Correlation between natural exposure and artificial ageing test for typical marine coating systems

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ABSTRACT: Natural exposure test at Sanya and artificial ageing test were carried out for four kinds of typical marine coating system, respectively. The gloss, surface morphology, coating impedance, and infrared spectra of the coatings were measured with methods of SEM, EIS, and FT-IR. The correlation between results from artificial ageing test and natural exposure test was discussed. The fluoro-carbon paint showed longer ability to remain high gloss than polyurethane paint. The xenon lamp ageing test and natural exposure test led to similar changes in surface morphology of the coatings. For polyurethane paint under both tests, with extended testing time micropores and cracks occurred gradually on the surface, meantime the coating gloss decreased. The increase of the defect area corresponded well to the decrease of the coating gloss, suggesting that the degradation extent of the coating surface could be reflected by the gloss measurement. The rank correlation study showed that the results by 60 d, 150 d, 300 d, 360 d, and 450 d natural exposure, respectively, were very close to those by 228 h, 443 h, 841 h, 1958 h, and 4013 h artificial ageing test. Hence, the artificial xenon lamp ageing test may be used to simulate natural exposure test at Sanya. © 2016 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 43893.

INTRODUCTION

In marine atmosphere environments, organic coatings suffer from the effects of light, temperature, humidity, oxygen, chlorides, and other contaminants, which result in defects and molecular degradation in the coatings and finally cause the failure of the coating systems.[1–4] Environmental exposure of coatings may really reflect the performance of the coating under typical environments; hence, it is an important method for coating evaluation. However, environmental exposure requires a long time, usually more than several years, and the reproducibility is relatively poor.[5,6] Therefore, during the development of corrosion resistant coatings, the accelerated test methods such as salt fog test and ultraviolet/immersion test are frequently used and the results are compared with those from the environmental exposures to obtain a reasonable correlation. Jelle et al.[5] reported the cracking failure of polypropylene and high density polyethelene coatings due to the formation of carbonyl groups under ultraviolet and salt fog environments. Howard et al.[6] studied the corrosion features at the side edge of a color plate with some accelerated methods including dry/wet alternate test, ASTM B-117[7] and standard Prohesion test. The result from B-117 was found different from the result by outdoor exposure, but Prohesion test provided good consistency. Prosek et al.[8]

studied the correlation among the bubbling times for hot-dip galvanizing materials under the conditions of outdoor exposure, immersion, continued water steam condensation, and dry/wet alternate exposure by EIS. The result from the condensation test was found best close to the data of 5 years outdoor exposure. Deflorian et al.[9] studied the correlation between the results by salt fog test, ultraviolet radiation, and periodic immersion and the results from outdoor exposure at five sites. The immersion time in chloride-containing solutions showed larger influence on the barrier performance of the coating. Fekete et al.[10] reported that the corrosion resistance of a water-base paint increased to some extent after natural exposure following by salt fog and periodic immersion tests. Salt fog test and humidity chamber test may be used to simulate outdoor exposure test for the water-base paint.

Good correlation requires that the electrochemical characteristics and corrosion products are consistent, and the mechanisms of the environmental action and the corrosion reaction dynamics are consistent by the two testing methods.[11] When several materials are evaluated by two different methods, the order of the corrosion rates should be consistent.[10] The statistical regression method, rank correlation method and gray relational degree method are frequently applied in correlation studies,

Table I. The Applied Coating Systems

Coating system	Coating	Thickness/μm
TC1	High build epoxy anti-corrosion paint	150
	Gray polyurethane heat reflection paint	80
TC2	High build epoxy anti-corrosion paint	150
	Gray fluorocarbon top paint	80
TC3	High build epoxy anti-corrosion paint with aluminum red	150
	Suboptical gray polyurethane top paint	80
TC4	Epoxy anti-corrosion paint with iron oxide red	150
	Gray polyurethane top paint	80

such as the corrosion dynamics regression[12–16] and environmental parameters regression[17] by the weight loss data with time. Ramanauskas et al.[18] pointed out that the weight loss data of zinc-rich coating in accelerated humid environment with Cl⁻ and SO_2 may be expressed by $C = At^b$, and also, neutral salt fog test may well simulate corrosion in marine atmosphere. Mendoza et al.[19] proposed a model including the effects of contaminants, wetting time and rain washing, which fitted well to the outdoor results in the rural, industrial and coastal areas in Cuba. Wen et al.[20] calculated the rank correlation coefficients of a steel in simulated sour rain environment, accelerated marine environment, and atmospheric tests. The results showed that the Spearman's rho (a Spearman correlation coefficient) values from the simulated sour rain test and corresponding sour atmosphere test were high during the first and the fourth year, indicating good correlation.

In this article, the natural atmosphere exposure test and the artificial ageing test were carried out for four coating systems which are used for marine ships. The correlation between results of the two tests was discussed according to analysis on the surface morphology, gloss, molecular structure, and electrochemical impedance parameters of the coatings.

EXPERIMENTAL

Materials

Q235 steel plates (250 mm × 150 mm × 3 mm) were used as the metallic substrate. They were treated by sandblasting up to Sa2.5 degree (SIS 05 5900), then rinsed with water, degreased with acetone, and dried with wind blowing. The steel plates were coated by brushing, then were placed in a dessicator at room temperature until complete drying had occurred (15 d). Table I shows the applied coating systems.

The gray polyurethane top paint (For TC1, TC3, and TC4 top coatings), was composed of urethane and hydroxy acrylic resins, with titanium dioxide, dryer, auxiliaries, and solvents, to obtain good color retention, weather-proof performance, good flexibility, and adhesive force. More titanium dioxide pigments were added to TC1 top paint to increase the heat reflection performance, and some delustering agents such as silicone dioxide and barium sulfate were added to TC3 top paint to decrease the glossiness of the coating. The film forming matter in TC2 top paint is urethane resin and hydroxy acrylic resin. The C—F bonds form a close protective layer surround the carbon chains,

which protects the main molecular chains and gives the fluorocarbon coatings good weather-proof performance, high chemical stability, and corrosion resistance.

The high build epoxy anti-corrosion paint was composed of bisphenol A epoxy resin and some curing agents. In TC1 and TC2 primers, the curing agents were tertiary amine and cardanol modified phenolic amine. In TC3 and TC4 primers, the curing agent was polyamide.

The outdoor exposure test was carried out in a typical marine atmospheric environment at Sanya, Hainan province. The test followed GB/T9276-1996 standard, and the period was 24 months. The gloss, chromatic aberration, and impedance spectra of the samples were measured each month. The artificial ageing test followed GB1865-2009 standard.[21] The samples were periodically exposed under a xenon lamp with a wave length of 340 nm for 102 min, then were sprayed with 3.5 wt % NaCl solution (the conductivity was 56.68 mS/cm) for 18 min. During the drying period the relative humidity was remained at 40–60% and the temperature was 38°C.

Measurements

The gloss and chromatic aberration were measured with a BYK Color-Guide instrument (Germany). The sample was illuminated by the ring-shaped lamps at 45°, and the observation was at 0° perpendicular to the coating surface. The other side of the sample that was not illuminated by the lamp was taken as the reference plane, and for each sample twelve parallel measurements were performed and the average value was automatically calculated.

A PARSTAT2273 electrochemical working station (Princeton) was used for electrochemical impedance measurement. The reference electrode was a saturated calomel electrode (SCE), the auxiliary electrode was a platinum plate, and the coating sample was the working electrode. A 10 mV perturbation was applied and the scanning frequency range was 100 kHz–10 mHz. The solution was 3.5 wt % NaCl and the impedance spectra were measured at the open circuit potential at room temperature.

A TENSOR27 Fourier transform infrared spectrometer was used to study the changes of the organic functional groups during the ageing process. 1–2 mg sample was mixed with 200 mg KBr and ground to fine powders, then was suppressed to a transparent sheet by a pressure larger than 15 MPa in an oil press. The measuring range was 600 cm⁻¹−4000 cm⁻¹.
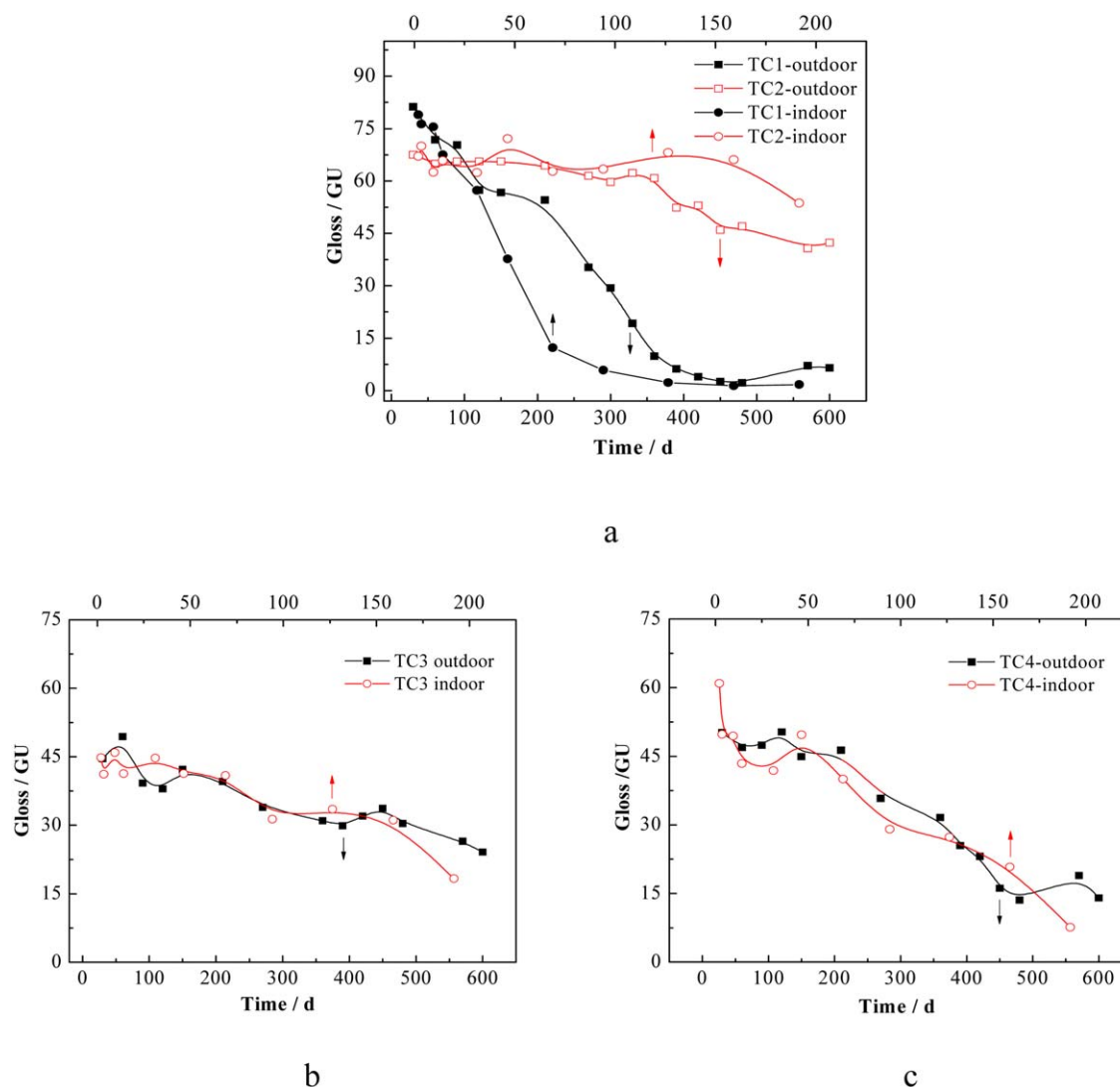
Figure 1. Variations of the coating gloss with testing time, (a) TC1 and TC2, (b) TC3, (c) TC4. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

A Hitachi S4700 field emission scanning electronic microscope (SEM) was used to observe the coating morphology after the indoor or outdoor tests.

RESULTS AND DISCUSSION

Gloss Measurement

The gloss refers to the ability of the surface to reflect the light cast on it. The larger the roughness of the coating surface, the lower the reflecting ability, and the gloss. Hence, gloss may be used to indicate the aging degree of coatings.[22] Figure 1 compares the gloss variations of the four types of coatings with time under natural exposure and artificial aging conditions, respectively. For all the four coatings the gloss showed decreasing tendency as the exposure time extended. For naturally exposed TC1 coating, the gloss remained relatively high values before 100 d, and then decreased rapidly [Figure 1(a)]. After natural exposure for 330 d, the gloss of TC1 coating was totally lost (by GB/T-1766-2008). For TC1 coating in the artificial ageing test, the gloss was totally lost after 69 d. TC2 coating

remained high gloss during both the natural exposure and artificial ageing tests, which may be due to that F—C bond has high bond energy and fluorocarbon coating possesses low surface energy and good hydrophobic property. TC3 coating was a semigloss coating,[5] the gloss of which showed a relatively low value at the beginning and decreased slowly with the exposure time [Figure 1(b)]. For TC4 coating, the gloss changed slowly in 210 d for natural exposure and 69 d for indoor test, respectively, and then decreased continuously [Figure 1(c)]. The above results show that for all the four coatings tested, the gloss change rules obtained by the artificial ageing tests are well correlated with those obtained by the natural exposure tests.

Surface Morphology of the Coatings

Figure 2 shows the morphologies of TC1 and TC2 coatings by SEM. After artificial ageing for 191 d, many cracks and micropores can be observed on the surface of TC1 coating [Figure 2(b)]. Figure 2(c) shows the morphology of TC1 coating sample
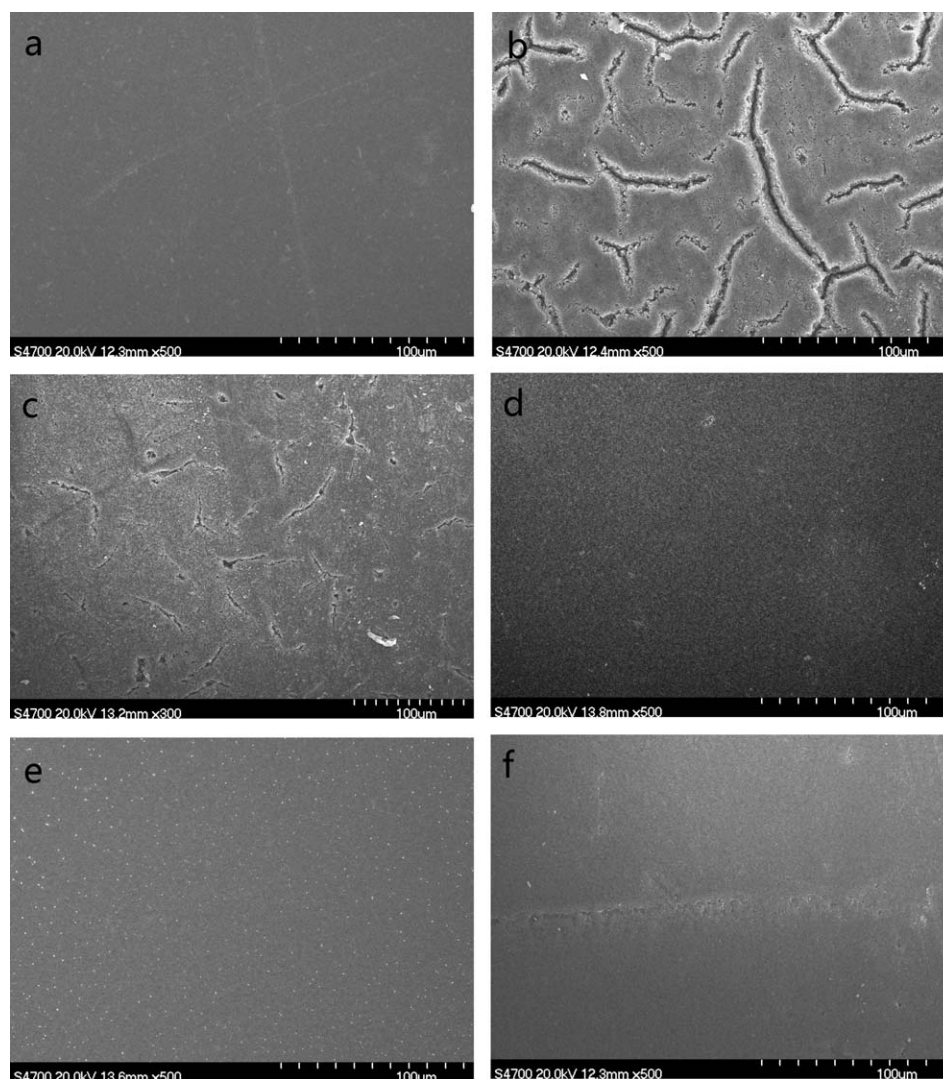
Figure 2. Surface morphologies of TC1 and TC2 coatings after different tests, (a) Original TC1, (b) TC1 after artificial ageing for 191 d, (c) TC1 after natural exposure for 600 d, (d) Original TC2, (e) TC2 after artificial ageing for 191 d, (f) TC2 after 600 d natural exposure.

after 600 d natural exposure. Also many cracks and defects can be seen, similar to the morphology by artificial ageing. Under the effect of violet radiation, molecules in the coating got degraded and micropores were produced, which may develop into microcracks.[23] The degraded products may also leave the surface, resulting in rough surface. The above results show that natural exposure and artificial ageing tests lead to similar sur-

face morphology for the coating, suggesting that the degradation mechanisms may be similar.

Figure 2(e,f) show the morphologies of the artificial aged and natural exposed coating samples, respectively, after 191 d and 600 d tests. Both samples show flat and intact surfaces and no obvious defects can be observed. This result means that the
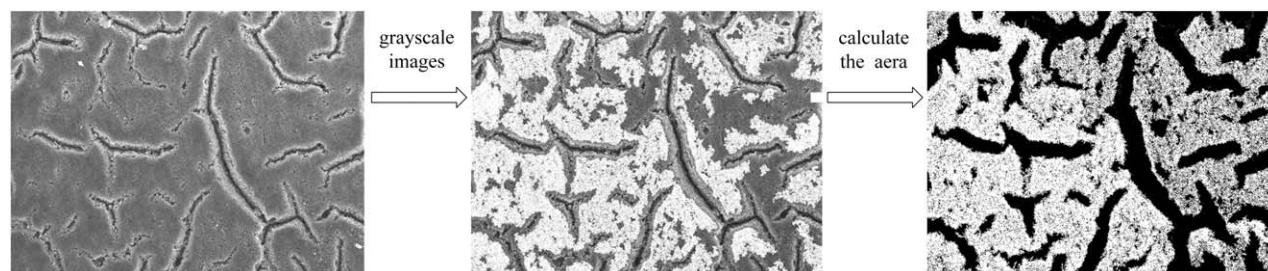

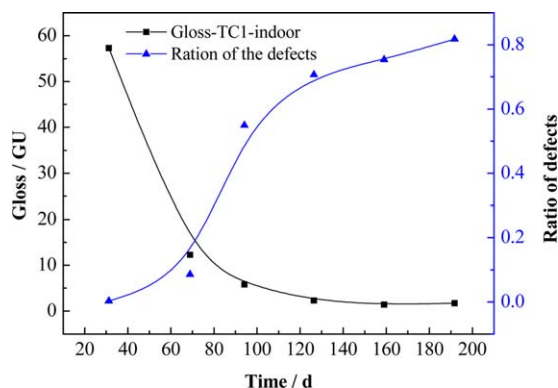**Figure 3.** The gray level image and calculation of defect areas.

Figure 4. Variations of gloss and defect area ratio with time. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary. com.]

fluorocarbon coating possesses much better resistance to violet radiation and ageing than polyurethane coating.

According to the image of TC1 coating by artificial ageing test [Figure 2(b)], grayscale images were obtained using Photoshop software, as shown in Figure 3. The area ratio of cracks and pores on the image was calculated using MATLAB and the result is shown in Figure 4. It is seen that the increase of the defect area corresponds well to the decrease of the coating gloss. This result indicates that the degradation extent of the coating surface may be reflected by the gloss measurement.

Chemical Changes of the Aged Coatings

Figure 5 shows the infrared spectra of TC1 and TC2 top coatings after natural exposure for 600 d or artificial ageing test for 191 d. In Figure 5(a), the broad band with peak at 3475 cm^{-1} could be assigned to the stretching vibration of —OH and N—H, the peaks at 2924 cm^{-1} and 2852 cm^{-1} are due to alkyl bands, the peak at 1729 cm^{-1} is due to the vibration of C=O bonds in esters, the peak at 1690 cm^{-1} is attributed to the

asymmetric C=O stretching mode of urethane, the peak at 1520 cm^{-1} belongs to the vibration of C—N in urethane, and the peak at 1452 cm^{-1} is due to the C—H bending vibration of CH$_2$.[24–26] In Figure 5(b), the peak at 1400 cm^{-1} is attributed to the deformation vibration absorption of —CH$_2$ linked with —CF$_2$, and the peak at 1115 cm^{-1} is due to the stretching vibration of C—F in fluorocarbon. For TC1 top coating, in Figure 5(a), compared with the original coating, after exposure for a long time the peak intensity at 3475 cm^{-1} increases, the peak intensity at 2924 cm^{-1} decreases, and the peak at 1520 cm^{-1} almost disappears. The increase of the absorption peak at 3475 cm^{-1} indicates the increase of hydroxyl, which can be related to water molecules trapped inside the alkyd network due to hydrogen bond interactions with carbonyl or other polar groups.[26] The decrease of 1520 cm^{-1} and 2924 cm^{-1} peaks are probably due to the loss of urethane and CH groups occurred during UV weathering.[27,28] Based on the literatures,[24,29,30] the decrease of C—N bond at 1520 cm^{-1} may result in the formation of amino free radical, alkyl free radical and CO$_2$, as shown in Figure 6. In Figure 5(a), we can see that the infrared spectra for TC1 top coating under two aged conditions show the same characteristics. Therefore, in the artificial ageing test the ageing mechanism is the same as that in natural exposure. The fracture of bonds in molecular chains would cause decreases of molecular weight and the binding force among different components, leading to increased porosity in the coating.

For TC2 top coating, the infrared spectra of the samples after artificial ageing test for 191 d and natural exposure for 600 d show no obvious difference to the spectra of the original sample without ageing [Figure 5(b)], indicating that during both artificial and natural exposure tests the fluorocarbon coating remains good chemical or structural stability.

Figure 7 shows the infrared spectra of TC3 and TC4 top coating samples after natural exposure and artificial ageing tests, respectively. The peak at 1640 cm^{-1} is related to the symmetric C=O
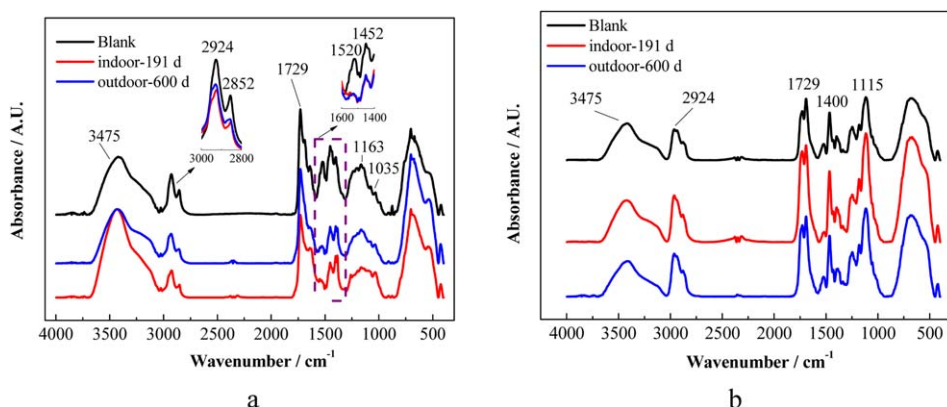

a

b

Figure 5. Infrared spectra for natural exposed and artificially tested TC1 and TC2 samples, (a) TC1, (b) TC2. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 6. Photodegradation mechanism of polyurethane.

Figure 7. Infrared spectra for natural exposed and artificially tested TC3 and TC4 samples, (a) TC3, (b) TC4. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
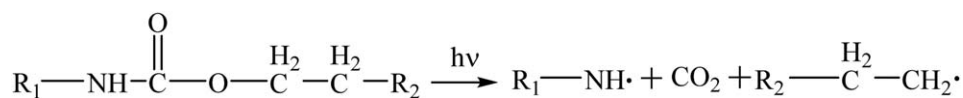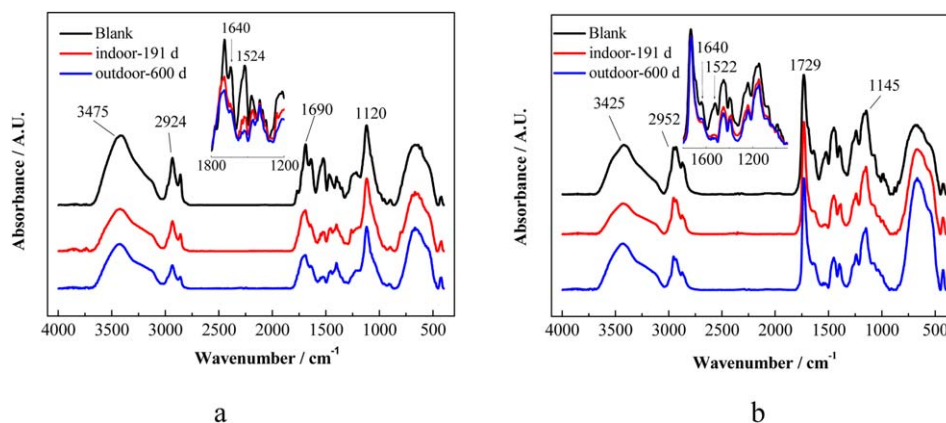
stretch in biuret linkages.[27] After natural exposure or artificial ageing test, for TC3 top coating, the peaks at 2924 cm^{-1} and 1690 cm^{-1} decrease, and the peaks at 1524 cm^{-1} and 1640 cm^{-1} almost disappear [Figure 7(a)]. For TC4 top coating, both 1640 cm^{-1} and 1524 cm^{-1} peaks reduce obviously [Figure 7(b)] after the ageing tests. These all indicate that oxidation may contribute to the degradation of urethane links.[25,27]

The above results are also consistent to those by the gloss measurements, showing that for the tested coatings the decrease of gloss is accompanied by degradation of the coating and microcracks and pores on the surface.

In addition, for each coating system, after the two ageing tests (artificial ageing test for 191 d and natural exposure for 600 d) FTIR measurement was also performed on the epoxy primer. The spectra results show no difference compared with the spectra of the original sample without ageing.

Rank Correlation Evaluation

Based on the gloss data of the four kinds of coatings in the artificial ageing and natural exposure tests, Spearman Rank was used to evaluate correlation between two test methods. The eq. (1) describes the calculation of Spearman Rank, R_s[31]:

$$R_s = 1 - \frac{6 \times \sum (\text{rank}_{\text{outdoor}} - \text{rank}_{\text{indoor}})^2}{n(n^2 - 1)} \quad (1)$$

where n is the number of samples (four in this study), rank is an ordinal value of the gloss of the four coatings in natural

Table II. The Calculated Rank Correlation Coefficients between the Results of Indoor Ageing and Outdoor Exposure

Outdoor exposure time (d)	Indoor ageing time				
	228 h	750 h	1655 h	3035 h	4600 h
60	1	0.6	−0.6	−0.6	−0.6
150	0.8	1	0.2	0.2	0.2
300	−0.2	0.6	0.8	0.8	0.8
360	−0.6	0.2	1	1	1
450	−0.6	0.2	1	1	0.8

exposure test (outdoor) and artificial ageing test (indoor), respectively. The sample with the highest gloss is ranked 1. The closer the R_s value to 1, the higher the correlation.

According to the gloss change of TC1 coating (GB 1766-2008), several natural exposure time points (60 d, 150 d, 300 d, 360 d, 450 d) and artificial ageing time points (228 h, 750 h, 1655 h, 3035 h, 4600 h) were selected. Then, the gloss values of the four coatings at the same testing time were ranked and the Spearman Rank R_s was calculated. The results are shown in Table II. It can be seen that, there is a good correlation between natural exposure results and artificial ageing results (a R_s value of > 0.8). The above SEM and infrared spectra results show that the aging rule for each coating in the artificial ageing test is consistent with that in natural exposure test, while the aging degrees of the four coatings are different. Based on the change of gloss values of 60 d, 150 d, 300 d, 360 d, and 450 d in natural exposure test, the according artificial ageing time was calculated, then the times averaged,[32] and the correlation between natural exposure and artificial ageing tests was obtained, shown in Table III. For 60 d, 150 d, 300 d, 360 d, and 450 d in natural exposure test, the correspondent artificial ageing time was 228 h, 443 h, 841 h, 1957 h, and 4013 h, respectively.

Impedance Measurements of the Coatings

Electrochemical impedance spectra were measured for the four coating systems after natural exposure or artificial ageing test, respectively. Figure 8 shows the Nyquist plots and Bode plots for TC1 coating under the two testing conditions as example, and in Figure 9 the impedance values at low frequency (0.01 Hz) of the four coating systems are displayed as a function of testing time. For all the four coating systems, in the first 360 d of natural exposure [Figure 9(a)] the coating impedance values

Table III. Corresponding Time between Natural Exposure Test and Artificial Ageing Test

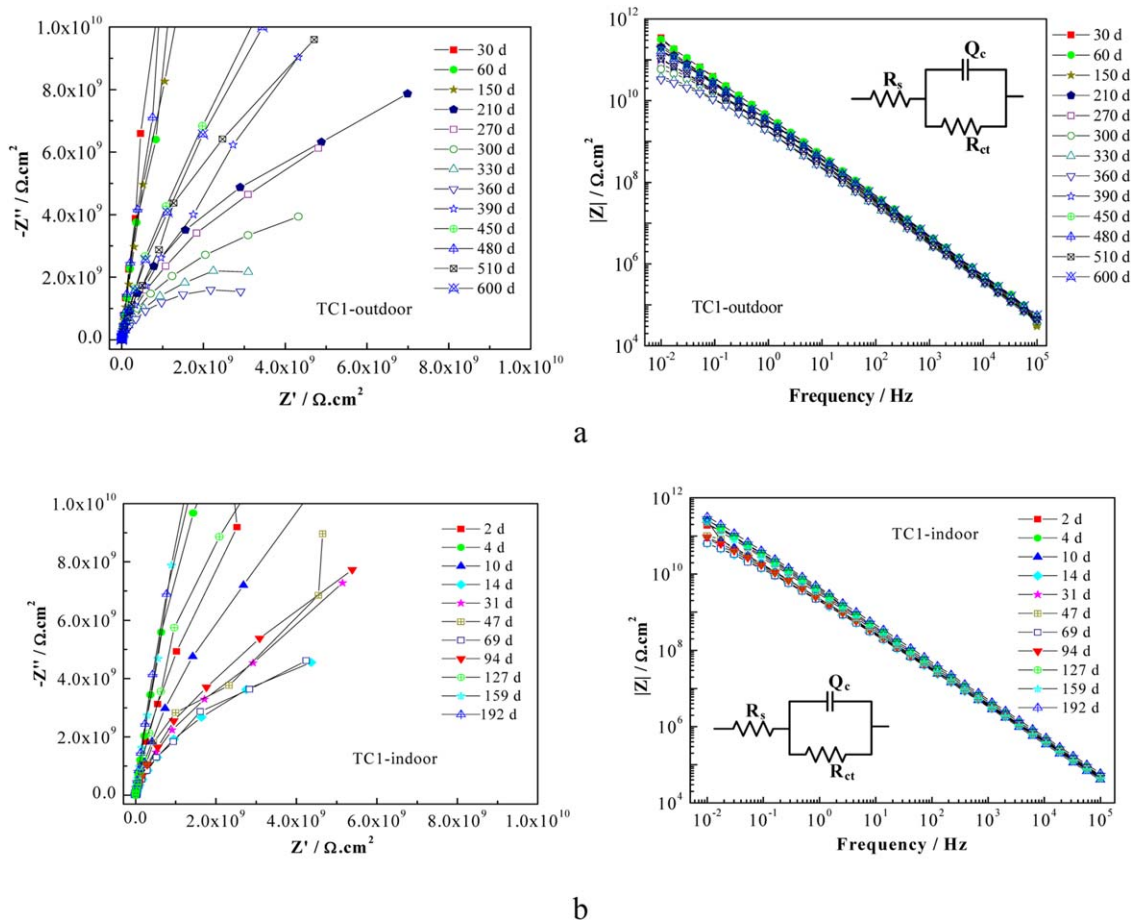| Outdoor exposure time	60 d	150 d	300 d	360 d	450 d
Indoor ageing time	228 h	443 h	841 h	1957 h	4013 h

Figure 8. Nyquist and Bode plots for TC1 coating systems under different test conditions, (a) natural exposure test, (b) artificial ageing test. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

decreased gradually with time, which may be attributed to the permeation of electrolytes and increased micropores in the coatings. After 360 d, the impedance values showed slightly increase, probably due to some micropores filled with the reacting products between electrolytes and the fillers in coatings. For all the tested coating systems, during the testing time the impedance remained high values above 10^{10} Ω cm^2, indicating good barrier performance.[33–35] The experimental data were analyzed using a

simple Randles circuit, shown in Figure 8, to derive information on coatings. In this model, a resistor R_c is in parallel with a capacitor Q_c and then in series with a resistor R_s, in which R_s represents the solution resistance, R_c is the coating pore resistance and Q_c is the coating capacitance. In the present EIS test, the electrolyte resistance R_s can be negligible due to the high conductivity of the alkaline solution. The coating capacitance Q_c is related to the behavior of electrolyte penetration in
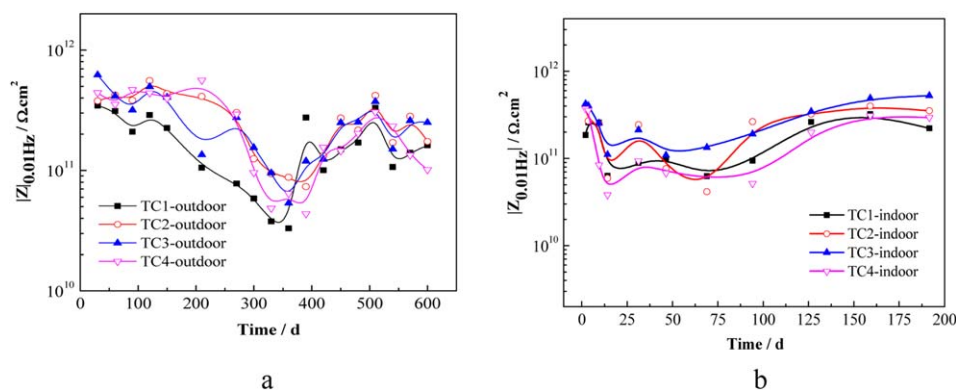

Figure 9. Low frequency impedance ($|Z|_{0.01Hz}$) values of the coating systems with testing time, (a) natural exposure test, (b) artificial ageing test. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 10. Coating capacitance values of four coating systems with testing time, (a) natural exposure test, (b) artificial ageing test. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
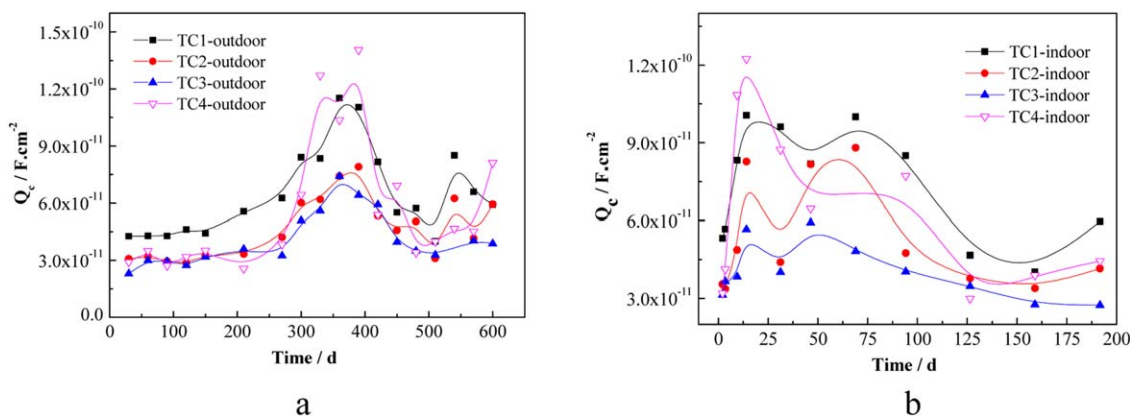
coatings. Figure 10 shows the coating capacitance values of four coating systems under two testing conditions with time. It can be seen that the Q_c of TC2 and TC3 coatings keep lower values and with relatively small change. The Q_c values of TC1 and TC4 coatings are relatively higher with bigger change, and among of them TC1 coating has the biggest Q_c. This probably is related to more defects occurred on the TC1 coating surface. The EIS results also show that although some defects occurred on the coating surface after natural exposure or artificial ageing test, the anti-corrosive primer as the bottom layer still remained good protective property. Therefore, the changes in coating gloss reflect mainly degradation of the top paint, but not the performance of the primer. This is also consistent with the FTIR spectra results of the four primers.

Recently, some authors[36–40] analyzed the frequency dependence of the evaluated coating parameters in some electrolytes and pointed out that the resistivity and permittivity varied with the applied frequency. This was attributed to an inhomogeneous uptake of electrolytic solution into the coatings. Further, we will try to study the variation of the coating properties by analyzing the frequency response of the coating parameters in future work.

CONCLUSIONS

1. For the four coatings tested, the change rules of the coating gloss under xenon lamp ageing test and natural exposure test at Sanya were similar. Fluorocarbon paint showed longer ability to remain high gloss than polyurethane paint. The addition of light stabilizer in suboptical polyurethane paint was helpful to keep the gloss.

2. The xenon lamp ageing test and natural exposure test led to similar changes in surface morphology of the coatings. For polyurethane paint under both tests, with extended testing time micropores and cracks occurred gradually on the surface, meantime the coating gloss decreased. The increase of the defect area corresponded well to the decrease of the coating gloss, suggesting that the degradation extent of the coating surface may be reflected by the gloss measurement.

3. The rank correlation study showed that the results by 60 d, 150 d, 300 d, 360 d, and 450 d natural exposure, respectively, were very close to those by 228 h, 443 h, 841 h, 1958

h, and 4013 h artificial ageing test. Hence, the artificial xenon lamp ageing test may be used to simulate outdoor exposure test at Sanya.

4. Although some defects occurred on the coating surface after natural exposure or artificial ageing tests, the whole coating systems remained high impedance during the test time. Therefore, the changes in coating gloss reflected mainly degradation of the top paint, but not the performance of the primer.

5. Above results show that the degradation process of coatings may be studied with EIS, SEM, and FT-IR. Further, the dynamic mechanical analysis, hardness test, pull-off adhesion test, and thermal analysis such as TG-DCS may be applied to analyze and predict the failure of coatings.

REFERENCES

1. Jacques, L. F. E. *Prog. Polym. Sci.* **2000**, *25*, 1337.

2. Armstrong, R. D.; Jenkins, A. T. A.; Johnson, B. W. *Corros. Sci.* **1995**, *37*, 1615.

3. Hao, M. L.; Cao, X. J.; Feng, X. H.; Shi, X. M.; Fang, X. Z. *Ordnance Mater. Sci. Eng.* **2006**, *29*, 28.

4. Brunner, S.; Richner, P.; Müller, U.; Guseva, O. *Polym. Test.* **2005**, *24*, 25.

5. Jelle, B. P.; Nilsen, T. N. *Constr. Build. Mater.* **2011**, *25*, 2122.

6. Howard, R. L.; Lyon, S. B.; Scantlebury, J. D. *Prog. Org. Coat.* **1999**, *37*, 91.

7. Standard Practice for Operating Salt Spray (Fog) Apparatus; ASTM: West Conshohocken, PA, 2007, ASTM B 117.

8. Prosek, T.; Nazarov, A.; Stoulil, J.; Thierry, D. *Corros. Sci.* **2012**, *61*, 92.

9. Deflorian, F.; Rossi, S.; Fedel, M. *Corros. Sci.* **2008**, *50*, 2360.

10. Fekete, É.; Lengyel, B. *Prog. Org. Coat.* **2005**, *54*, 211.

11. Montoya, P.; Díaz, I.; Granizo, N.,; Fuente, D.; Morcillo, M. *Mater. Chem. Phys.* **2013**, *142*, 220.

12. Luo, H.; Li, X. Gg; Dong, C. F.,; Xiao, C. K. *J. Chin. Soc. Corros. Prot.* **2013**, *33*, 193.

13. Wang, Z. Y.; Yu, G. C.; Han, W. *Corros. Sci. Prot. Technol.* **2004**, *16*, 70.

14. Liu, J. R.; Zhang, W. L.; Ma, Y.; Huang, X. Q.; Lang, F. J.; Shi, Z. *Res. Iron Steel* **2013**, *41*, 35.

15. Wang, J.; Wang, Z. Y.; Ke, W. *Mater. Chem. Phys.* **2010**, *124*, 952.

16. Corvo, F.; Perez, T.; Dzib, L. R.; Martin, Y.; Castañeda, A.; Gonzalez, E.; Perez, J. *Corros. Sci.* **2008**, *50*, 220.

17. Deflorian, F.; Rossi, S.; Fedrizzi, L.; Zanella, C. *Prog. Org. Coat.* **2007**, *59*, 244.

18. Ramanauskas, R.; Muleshkova, L.; Maldonado, L.; Dobrovolskis, P. *Corros. Sci.* **1998**, *40*, 401.

19. Mendoza, A. R.; Corvo, F. *Corros. Sci.* **1999**, *41*, 75.

20. Wen, B. W.; Zhang, L. W.; Li, J. H.; Lai, J. P. *Equipment Environ. Eng.* **2006**, *3*, 97.

21. GB1865-2009 Standard Test Method for Paints and varnishes—Artificial Weathering and Exposure to Artificial Radiation—Exposure to Artificial Radiation Exposure to Filtered Xenon-Arc Radiation; The National Coating and Paint Standardization Technical Committee: Beijing, **2009**.

22. Yang, X. F.; Vang, C.; Tallman, D. E.; Bierwagen, G. P.; Croll, S. G.; Rohlik, S. *Polym. Degrad. Stabil.* **2001**, *74*, 341.

23. Ye, M. Q.; Jin, X. H.; Chen, N. H.; Lin, C. J.; Li, K.; Yao, J. H.; Wang, J. J.; Ye, Z. J.; Xu, C. Q.; Huang, S. Z.; Tao, N. W.; Guo, A. H. *Equipment Environ. Eng.* **2012**, *8*, 4.

24. Perrin, F. X.; Irigoyen, M.; Aragon, E.; Vernet, J. L. *Polym. Degrad. Stabil.* **2000**, *70*, 469.

25. Colin, A.; Baba, M.; Bussiere, P. O.; Cavaletti, E.; Nizeyimana, F.; Therias, S. *Polym. Degrad. Stabil.* **2015**, *114*, 115.

26. Perrin, F. X.; Merlatti, C.; Aragon, E.,; Margaillan, A. *Prog. Org. Coat.* **2009**, *64*, 466.

27. Merlatti, C.; Perrin, F. X.; Aragon, E.; Margaillan, A. *Polym. Degrad. Stab.* **2008**, *93*, 896.

28. Fratričová, M.; Šimon, P.; Schwarzer, P.; Wilde, H. W. *Polym. Degrad. Stab.* **2006**, *91*, 94.

29. Gao, J.; Li, X. G.; Ge, S; Wang, L. Presented at the 5th Sino-American Academic Symposium on Environmental Corrosion and Degradation Test of Material, Shang Hai, **2008**; p 3.

30. He, C. L.; Deng, J. G.; Zhang, Y. S. *Polyurethane Ind.* **2002**, *17*, 1.

31. Cocuzzi, D. A.; Pilcher, G. R. *Prog. Org. Coat.* **2013**, *76*, 979.

32. Yuan, H. H.; Sun, X. L.; Zhang, H. *Tech. Res. Dev.* **2009**, *24*, 35.

33. Bierwagen, G. P.; He, L.; Li, J.; Ellingson, L.; Tallman, D. E. *Prog. Org. Coat.* **2000**, *39*, 67.

34. McIntyre, J. M.; Pham, H. Q. *Prog. Org. Coat.* **1996**, *27*, 201.

35. Jr, L. H. *Prog. Org. Coat.* **1979**, *7*, 79.

36. Touzain, S. *Electrochim. Acta* **2010**, *55*, 6190.

37. Alexander, C. L.; Tribollet, B.; Orazem, M. E. *Electrochim. Acta* **2015**, *173*, 416.

38. Amand, S.; Musiani, M.; Orazem, M. E.; Pebera, N.; Tribollet, B.; Vivier, V. *Electrochim. Acta* **2013**, *87*, 693.

39. Nguyen, A. S.; Musiani, M.; Orazem, M. E.; pebera, N.; Tribollet, B.; Vivier, V. *Electrochim. Acta* **2015**, *179*, 452.

40. Schiller, C. A.; Strunz, W. *Electrochim. Acta* **2001**, *46*, 3619.