

Corrosion behavior of weathering steel in tropical atmosphere of Vietnam

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Abstract— Weathering steel (WS) of Corten B type was exposed for three years at three test sites including urban and marine sites. After one year-exposure, WS showed the protective ability better than carbon steel (CS) did. The corrosion loss of WS fitted well power law with high correlations. The characterization of rust layer was studied by using X-ray diffraction, optical microscope and SEM-EDX. Goethite (α -FeOOH), akaganeite (β -FeOOH), lepidocrocite (γ -FeOOH), maghemite and magnetite were founded in corrosion product formed on WS for 3, 6, and 12 month-exposure; The structure of rust layer of WS composed of two layers – the inner layer dark-grayish, dense and good adherent to steel substrate; and the outer layer porous and reddish; The morphologies of rust surface showed the sandy and flowery structure typical for lepidocrocite and cotton balls structure typical for goethite; Cr and Cu distributed in the inner layer of rust – at distance of 40-60 μ m from steel substrate. The resistance of rust layers was evaluated by using electrochemical impedance spectroscopy measurement. This proved the predominance of WS over CS in corrosion resistance. The influence of environmental factors on protectiveness of WS was also discussed in this paper.

Keywords: weathering steel, rust layer, corrosion loss, rust resistance, protectiveness.

I. INTRODUCTION

Weathering steels (WS), also known as low-alloy steels, are mild steels containing a small amount of Cr, Cu, Ni and P (less than 3-4%) [1], they have been used very popular in the world from the viewpoint to reduce maintenance cost of steel structure. The enhanced corrosion resistance of WS is due to the formation of protective rust layer (patina), which is dense and good adherent to steel substrate, and therefore they impede the corrosion process. The formation of such protective rust layer has been shown to depend upon the existence of certain alloying elements in the steel and on exposure conditions [2, 3].

The environmental conditions which promote the formation of protective rust layers are existence of wet-dry cycling, absence of very long time of wetness, absence of marine component in atmosphere. It is now accepted that wet/dry cycling is necessary to form a dense and adherent rust layer, with rainwater washing the steel surface well, accumulated moisture draining easily, and fast drying action (low time of wetness) [2]. Surfaces protected from sun and rain (sheltered) tend to form the loose and poorly compact rust, while surfaces freely exposed to the sun and rain produce more compact and protective rust layers. The presence of atmospheric SO₂ within certain limits favors the development of protective patina, while the chlorides suspended in marine atmosphere impair its protective properties [3,4]. For instance, in Japan, the use of unpainted conventional WS is not recommended in marine atmosphere with chloride deposition rate higher than 6mg/m².day [5].

Vietnam is located in wet tropical area with very long coastal line of more than 3000km, this condition seems not advantageous to exploring steels in atmosphere. However, the great total of rainfall with high speed and the longtime of sunshine in summer may promote the existence of protective rust layer. For example, the longtime of sunshine improves the compactness of rust layer; and the great total of rainfall both washes away corrosive pollutants on steel surface and partly washes them away from the porous outer rust layer. In addition, unstable and porous outer rust layer may be removed under high speed raining, according to [6] that shortens wetting time in wet/dry cycles and slows down corrosion.

The corrosion behavior of carbon steel (CS) in tropical climate of Vietnam has been studied for long time with the summary of results given in [7, 8]. Up to now, WS is used not so popularly in Vietnam, thus there is very lack of information about atmospheric corrosion behavior of WS. Nevertheless, some results of characteristics of corrosion product layer formed on WS in the initial stage of exposure were published recently in [9]. It was founded that the dense α -FeOOH phase appeared very soon - just after 1-3 days exposure, and the alloyed elements as Cr and Cu appeared in the rust formed in the early stage of exposure (1-7 days) which were found to be rich in the rust layer closest steel substrate.

This paper introduces the corrosion behavior of WS (Corten B type) exposed for three years at three test sites including urban and marine sites. The study focused on: (1) The evolution of WS corrosion with time; (2) The chemical composition and structure of corrosion product formed on WS; (3) The distribution of alloying elements (Cr, Cu) in rust layer; and (4) Corrosion resistance of rust layers formed on WS exposed for three years. The mild carbon steel was investigated in order to compare with WS. The influences of climatic and environmental factors were discussed in the paper.

II. EXPERIMENT

2.1 Material and exposure test

WS of Corten B type and CS with chemical compositions shown in tab.1 were used for study. The samples with size of 100x75x 2mm were exposed on the racks inclined 45°, the skyward surface of samples were faced to South at the inland test sites, and faced to the sea at the coastal test sites.

The test sites were Hanoi (HN), Dong Hoi (DH), and Phan Rang (PR) with the main climatic and environmental factors shown in tab 2. The chloride deposition rates ($\text{mgCl}^-/\text{m}^2\cdot\text{day}$) were determined by “dry gauze” according to standard JIS Z2382/1998 [10]. The above parameters were collected during exposure time.

The tested samples were taken off after 1, 3, 6, months and 1, 2, 3 years.

TABLE 1
THE CHEMICAL COMPOSITION OF WEATHERING STEEL, MASS

Elements	C	Mn	Si	Ni	Cr	Cu	Mo	V	Ti	Al	W	Co	Fe
WS	0.111	1.06	0.236	0.186	0.528	0.320	0.048	<0.005	< 0.005	0.0158	< 0.05	< 0.005	97.4
CS	0.135	1.35	0.341	0.059	0.024	0.061	0.048	< 0.005	< 0.005	0.0243	< 0.05	< 0.005	97.9

TABLE 2
THE CLIMATIC AND ENVIRONMENTAL FACTORS OF TEST SITES

Test sites	Characteristics	T, °C	Total of rainfall, mm/y	RH, %	TOW, h/y	TOW, %/y	Ratio of dry /wet time	Time of sunshine h/y	Cl, $\text{mg}/\text{m}^2\text{d}$
Hanoi	Urban	24.2	1606	78.6	4616	52.7	0.9	1103	3.0
Dong Hoi	Coastal	24.8	2445	83.1	5705	65.1	0.54	1606	18.7
Phan Rang	Coastal	27.1	1130	76.5	3719	42.4	1.35	2751	8.5

(*): TOW is Time of Wetness, TOW, % = TOW/8760 hours

2.1.1 Corrosion loss

The corrosion losses (CL) of steel were determined by mass loss of tested samples after removing corrosion product according to the standard ISO 8407 [11].

2.1.2 Characterization of corrosion product

The morphology of rust surface was observed using Scanning Electron Microscope. The cross-sections of exposed WS sample were mounted in epoxy resin to prevent the removal of corrosion products. These sections were prepared by grinding with 200-1200 silicon carbide discs, using ethanol as lubricant and polished with diamond pastes with 9 to 3 μm . Finally the surfaces were etched by 2% Nital solution (98% ethanol and 2% nitric acid), rinsed in cold tap water, dipped in ethanol and dried in forced hot air. The prepared cross-sections were observed under an optical microscope. The composition of compounds in the rust were determined by X-ray diffraction

2.1.3 Distribution of alloying elements

In order to investigate the distribution of Cr and Cu elements in the rust, the cross-section of rust prepared as above were analyzed by energy dispersive spectroscopy (EDS) using line scan technique.

2.2 Electrochemical impedance measurement

The electrochemical impedance spectroscopy (EIS) measurement was carried out in 0.1 M aqueous NaCl solution. A three electrodes cell was used with platinum as counter electrode, saturated calomel electrode as reference electrode, and the rusted

samples with an exposed area of 10.84cm^2 as the working electrode. The frequency range for EIS covered from 100 KHz to 10 mHz with a 10mV amplitude signal at open circuit potential.

III. RESULTS AND DISCUSSION

3.1 Corrosion loss of weathering steel

At the early stage of exposure (less than one year) the CL of CS and WS are similar at all test sites. After one year-exposure the CL of WS become lower than that of CS, this was expressed clearly at Hanoi and Dong Hoi sites (fig. 1a, 1b), whereas at Phan Rang site the CL of WS and CS are negligible different (fig. 1c). After three year-exposure, the CL of WS become obviously lower in comparison with CS.

The above results can be explained as follows: the higher humidity and longer TOW at Hanoi and Dong Hoi promoted the existence of Cu, Cr in wet period which produced the more protective rust layer on WS than that on CS. Meanwhile, in the drier climate with shorter TOW of Phan Rang, the alloying elements existed slower, thus WS has not yet shown the preminent protectiveness in comparison with CS.

By the same reasons, the CL of WS are in order as follows: Dong Hoi > Hanoi > Phan Rang (fig.2). It can be explained in detail that the atmospheric condition at Dong Hoi site with highest RH (83.1%), longest TOW (5705h/year), and highest airborne salinity (tab.2) accelerated corrosion of WS. Whereas, the climate at Hanoi is less humid with shorter TOW and very low chloride content in the air, thus the CL of WS is lower than that at Dong Hoi. Especially, the CL of WS is lowest at Phan Rang where the climate is very dry with TOW of 3719h/y and very little rain (1130mm/y), even though the airborne salinity at Phan Rang ($8.5\text{ mgCl}^-/\text{m}^3$) is higher than that in Hanoi ($3.0\text{ mgCl}^-/\text{m}^3$), it may not dominate corrosion.

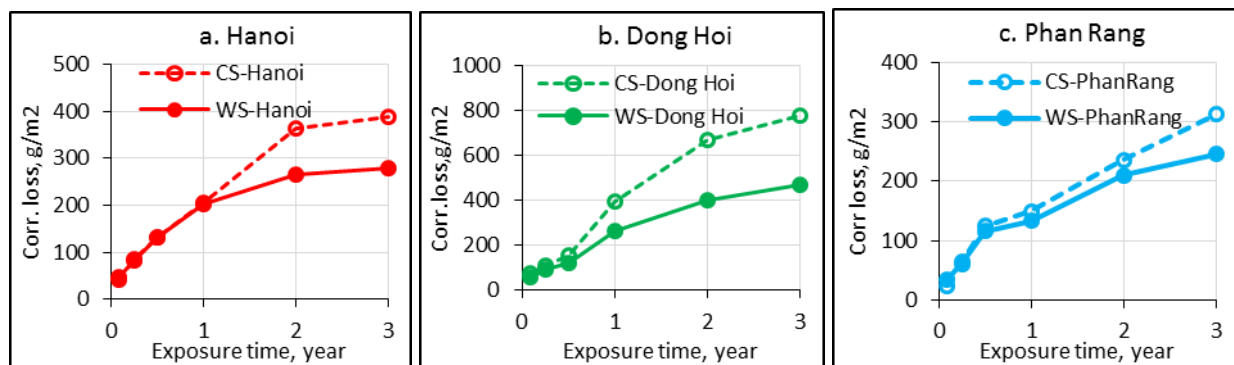


FIG 1. CORROSION LOSS OF WS AND CS AT: A) HANOI, B) DONG HOI, C) PHAN RANG

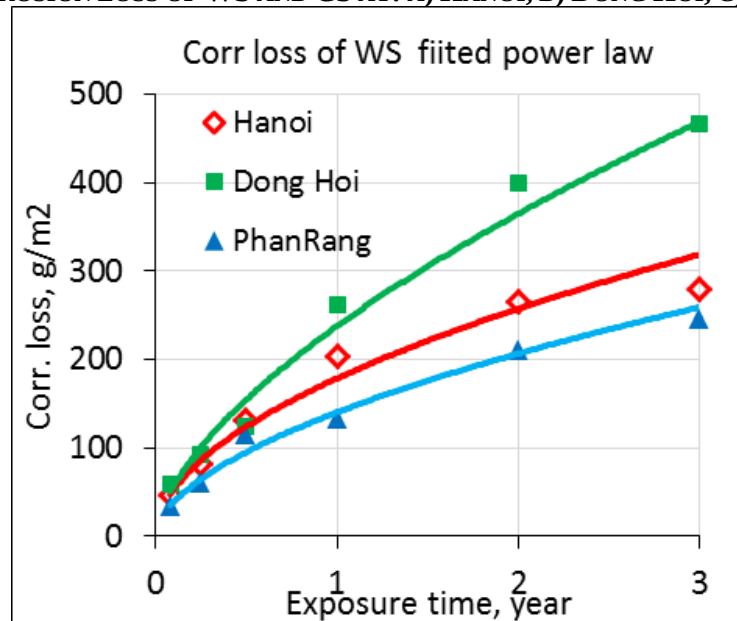


FIG. 2 CORROSION LOSS OF WS FITTED POWER LAW

TABLE 3
A AND N VALUES OF THE POWER LAWS

Sites	A	n	R ²
Hanoi	178.86	0.526	0.98
Dong Hoi	237.89	0.618	0.97
Phan Rang	140.52	0.557	0.98

Many authors [12-21] have founded that the evolution of atmospheric corrosion process follows power law (also called the logarithmic law) in the first stage of exposure (3-5 years), and for longer- term exposure, it fits better the power-linear model. In this study, the fitting data of CL depended on exposure time indicated that the atmospheric corrosion of WS at the above test sites obeys well the power law with high correlation $CL = At^n$ (here: t is exposure time, A and n are constants). The values of A and n are given in tab.3. The authors [12-13, 21] suggested the exponent n as an index of the physico-chemical of the corrosion product (or rust) layer, which can provides a criterion for gauging long term atmospheric corrosion susceptibility. It gives a measure of the resistance to transport processes within the corrosion product oxide once it has formed. When n is close to 0.5, it is the special case of an ideal diffusion-controlled mechanism when all corrosion product remains on the steel surface as the imperturbable layer. However, if the rust layer becomes more compact with time, then the diffusion coefficient decreases, and n -value will be lower than 0.5. On the contrary, if the diffusion process is accelerated by removing the rust layer, the exponent n will be higher than 0.5, attaining the limiting value of 1.0. Therefore the value of n depends both on the metal nature, the local atmosphere, the maximum exposure time, and the exposure condition (out-door, in-door, position of samples...). On the other hand, the parameter A provides the criterion for gauging short-term atmospheric corrosion susceptibility. It provides a measure of the inherent reactivity of a metal surface to produce a corrosion product layer in a short-term atmospheric exposure.

The n -values at Hanoi and Phan Rang are close to 0.5 (tab.3), as mentioned above, this indicated the diffusion-control of WS corrosion (which was confirmed by EIS results below). Conversely, the n value of 0.62 at Dong Hoi is typical of marine atmosphere where the airborne salinity content is relative high (18.7 mg Cl/m².day) and TOW is rather long (5705h/year). The evolution of WS corrosion at the test sites can be explained as follows:

The weather conditions at Dong Hoi (high RH, long TOW, great total of rainfall) produced the wet-period longer than dry-period (the ratio of drying-time/wetting-time is 0.54, see tab. 2), this accelerated the corrosion process, thus the rust was generated very fast in wetting period, however, this layer was slowly compacted due to very short drying-time. In addition, the high chloride deposition rate in Dong Hoi is an aggressive agent that makes the rust layer porous. According to many authors [3, 4, 22-26], chloride ion promotes the formation of β -FeOOH in rust, which is porous and unstable, consequently, the n -value is highest at Dong Hoi site.

Different from Dong Hoi, the climate at Hanoi is less humid with shorter TOW and less rain which created the similar duration of wetting time and drying time (with the ratio of 0.9), this supported both generation and improvement of rust layer. On the other hand, the negligible airborne salinity at Hanoi also favored the rust layer more compact; as a result, n -value at Hanoi is lower than that at Dong Hoi.

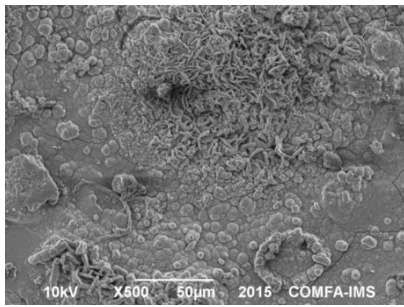
Meanwhile, the climate at Phan Rang is driest due to very little rain (the total of rainfall is 1130 h/y) and the highest time of sunshine which results in the drying-time/wetting-time ratio of 1.35. For these reasons, the corrosion loss at Phan Rang is lowest, however, n -value is a little higher than that in Hanoi, may be due to the influence of chloride ion.

It is clear from the above discussions that the evolution of protective rust layer on WS strongly depends on the ratio of drying time and wetting time. The airborne salinity at certain content showed the dominated influence on the corrosion and the protective ability of rust layer.

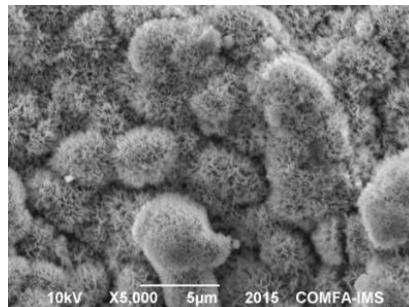
3.2 Characteristic of corrosion product layer on weathering steel

3.2.1 Morphology of rust surface

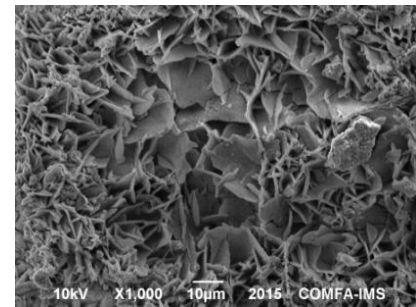
Fig.3 expresses SEM micrographs of the rusted WS samples those were exposed for six months at the test sites. As reported by the authors [27-28], the typical morphologies of lepidocrocite look like sandy and flowery, and typical morphology of goethite looks like cotton balls.



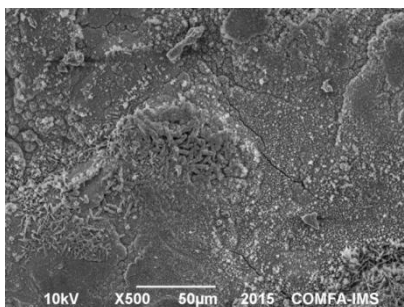
The over view



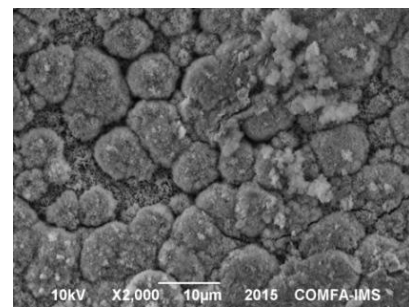
Cotton ball structure of goethite



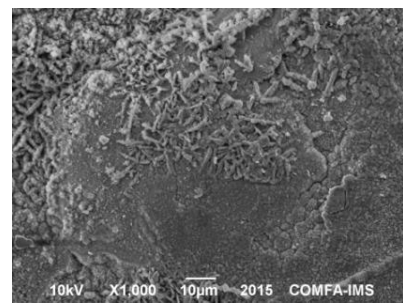
Flowery structure of lepidocrocite

FIGURE 3A. MORPHOLOGY OF RUST FORMED ON WS AFTER SIX MONTH-EXPOSURE IN HANOI

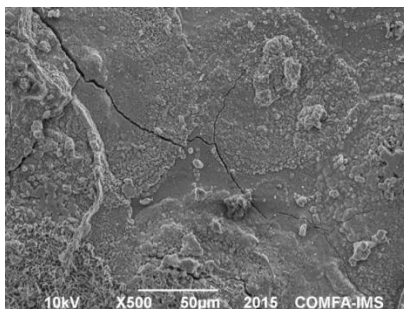
The over view



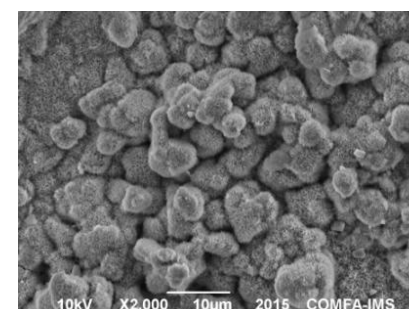
Cotton ball structure of goethite



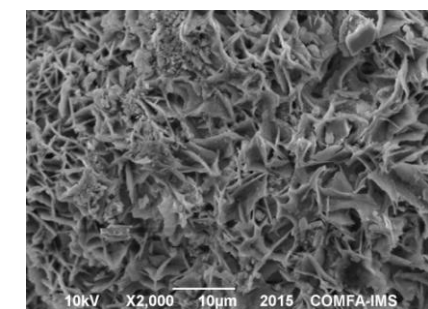
Flowery structure of lepidocrocite

FIGURE 3B. MORPHOLOGY OF RUST FORMED ON WS AFTER SIX MONTH-EXPOSURE IN DONG HOI

The over view



Cotton ball structure of goethite



Flowery structure of lepidocrocite

FIGURE 3C. MORPHOLOGY OF RUST FORMED ON WS AFTER SIX MONTH-EXPOSURE IN PHAN RANG

3.2.2 Cross-section structure of rust layer

The cross-section structures of rust layer formed on WS that was exposed for two years (fig.4) obviously include two layers: The inner layer looks dark grayish, dense and good adherent to steel substrate; And the porous outer layer is reddish and mottled. The rust layer formed at Phan Rang looks more compact in comparison with the other sites; meanwhile it is more porous at Dong Hoi. This can be explained by the different climatic conditions at these test sites (tab.1), thus long drying time, little rain and longtime of sunshine at Phan Rang favored the rust layer more compact, whereas long wetting time, great total of rainfall and shorter time of sunshine at Dong Hoi made the rust layer more porous. The rust layer at Hanoi looks more porous than that at Phan rang, but denser than that in Dong Hoi. The results of the rust cross-section morphology are confirmed the information reported by many authors [3, 12, 23-25, 29,] as well as suitable with the results of corrosion loss presented above.



FIGURE 4. THE CROSS-SECTION OF RUST LAYER FORMED ON WS EXPOSED FOR TWO YEARS AT TEST SITES

3.2.3 Phase composition of rust layer by X-ray analysis

TABLE 4
COMPOSITION OF CORROSION PRODUCT OF WS FORMED AT DIFFERENT EXPOSURE PERIOD

Exposure period	3 months					6 months					12 months				
Phases	α	β	γ	Mg	Mh	α	β	γ	Mg	Mh	α	β	γ	Mg	Mh
Hanoi	x	o	x	o	o	x	o	x	o	o	x	x	x	x	o
Dong Hoi	x	o	x	o	x	x	o	x	o	x	x	x	x	x	o
Phan Rang	o	x	x	o	x	x	x	x	o	x	x	x	x	x	o

Note: x-appeared, o-not appeared, α - α -FeOOH (goethite), β - β -FeOOH (akaganeite), γ - γ -FeOOH (lepidocrocite), Mg - Magnetite, Mh - Maghemite

The qualitative X-ray analysis results of corrosion product formed on WS after different exposure periods are given in tab.4 which showed the present of goethite (α -FeOOH), akaganeite (β -FeOOH), lepidocrocite (γ -FeOOH), maghemite and magnetite. These are main compounds of rust on WS [3-4, 21-26]. Goethite appeared early (for three month-exposure) in the rust formed at Hanoi and Dong Hoi, perhaps due to high humidity and long TOW which promoted the existence of goethite, whereas the dry climate with the long drying time at Phan Rang impeded its generation, accordingly, it was not detected by X-ray analysis. Akaganeite appeared in the rust at Phan Rang due to the impact of airborne salinity [3, 4], however it was not found in the rust at Dong Hoi where the chloride content is higher than that at Phan Rang. This can be explained by the great total of rainfall at Dong Hoi (2445 h/y) during the exposure time which washed the pollutants on the sample surface away and reduced the real content of chloride on the sample surface. During the early stage of exposure (till six months), magnetite was not founded at all sites, it is only detected in the rust formed after one year exposure, this time duration may be long enough for transformation of γ -FeOOH to α -FeOOH, then to magnetite. It is also possibility that X-ray analyses were done directly on the surface of three and six month-samples, while magnetite is located in the bottom of rust layer, thus it was difficult to detect. In fact, the rust formed for one year exposure was X-ray analyzed using powder, therefore the phases were discovered with higher accuracy.

3.3 Distribution of alloying elements in the rust

The line scan analyses using SEM-EDX were done on cross-sections of rust formed after two years exposure (as showed in fig. 5). The scan was performed several times for each sample, and the distribution of Cr and Cu content depending on distance from steel substrate are presented in fig. 6. It can be recognized that Cr element was concentrated in the rust at the distance of about 10-40 μ m, except at Dong Hoi, the dissolved Cr content in the rust was higher than that at other sites, and it distributed till 60 μ m far from steel base, perhaps the long wetting time at Dong Hoi promoted Cr more existence and faster diffusion. Contrary to Cr, Cu element was less dissolved, but faster diffused than Cr did – till 60 μ m distance. However, both of them were heterogeneously distributed. The distance of 40-60 μ m is similar to thickness of the inner layer showed in fig.4, as reported by many authors [3-4, 12, 30-34], Cr and Cu elements were strongly enriched in the inner compact layer of rust, Cr could replace Fe partly in α -FeOOH to form $\text{Fe}_x\text{Cr}_{x-1}\text{OOH}$ and attributed to protectiveness of WS. Meanwhile, Cu retards the growth of rust and suppresses the supply of oxygen to the steel surface; in addition, it reduces rust conductivity, retards the crystallization of rust as well as contributes to a uniform dissolution of steel and formation of the rust layer at the initial stage. Heidis Cano et al [30] studied characterization of corrosion products formed on WS exposed in marine atmospheres and found that Cr was distributed in the rust at the distance of 40-60 μ m from the steel substrate. The content of Cr dissolved in the rust is higher in comparison with content of Cu, may be because Cr content is higher Cu content in steel.

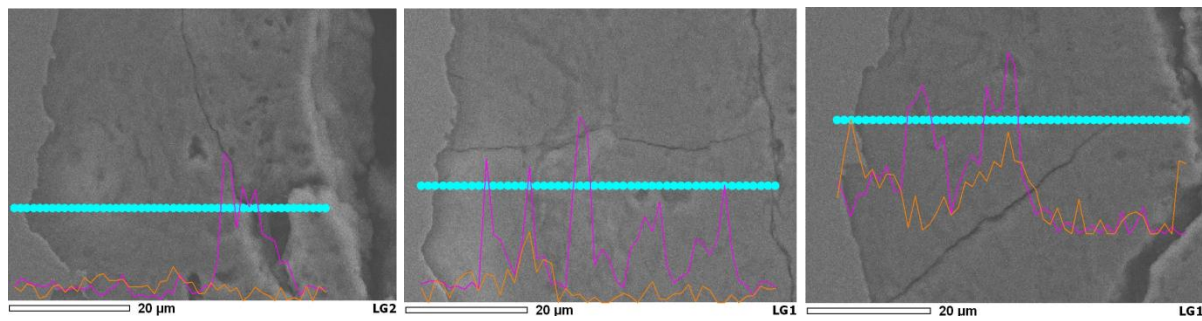


FIGURE 5. THE EDX LINE SCAN PICTURES OF RUST CROSS-SECTION FORMED FOR TWO YEARS-EXPOSURE

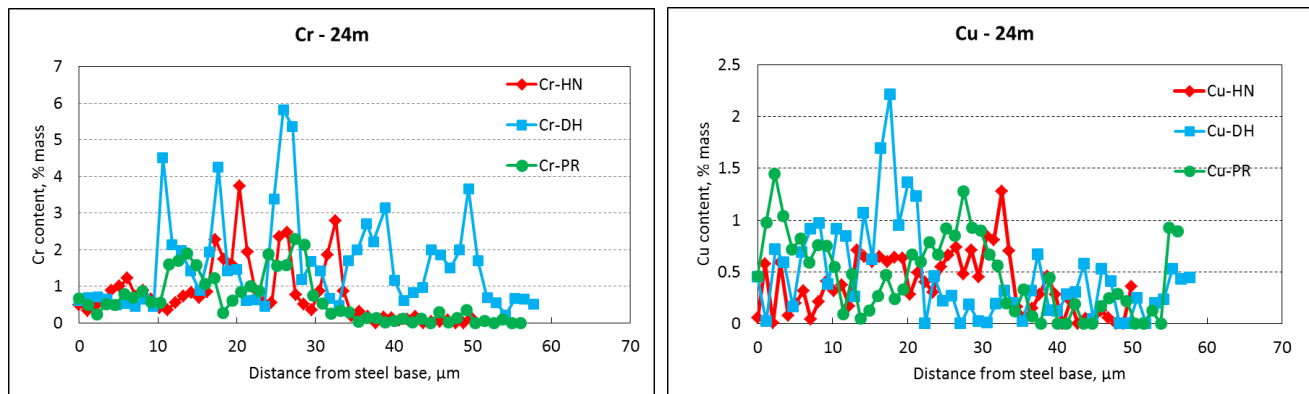


FIGURE 6. DISTRIBUTION OF Cr and Cu ELEMENTS IN RUST LAYER FORMED ON WS EXPOSED FOR 2 YEARS AT HANOI (HN), DONG HOI (DH) AND PHAN RANG (PR)

3.4 Electrochemical Impedance Spectroscopy

Fig.7 shows the Niquist diagram of WS exposed for three years at the test sites. The impedance diagrams are composed of the incomplete compressed semicircles at high frequency region and a diffusion tail at low frequency region. The equivalent electrical circuit (fig.8) is suitable to simulate the electrochemical of rusted WS [6, 12, 34-36]. Here, R_{sol} is resistance of solution, R_{rust} is resistance of the rust layer and C_{rust} , C_{dl} are capacitances of the rust layer and double layer, respectively. In addition, because the corrosion process of WS is controlled by diffusion of elements, the Faraday impedance is divided into two parts: the charge transfer resistance R_{ct} and Warburg impedance W . The resistance of rust is most useful for estimating protective properties of the rust layer.

The radius of high frequency semicircle increases in the order: Hanoi<Dong Hoi<Phan Rang (fig.6), as a result, the R_{rust} values obtained from fitted data also follow the same order (tab. 5). This indicates that for three years-exposure, the rust formed on WS exposure at Phan Rang atmosphere showed the best protective ability, may be the long drying time there predominated over other environmental parameters, irrespective of chloride presence, it promoted the rust dense and compact. Whereas, the rust layer formed on WS at Hanoi showed the lowest resistance and at Dong Hoi, R_{rust} value was in between. It is possibility that the high corrosion rate at Dong Hoi generated a thicker rust layer as barrier to protect steel base. Similarly, the thinner rust layer formed at Hanoi showed the lower resistance.

It can be seen from fig. 7 that at Phan Rang and Hanoi, the radius of high frequency semicircle of rusted WS is bigger than that of rusted CS, in contrary, at Dong Hoi, the semicircle of rusted WS shows the radius a little bit smaller than that of rusted CS, the fitted data of R_{rust} for CS are shown in tab. 5. As discussed above, for three years-exposure, WS obviously showed protectiveness predominant over CS did, except at Dong Hoi site, the rust of WS and CS showed similar resistance. Therefore, it can be suggested that the drying time is most important factor which determines protectiveness of WS in the atmosphere.

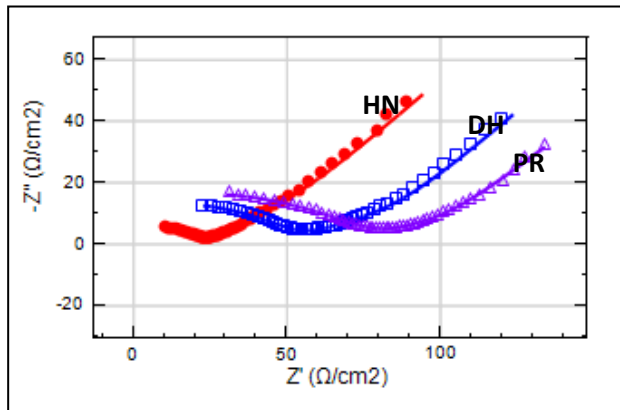


FIGURE 7. THE NIQUIST DIAGRAM OF WS exposed for three years at Hanoi (HN-●), Dong Hoi (DH-□) and Phan Rang (PR-Δ) which were measured in 0.1M NaCl. The bullets are measured data, and the lines are fitted data

TABLE 5
THE PARAMETERS EXTRAPOLATED FROM FITTED DATA

Rust resistance	Hanoi	Dong Hoi	Phan Rang
R_{rust} of WS, ohm.cm ²	44.2	88.2	118
R_{rust} of CS, ohm.cm ²	23.4	95	78

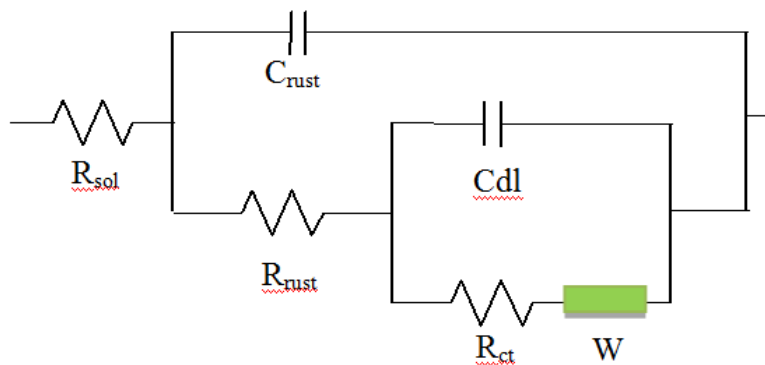


FIGURE 8. THE EQUIVALENT ELECTRICAL CIRCUIT OF RUSTED WS IN 0.1M NaCl SOLUTION

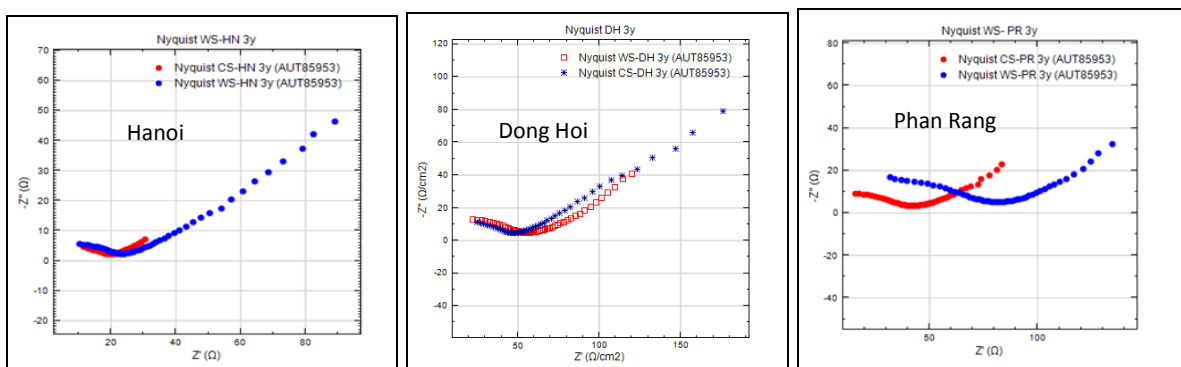


FIGURE 9. THE NIQUIST DIAGRAM OF WS AND CS EXPOSED FOR THREE YEARS AT HANOI (HN), DONG HOI (DH) AND PHAN RANG (PR) WHICH WERE MEASURED IN 0.1M NaCl.

IV. CONCLUSION

The weathering steel was exposed for three years at test sites of Vietnam. The results showed that:

- After one year-exposure, corrosion of weathering steel becomes lower than that of carbon steel. This showed the predominance of weathering steel over carbon steel in corrosion resistance. Corrosion loss of weathering steel increased in the order: Phan Rang < Hanoi < Dong Hoi and fitted well power law with high correlation.

- The morphology of WS surface exposed for six months showed sandy and flowery structure which is typical for lepidocrocite and cotton balls structure typical for goethite. The cross-section structures of rust formed on weathering steel exposed for two years obviously composed of two layers: the inner layer looks dark greyish, dense and good adherent to steel substrate; and the porous outer layer is reddish and mottled.
- X-ray analysis discovered the present of goethite (α -FeOOH), akaganeite (β -FeOOH), lepidocrocite (γ -FeOOH), maghemite and magnetite in corrosion product formed on weathering steel for 3, 6, and 12 months exposure. Cr and Cu were strongly enriched in the inner rust layer with distance of about 40-60 μ m from the steel substrate.
- The EIS measurements of rusted weathering steel which was exposed for three years indicated that the rust resistances (R_{rust}) increased in the order: Hanoi < Dong Hoi < Phan Rang. At Phan Rang and Hanoi, the resistances of rust formed on weathering steel was higher than that formed on carbon steel; Meanwhile, at Dong Hoi the rust of weathering steel and carbon steel showed similar resistance.
- It can be suggested that the drying time is most important to produce the rust layer with good resistance.

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