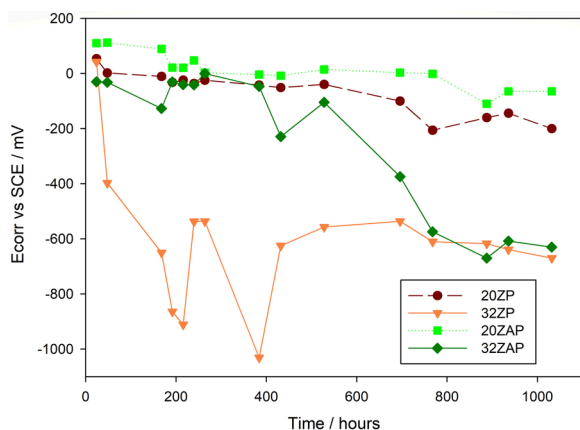


Fig. 5. Corrosion potential ( $E_{corr}$ ) of the painted panels.

case of panels 32ZAP (Fig. 4). In this sense, higher PVC could have brought out ZAP inhibitory activity against fungal growth by increasing the relative concentration of the solids by volume. This could produce, consequently, an increase in the exposed surface area of the solids and a more porous surface of the film. In this sense, Lin and Chen, 2017, noted that higher pore volume of non-biodegradable substrate is beneficial to prevent fungal growth [61].

After isolation and purification of the strains, it was detected that *Aspergillus fumigatus* (Fig. 4, detail) was the strain with a 95 % frequency of appearance, followed by *Aspergillus* sp. (Fig. 4, detail) with 80 % relative frequency. Biofilm formation is a process that occurs in stages in which the irreversible adhesion of the microorganism is fundamental. Filamentous fungi are known to highly biodeteriorate materials [62]. These fungi have an invasive growth that many times manages to break the material on which they grow and at the same time release large amount of acids that favour corrosion processes [63]. Besides, *Aspergillus* as is one of the most common genera associated with biodeterioration of coatings and paints [64].

### 3.2.3. Electrochemical tests

**3.2.3.1. Corrosion potential.** In Fig. 5, the corrosion potential values of panels protected with the different paints can be seen. In the cases of panels protected with paints with lower PVC, the corrosion potentials

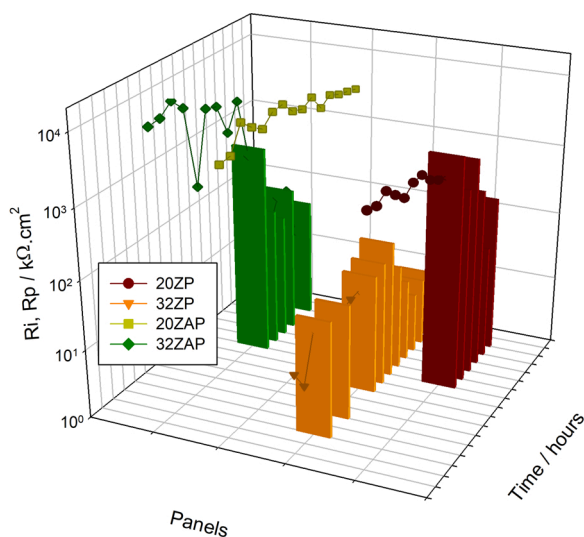


Fig. 6. Ionic resistance ( $R_i$ ), as dots, and polarization resistance ( $R_p$ ), as bars, of the painted panels.

were depleted to more positive values along the test, being slightly more positive in the case of ZAP as anticorrosive pigment. After 700 h,  $E_{corr}$  was near -120 mV. In the case of the panels 32ZP the values oscillated in an important way, probable due to changes in the cathodic / anodic surface area ratio and / or to the instability of pores plug by the corrosion products [65]. However, the values were around -600 mV after 500 h. At longer times, the values were stabilized [66]. In the case of panels 32ZAP, the corrosion potential values were more positive than -200 mV for 400 h but as time elapsed, the values drop, reaching -600 mV after 768 h.

**3.2.3.2. Ionic resistance.** In Fig. 6 the ionic resistances values can be seen. It is observed that panels protected with the paints with lower PVC (panels 20ZP and 20ZAP) presented values higher than  $1000 \text{ k}\Omega \cdot \text{cm}^2$  along the test.  $R_i$  values for panels 32ZAP diminished after 528 h. Panels 32ZP had values lower than  $10 \text{ k}\Omega \cdot \text{cm}^2$  along the tests, indicating very low barrier properties [49,67]. In these last cases, the low barrier properties were expected due to the higher PVC values of the paints [68–70].

**3.2.3.3. Polarization resistance.**  $R_p$  values obtained from linear polarization curves were calculated as

$$R_p = \frac{\Delta E}{\Delta J} \quad (3)$$

being E the potential and J the current density.  $R_p$  values against time can be seen in Fig. 6 while selected polarization curves can be seen in Fig. 7.

$R_p$  values could be determined after 384 h in the case of 32ZP, and after 528 h in the cases of 20ZP and 32ZAP, when the ionic resistances values were lower than  $10^3 \text{ k}\Omega \cdot \text{cm}^2$  (Fig. 6). It can also be seen that  $R_i$  values were lower than  $R_p$  indicating that, after the paint was penetrated by the electrolyte, the metal is protected by the anticorrosive pigment [50] as are the cases of 20ZP and 32ZAP.  $R_p$  values diminished around one order of magnitude along the 43 days of experiment.  $R_p$  values of panels 20ZAP could not be determined as  $R_i$  values were never lower than  $10^3 \text{ k}\Omega \cdot \text{cm}^2$  [50].

## 4. Conclusions

- 1 Alkyd resin based on soybean oil is compatible with polysaccharide resin and anticorrosive alkyd reducible coatings can be prepared mixing both resins.
- 2 Zinc phosphate and zinc aluminium phosphate can be incorporated in the paints.
- 3 Paints with PVC 20 protected better steel than paints with PVC 32, and paints with zinc aluminium phosphate behaved better than those with zinc phosphate.
- 4 Fungal growth was inhibited on paints with PVC 32 and zinc aluminium phosphate as anticorrosive pigment.
- 5 Good selection of the type of pigment and the PVC are important to prevent corrosion and fungi growth in a high humid environment.

### CRedit authorship contribution statement

**M. Martínez:** Investigation, Writing - original draft. **E. Gámez:** Investigation. **N. Bellotti:** Methodology, Supervision. **C. Deyá:** Conceptualization, Methodology, Resources, Visualization, Project administration, Writing - review & editing, Supervision, Funding acquisition.

### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

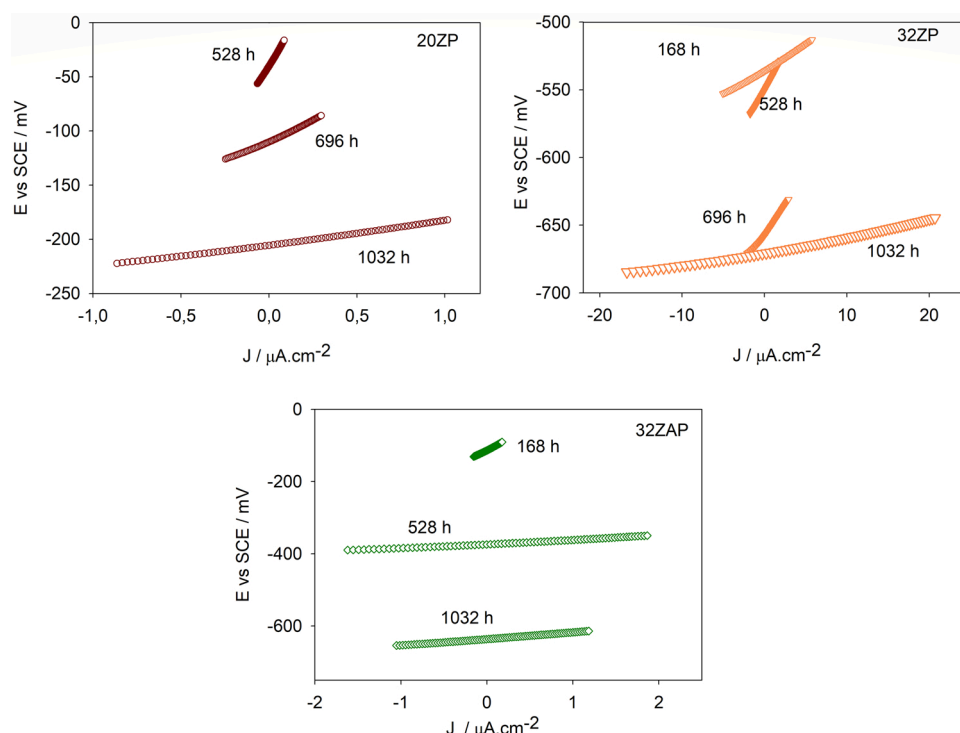


Fig. 7. Polarization curves at different immersion times.

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## Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:<https://doi.org/10.1016/j.porgcoat.2020.106069>.

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