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The influence of rare earth elements lanthanum on corrosion resistance of steel plate for offshore platform

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Abstract

Two kinds of offshore platform steels without rare earth elements and with trace rare earth elements lanthanum (less than 20ppm) were selected as the research objects. The corrosion of the two groups of steels was simulated by salt spray corrosion chamber. The macro/micro corrosion morphology of the two groups of steels was studied by scanning electron microscope; The periodic immersion corrosion experiment was used to accelerate the corrosion, the weight loss method was used to study the corrosion kinetics, and the morphology of corrosion rust layer was observed by scanning electron microscope; The composition and content of corrosion products were analyzed by XRD; The electrochemical behavior of the steel after corrosion was analyzed by electrochemical polarization curve and Nyquist diagram. The results show that the addition of trace rare earth elements lanthanum (less than 20ppm) can reduce the corrosion rate and corrosion current of two kinds of offshore platform steel with different strength, and the corrosion layer is more compact, which is a corrosion product with protective performance FeOOH increased. The addition of trace rare earth elements lanthanum (less than 20ppm) improves the corrosion resistance of two kinds of offshore platform steels with different strength.

Rare earth has the function of purifying the liquid steel, refining the grain, improving the strength and corrosion resistance of the material. In the early years, most scholars systematically studied the effect of rare earth elements lanthanum on the corrosion resistance of test steel from the aspects of rust layer structure and inclusion modification by means of field atmospheric exposure, laboratory accelerated corrosion and electrochemistry [1–3]. According to the research of MI Fengyi et al [4], in the marine atmospheric environment, steel with rare earth content above 430ppm has fewer rust layer defects, which can effectively prevent the invasion of corrosive examples in the environment, and rare earth can promote the conversion of active components in corrosion products into α-FeOOH comprehensively improves the marine corrosion resistance of the test steels. Yue Lijie et al [5] studied the effect of 0–210ppmr on the corrosion resistance of weathering steel and found that rare earth elements can inhibit long MnS inclusions, form spheroidal rare earth sulfur oxides and rare earth sulfides, reduce the size of inclusions in steel and improve the corrosion resistance of weathering steel. However, the rare earth content in the steel studied at present is generally large (40–200ppm) [6–11], and more rare earth content will form large particles of rare earth inclusions, which block the casting nozzle in the production of continuous casting process, which seriously affects the production efficiency. The addition of ultra-micro (less than 20ppm) rare earth in steel can effectively avoid ‘nodule’ phenomenon, and it is suitable for continuous casting process of steel for offshore platform, which is conducive to the improvement of production efficiency in the factory. The ultra-micro rare earth elements can replace some corrosion-resistant precious metal elements, reduce the alloy
cost, improve the corrosion resistance of steel used in offshore platform to some extent, reduce the self-weight of steel used for offshore platform and improve its load capacity. Combined with the actual production of continuous casting process, the influence of trace (less than 20ppm) rare earth elements lanthanum on the corrosion performance of offshore platform steel is studied by means of salt spray corrosion, weekly immersion corrosion, electrochemical corrosion, XRD and SEM. It provides some reference for the full use of rare earth resources and the development of new type of rare earth offshore platform steel.

1. Experimental materials and methods

1.1. Experimental materials
EH690 and EH36 high strength offshore platform steels produced by a steel plant are used as experimental materials. Rare earth elements lanthanum less than 20ppm are added into the two groups of experimental steels as comparative experimental steels. Table 1 shows the main chemical composition of the experimental steels.

The four kinds of test steel are made of 210t top bottom combined blowing converter and LF + RH refining process and are cast into 230 × 1800 mm continuous casting billet, the first group of ultra-high strength test steels A and B are hot rolled and tempered. The second group of high strength test steel e, f hot rolled after the heat treatment.

1.2. Experimental methods
The samples were polished, and the grain boundaries were corroded by picric acid solution (30 ml distilled water + 2 g picric acid + 1.5 g Haiou Shampoo + 6 drops of hydrogen peroxide). The metallographic photos were taken by Olympus inverted system metallographic microscope, and the average grain size was determined by IPP software. According to the requirements of GB/T10125-2012 neutral salt spray. The test steel was processed into a sample by molybdenum wire cutting machine, and the sample size was 30 × 40 × 2 mm. The surface of the sample shall be polished evenly with sandpaper. The four test steels A, B, E and F were placed in the salt spray corrosion box of IRHS-270-RY, and the salt spray corrosion was carried out in the cycles of 48 h, 96 h, 144 h, 192 h and 240 h; The macro morphology changes of the test steel before and after corrosion were photographed by digital camera under different corrosion cycles; The rust removal solution (3.5 g hexamethylenetetramine, 500 ml hydrochloric acid and 1000 ml distilled water) was used to soak for 10 min. The corrosion products on the surface of the corrosion sample were removed with clean water, and the macro morphology after rust removal was photographed by digital camera. SEM was used to observe the micro morphology of salt spray corrosion; The corrosion products on the surface of the samples under different corrosion cycles were gently scraped off with a knife, and the composition and content of the rust layer were measured by XRD. The specimen is cut into a thin sheet of size 10 × 10 × 3 mm in each group were placed in a beaker with 3.5% NaCl aqueous solution. The beaker was placed in a constant temperature water bath at 25 °C, and the soaking period was set as 5d, 10d, 20d, 30d, 45d and 240d. SEM was used to observe the cross-section morphology of the rust layer of the test steel after 45 days and 240 days of immersion corrosion. The sample was cut into 10 parts by molybdenum wire cutting machine which size was 10 × 10 × 3 mm. The polarization curve and Nyquist diagram were measured by Zennium electrochemical workstation. The three-electrode system was used in the test system. The working electrode was the sample with corrosion period of 6d, 12d and 18d. The reference electrode was saturated calomel electrode, and the auxiliary electrode was platinum electrode. The excitation signal of AC impedance was a sine wave with amplitude of 50 mv. The measurement frequency range was ~800 mv ~ 400 mv. The effect of trace rare earth elements lanthanum on corrosion resistance of offshore platform steel was analyzed.

2. Experimental results and analysis

2.1. Morphology of original austenite
Figure 1 shows the original austenite metallographic photos of two groups of test steels with different strength under quenching and tempering condition. According to the requirements of GB/T 6394-2017, The grain size of the first group of ultra-high strength steel A and B was measured by IPP software. The average grain size of the test steel A with 9 ppm rare earth element was 13.83 μm. The average grain size of blank test steel B corresponding to a is 15.37 μm.
Figure 1. Original austenite of A, B, E and F samples (a) 9ppmRE (b) 0RE (E) 10.2ppmRE (f) 0RE.

About 43.66% of the grain size of A test steel containing 9ppmRE is 10–15 μm, about 11.26% of the grains are larger than 20 μm. The grain size of blank test steel B is about 32.47%, which is distributed in 10–15 μm, 51% of the grains are larger than 20 μm. In the second group of high strength steels E and F, the average grain size of E test steel with 10.2 ppm RE is 11.30 μm. The average grain size of the corresponding blank test steel F is 13.99 μm. About 45.39% of the E grain size of the experimental steel with trace rare earth is distributed in the range of 10 ~ 15 μm, 65% of the grains are larger than 20 μm. About 30.57% of the F grain size is distributed in the range of 10 ~ 15 μm. About 17.19% are larger than 20 μm [12, 13]. After adding trace rare earth elements lanthanum, the original austenite grains of two kinds of offshore platform steels with different strength are refined, the large-size grains are reduced, and the grain distribution is more uniform. The research shows that the corrosion uniformity of the test steel is better after grain refinement, and the probability of forming deeper cracks and voids is smaller, which can greatly improve the compactness of the rust layer, to improve its corrosion resistance [14].

2.2. Salt spray corrosion

Figure 2 macro morphology of salt spray corrosion of A, B, E and F test steels in the corrosion cycle of 48 h, 96 h, 144 h, 192 h and 240 h. It can be seen from the figure that the corrosion products of the first group of ultra-high strength steels A and B at the initial stage of corrosion are brown, dark brown and a small amount of black corrosion products appeared in the later stage of corrosion, and large corrosion pits appeared on the surface of the substrate after 240 h of corrosion. The color change of rust layer is due to the change of free iron ions into ferrous hydroxide, ferrous oxide and hydroxyl oxide of iron at first, and the formation of stable ferric oxide and ferric oxide at the later stage of corrosion, α- Corrosion products of FeOOH et al [15].

It can be seen from the figure that the corrosion period is 48 h and 96 h, and the rust layer is loose, which provides a transmission channel for water, oxygen and other corrosive media, so that the initial corrosion rate is higher. After 144 h, a dense rust layer is formed on the surface of the test steel, which effectively protects the matrix and reduces the corrosion rate. The corrosion products of the second group of high strength steels E and F are basically the same as those of A and B. To further study the effect of trace rare earth on the corrosion resistance of offshore platform steel, the test steel under different corrosion cycles was deducted with degusting solution. The macro morphology after degusting is shown in figure 3.

It can be seen from the figure that at the initial stage of corrosion, corrosion lines of different depths appeared on the surface of the substrate. With the extension of corrosion time, corrosion pits with more serious corrosion degree gradually appeared. It can be seen from the figure that compared with the corresponding blank test steel, the corrosion lines of a and e steels added with trace rare earth elements lanthanum are relatively shallow, and the corrosion pits are also less. According to the measurement with Image Pro Plus software, the average area of corrosion pits in E test steel containing rare earth element lanthanum is 11.2 μm², the average area of corrosion pit in F test steel is 44.3 μm², after adding lanthanum into low carbon steel, the average area of corrosion pit on the surface of test steel is reduced by about 74% compared with blank test steel. Lanthanum can effectively improve the corrosion resistance of low carbon steel. Figure 4 shows the micro morphology of two kinds of offshore platform steels with different strength after 240 h salt spray corrosion. It can be seen from the figure that the corrosion pits of test steel a with trace rare earth elements lanthanum are more uniform and denser than that of test steel B; The corrosion pit size of test steel E with trace rare earth elements lanthanum is obviously smaller than that of blank test steel F. The research shows that lanthanum can make the inclusions in steel evenly distributed, and the evenly distributed inclusions can be used as active dissolution points to promote uniform corrosion distribution, to effectively improve the corrosion resistance of steel [16]. FeOOH, Fe₃O₄ and Fe₂O₃ are ionic crystals. When the
stress accumulation in the crystal reaches a certain degree, the energy is usually released in the form of microcracks. The more microcracks on the material surface, the larger the corrosion area, and the worse the corrosion resistance of the material. The ion radius of lanthanum is large and entering the rust crystal will lead to lattice distortion, increase the internal energy of the material, increase the micro stress, hinder the dislocation slip deformation, effectively prevent the generation of cracks and effectively improve the corrosion resistance of the material.

Combined with the macro and micro morphology of the test steel after salt spray corrosion, the addition of trace rare earth elements lanthanum makes the corrosion pit of the steel for offshore platform more uniform and the rust layer more compact, which prevents the corrosive substances from invading the matrix and reduces the size of the corrosion pit. Because of the large radius of rare earth ions, lattice distortion and lattice stress will be caused when they enter the rust crystal.

Figure 2. Macro morphology of salt spray corrosion of A, B, E and F test steels with corrosion cycles of 48 h, 96 h, 144 h, 192 h and 240 h.
Figure 5 shows the microstructure of two groups of steel for different strength offshore platforms. The microstructure of A and B of the first group of ultra-high strength steel is tempered Soxhlet, and the second group of high strength steel E and F are ferrite and pearlite. The microstructure of super high strength steel A and B is tempered Soxhlet, and the potential difference is small, and the corrosion effect of steel is small. Ferrite and pearlite in high strength steel E and F are two kinds of structures, with large potential difference. Different types of metallographic structure are easy to form corrosion micro cells, which makes the tendency of anode dissolution larger. The corrosion rate between the two phases is different, which makes the growth rate of rust on the steel surface inconsistent in each part. The rust layer produces a large internal stress during the growth process, and the stress accumulation to a certain extent will produce micro cracks in the rust layer, release energy, and the formed penetrating cracks will destroy the

Figure 3. Macro morphology of A, B, E and F test steels after salt spray corrosion and rust removal with corrosion cycles of 48 h, 96 h, 144 h, 192 h and 240 h.
density of the rust layer, and lead to a larger corrosion rate [17]. After adding rare earth elements lanthanum, the microstructure of the two groups of steel is refined, which makes the steel get better corrosion uniformity, and is conducive to forming a dense rust layer, thus improving the corrosion resistance of the steel.

Figure 6 is the salt spray corrosion rate diagram of the second group of high strength steel E and F. It can be seen from the diagram that with the increase of corrosion time, the corrosion rates of E and F test steel show a trend of first decreasing and then increasing, and the corrosion rates of E sample steel with 10.2ppmRE addition in different corrosion cycles are less than that of F test steel. With the increase of corrosion time, the surface of the substrate is gradually covered with a layer of corrosion products, which can prevent Cl\(^{-}\) from continuing to invade the substrate, intensify the corrosion, and gradually reduce the corrosion rate. In the later stage of
corrosion, the corrosion products on the surface of the substrate will peel off to a certain extent, which will reduce the protection of the substrate and increase the corrosion rate again. The concentration of Cl\(^-\) in the solution decreases after reacting with the matrix in the early stage of corrosion, so the corrosion rate in the late stage of corrosion is lower than that in the early stage of corrosion.

In order to further analyze the influence of trace rare earth elements lanthanum on the corrosion resistance of offshore platform steel, the rust layer of high strength test steel E and F after 192 h salt spray corrosion was selected for XRD corrosion product analysis, as shown in figure 7. It can be seen from the figure that the corrosion products of the two kinds of test steels are all corrosion products \(\alpha\)-FeOOH, \(\gamma\)-FeOOH, Fe\(_3\)O\(_4\), Fe\(_2\)O\(_3\). Table 2 shows the phase ratio of rust layer of test steel. It can be seen from the table that when 10.2 ppm RE is added to test steel E, The content of \(\alpha\)-FeOOH is higher. Studies by other scholars have shown that among various products of atmospheric corrosion of steel, \(\gamma\)-FeOOH can be used as depolarizer in the process of cathode with rusty electrode, and it can be transformed into stable one through dissolution crystallization.
process with the extension of corrosion time \( \alpha \)-FeOOH [18]. \( \alpha \)-FeOOH can inhibit the corrosion of steel. The more content of \( \alpha \)-FeOOH, the better the protective property of the matrix. The protection of rust layer can be determined by parameters \( \alpha/\gamma^* \). The larger the parameter is, the stronger the protection of rust layer is [19]. The total content of \( \gamma \)-FeOOH, Fe\(_2\)O\(_3\) and Fe\(_3\)O\(_4\) is determined as \( \gamma^* \), E test steel containing 10.2ppmRE, \( \alpha/\gamma^* \) 193; F test steel \( \alpha/\gamma^* \) 159. The results show that the addition of trace rare earth makes the rust layer protection of the test steel better than that of the blank test steel, and it can be inferred that rare earth can promote the active components in the rust to stabilize \( \alpha \)-FeOOH phase transformation, change the phase composition of rust layer, improve the protection performance of offshore platform steel rust layer [4].

Figure 8 shows the corrosion rate of E test steel with 10.2ppmRE and blank test steel F after cycle immersion corrosion. It can be seen from the figure that the corrosion rate of E test steel added with trace rare earth elements lanthanum is lower than that of F test steel in all set cycles. With the increase of corrosion cycle, the corrosion rate first decreases and then increases, which is the same as that of salt spray corrosion.

To further study the effect of trace rare earth elements lanthanum on the corrosion resistance of offshore platform steel, E and F test steels with corrosion cycle of 45 days were selected, and the corrosion section morphology was observed by scanning electron microscope, as shown in figure 9. It can be seen from the figure that the corrosion section of E test steel is more uniform, and the corrosion degree of F sample steel is more serious, which goes deep into the matrix and intensifies.

To comprehensively analyze the corrosion resistance, E and F test steels with a corrosion cycle of 240 days were selected to observe the corrosion section morphology, as shown in figure 10. It can be seen from the figure

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**Figure 8.** Corrosion rate of E (10.2ppmRE) and F test steel.

**Figure 9.** Corrosion section morphology of test steel E (10.2ppmRE) and F after 45d immersion corrosion.
that under long-term corrosion, the corrosion rust layer of E test steel containing trace rare earth elements lanthanum is shallower than that of F test steel, and the depth of rust layer measured at the deepest corrosion place of e sample steel is 3.39 μm. The depth of the deepest rust layer in F test steel is 3.76 μm. The addition of trace rare earth can still improve the corrosion resistance of offshore platform steel under long-term corrosion.

2.3. Electrochemical corrosion

Figure 11 shows the polarization curves of E and F test steels at 6d, 12d and 18d. It can be seen from the figure that the change trend of cathode and anode of E and F test steels is basically the same. The self-corrosion potential of E test...
steel added with trace rare earth elements lanthanum is higher than that of F test steel. Rare earth improves the thermodynamic stability of rusty samples in corrosive medium and reduces the tendency of material to undergo anodic dissolution \[18\]. Under the same polarization voltage, the polarization curve of E sample steel with trace rare earth elements lanthanum is closer to the left end than that of F sample steel, and the corrosion current density is lower, which indicates that the addition of rare earth elements lanthanum can slow down the anodic dissolution of steel matrix during the corrosion period. In terms of corrosion kinetics, the corrosion current density in polarization curve represents the corrosion rate; The lower the self-corrosion current, the better the corrosion resistance \[20\].

Figure 12 shows Nyquist diagram of E and F sample steel at 6d, 12d and 18d. Electrochemical impedance method is an effective experimental method to study corrosion mechanism of metal materials and analyze resistance of rust layer on corrosion surface. It can be seen from figure 12 that Nyquist spectrum of rust test steel consists of two resistance arcs located in high frequency and medium-low frequency areas. The resistance arc in high frequency region is compressed and deformed, which is caused by the charging and discharging process of rust layer on the corrosion surface. By analyzing Nyquist diagram, the strength and weakness of the rust layer material on the matrix protection can be compared. The larger the curve radian in the drawing, the stronger the corrosion resistance of the corresponding test steel; The shape of the resistance arc in low frequency region is caused by the change of the concentration of the corrosion liquid by the charge transmission, and the change of concentration difference and diffusion causes the change of electrode reaction. The surface of the steel is not smooth after corrosion, which causes the electric field distribution in the electrode solution interface is uneven, which leads to the difference between the capacitance resistance arc and the ideal arc shape in the high frequency region \[21\]. It can be seen from the figure that the curve arc of E test steel added with trace rare earth elements lanthanum is larger, which indicates that the corrosion resistance of E-sample steel is stronger.

3. Conclusion

(1) The addition of trace rare earth elements lanthanum can refine the original austenite structure of offshore platform steel, make the structure more uniform and reduce the number of large-size grains.
(2) The addition of trace rare earth elements lanthanum can effectively reduce the corrosion rate of offshore platform steel, and promote the protection of corrosion products. The production of α-FeOOH; It makes the rust layer of the test steel more compact, prevents Cl\(^{-}\) from invading continuously, and effectively protects the matrix.

(3) Trace rare earth elements lanthanum make the self corrosion potential of offshore platform steel move forward, the anode current density decreases, and the Nyquist diagram curve increases, which effectively improves the corrosion resistance.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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