Crevice Corrosion of Several Supper Stainless Steels in the Simulated LT-MED Environment

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Abstract

Susceptibility and morphological characteristics of crevice corrosion for SS316, SS904L, SS254sMo and SS2507 in the simulated low-temperature multi-effect distillation environment were investigated by cyclic polarization test, scanning electron microscope and laser microscope. The results show that the crevice corrosion resistance of four kinds of stainless steel is ranked as SS254sMo $\approx$ SS2507 > SS316 > SS904L. There are “cover” structures over the edge of active crevice corrosion regions of SS904L, SS254sMo and SS2507, but SS316 is an exception. Galvanic corrosion characteristics appeared in the crevice of duplex supper stainless steel SS2507.

Keywords Supper austenite stainless steel · Super duplex stainless steel · Low-temperature multi-effect distillation (LT-MED) · Crevice corrosion · “Cover” structure

1 Introduction

Due to the uneven distribution of fresh water resources, many countries and regions all over the world are experiencing water shortage. As a technology to increase fresh water supply, desalination has become an important method to solve the crisis of water resource shortage [1, 2]. Several technologies such as low-temperature multi-effect distillation (LT-MED), multistage flash distillation and reverse osmosis are used in the desalination process [3–6]. Among these, LT-MED is one of the most efficient thermal desalination technologies, which run at the temperature below 75 °C and produce a lot of high concentrated seawater [2]. In such a harsh corrosive environment, materials used for LT-MED equipment often suffer from serious corrosion damage.

Supper stainless steels are being increasingly used as structural materials in desalination facilities, due to their high resistance to local corrosion and stress corrosion

cracking as well as their high strength and toughness [7, 8]. The high corrosion resistance of supper stainless steels comes from their high alloy contents, such as chromium, molybdenum and nitrogen, which can stabilize the passive film of stainless steels [9]. Based on the contribution of the three kinds of alloying elements, pitting resistance equivalent number (PREN) is used as a criterion to evaluate the stability properties of passive films, as shown in Formula (1) [9–11]. Normally, supper stainless steels have high values of PREN, and some of them can even exceed 40 (SS254sMo and SS2507, etc.).

$$\text{PREN} = 1 \text{ Cr}\% + 3.3 \text{ Mo}\% + 30 \text{ N}\%. \quad (1)$$

Austenite and duplex supper stainless steels are two main types of supper stainless steels. Austenite supper stainless steels (SS904L and SS254sMo, etc.) are made up of austenite phase. Their corrosion behaviors are similar to SS316, while the corrosion resistances are much higher than it [12]. Duplex supper stainless steels (SS2507, etc.) composed of ferrite (α) and austenite (γ) phase offer high strength and corrosion resistance, particularly pitting and crevice corrosion [13, 14]. In recent years, duplex supper stainless steels, characterized by their lower nickel contents, have attracted a lot of attention.

Currently, there have been a number of studies on the application evaluation of supper stainless steel to

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desalination equipment. Oldfield and Todd reported that, compared with ordinary stainless steel (SS316 and SS317, etc.), super stainless steel (SS31254, etc.) was more able to meet the requirements of seawater reverse osmosis equipment for corrosion resistance [15]. However, in the case of screws and flanges, crevice corrosion is still impossible to avoid at the interface between the tightly contacted plate and the screw or flanges made up of super stainless steel. For example, Wallen et al. found that flanges made of SS254sMo suffer severe crevice corrosion in continuously chlorinated seawater at 45 °C, while the longitudinally welded pipes made of SS254sMo did not suffer any corrosion [16]. Mahk et al. [16] considered that, in seawater reverse osmosis equipment, SS2507 and SS254sMo have high resistance to crevice corrosion.

The PREN indexes of SS2507 and SS254sMo are significantly higher than that of SS904L, while the cost of SS904L was lower than those of the former two. Therefore, compared with SS2507 and SS254sMo, there are more promising applications in LT-MED equipment for SS904L. However, the application evaluation of the crevice corrosion resistance for SS904L, SS2507 and SS254sMo in LT-MED equipment has not been reported.

SS904L, SS254sMo and SS2507 are the main candidates for LT-MED equipment, while there are still some crevice corrosion problems and they need to be clarified. For example, there is obvious difference between austenite (SS904L and SS254sMo, etc.) and duplex (SS2507, etc.) super stainless steel in phase composition. Whether the crevice corrosion characteristics will be affected by the difference between the two phases and whether this phenomenon will limit their use in LT-MED equipment are still lack of knowledge. Moreover, the PREN of SS904L is greater than ordinary stainless steel (SS316), while its cost is obviously lower than that the super stainless steel with high PREN (SS254sMo, etc.). Therefore, there are more promising applications in LT-MED equipment for SS904L. However, the evaluation on crevice corrosion for SS904L using for LT-MED equipment has not been carried out.

In view of the above problems, the aim of this work is to examine the crevice corrosion sensitivity of SS904L, SS254sMo and SS2507 and meanwhile assess their suitability for LT-MED equipment. In addition, this work is to reveal the crevice corrosion morphological characteristics of austenite (SS904L and SS254sMo, etc.) and duplex (SS2507, etc.) super stainless steel.

2 Experimental

The chemical compositions and the metallographic morphologies of SS316, SS904L, SS254sMo and SS2507 used in this research are given in Table 1 and Fig. 1. SS316,

SS904L and SS254sMo are austenitic stainless steel consisting of single γ -phase, while SS2507 is duplex stainless steel composed of α -phase and γ -phase. A multiple crevice assembly based on the standard guide for crevice corrosion testing, ASTM G78 [17, 18], was used. The modified crevice formers as shown in Fig. 2a were made up of polyethylene. There are 20 of plateaus on every multiple crevice assembly, and the design details of multiple crevice assembly can be seen from Fig. 2a. The stainless steel specimens (30 mm $\times$ 30 mm $\times$ 6 mm with a 7-mm-diameter hole) were cut from commercial plates (Fig. 2b). Electrical connection to the specimens was established via an isolated Cu wire, and epoxy resin was used for insulation treatment of welding points. Before starting the crevice corrosion test, all the tested specimens were abraded with wet 2000 grit SiC paper, degreased with acetone followed by alcohol and then dried in air. The specimen assembly was tightened with a titanium bolt, nut and washer using a torque wrench with the applied torque of 0.1 Nm [17, 18], so that 20 small crevice sites were formed on each side of the specimen (Fig. 2c). PTFE tape was used to insulate the bolt to prevent electrical connection between the titanium bolt and the stainless steel specimen.

The crevice corrosion test was performed in two times concentrated artificial sea water, and the detailed compositions of the solution are given in Table 2 [17, 19]. The test temperature is 70 °C.

The standard three-electrode cyclic potentiodynamic polarization method was employed according to ASTM G61 [17, 20]. The test cell was composed of a reference electrode (saturated calomel electrode), a platinum sheet as the electrode (20 mm $\times$ 20 mm) and a working electrode (stainless steel specimen assembly), as shown in Fig. 3. Prior to carrying out the cyclic potentiodynamic polarization test, the specimens were kept in solution for 30 min. The scan was initiated from 100 mV below the open-circuit potential (OCP) forward to 400 mV_{SCE} and then backward to 100 mV below OCP. The sweep rate was controlled at 0.166 mV/s.

All the electrochemical measurements were taken by using a Gamry 600 electrochemical workstation. The corrosion depth profile of the crevice corrosion site was measured using a LEXT laser microscope OLS 4000. The corrosion morphology was observed using a Phillips FEI INSPECTF scanning electronic microscope (SEM).

3 Results and Discussion

Figure 4 shows the cyclic potentiodynamic polarization curves of SS316, SS904L, SS254sMo and SS2507 crevice assembly specimens in the two times concentrated artificial seawater at 70 °C. In the cyclic potentiodynamic

Table 1 Chemical compositions of SS316, SS904L, SS254sMo and SS2507 stainless steels (wt%)

	C	Cr	Ni	Mn	Si	P	S	Mo	N	Cu
SS316	0.05	16.9	10.6	1.6	0.5	0.031	0.006	2.05	–	–
SS904L	0.02	20.1	25.3	2.00	1.00	0.045	0.035	4.04	–	1.32
SS254sMo	0.02	19.7	17.8	1.0	0.8	0.03	0.01	6.2	0.19	0.75
SS2507	0.03	25.4	7.01	1.00	0.80	0.035	0.020	4.2	0.27	0.50

Fig. 1 Metallographic morphologies of **a** SS316, **b** SS904L, **c** SS254sMo, **d** SS2507 stainless steels

polarization curves, three important corrosion parameters which are, respectively, called as corrosion potential (E_{corr}), breakdown potential (E_b) and repassivation potential (E_{rp}) can be seen. The values of E_{corr} , E_b and E_{rp} for SS316, SS904L, SS254sMo and SS2507 stainless steels are shown in Table 3. The measured E_{corr} values are in the following sequence: SS316 ($-325 \text{ mV}_{\text{SCE}}$) < SS904L ($-307 \text{ mV}_{\text{SCE}}$) < SS254sMo ($-231 \text{ mV}_{\text{SCE}}$) < SS2507 ($-213 \text{ mV}_{\text{SCE}}$), respectively. When the scanning potential reaches the breakdown potential (E_b), the passive film breaks down and localized corrosion occurs on the stainless steel surface. The more negative the breakdown potential (E_b) is, the easier for the initiation of localized corrosion (pitting or crevice corrosion) occurs [20]. Normally, this is attributed to the easier initiation for crevice corrosion than

pitting corrosion, and the breakdown potential (E_b) of crevice corrosion is more negative than that one [21–23]. Therefore, the value of E_b is considered as the breakdown potential of crevice corrosion in this study. The measured E_b values of SS316, SS904L, SS254sMo and SS2507 crevice corrosion specimens are, respectively, 86, 114, 175 and 178 mV_{SCE} , which mean that the susceptibility of crevice corrosion can be sequenced as follows: SS316 > SS904L > SS254sMo $\approx$ SS2507. In addition, the reverse scanning curve intersects the forward scan curve at one point, and the potential of this point is called repassivation potential (E_{rp}) [24]. The more negative of this potential is, the more difficult to repassivate of the steel is [20]. The measured E_{rp} values of SS316, SS904L, SS254sMo and SS2507 crevice corrosion specimens are, respectively,

Fig. 2 Multiple crevice assembly with the specimen [13, 14]: **a** dimensions of crevice former; **b** dimensions of test specimen; **c** specimen assembly

Table 2 Compositions of the two times concentrated artificial seawater (g/L)

NaCl	MgCl ₂	Na ₂ SO ₄	CaCl ₂	NaHCO ₃	KBr	H ₃ BO ₃
49.06	10.4	8.18	2.32	0.402	0.202	0.054

– 373, – 242, – 189 and – 165 mV_{SCE}. It is necessary to concern that the value of E_{tp} is more negative than E_{corr} for SS316, which means that SS316 occurs to crevice corrosion more easier [21, 25]. The value of E_{tp} is more positive than E_{corr} for the other three kinds of stainless steels. The current densities on the polarization curve at 400 mV_{SCE} are in the following sequence: SS254sMo (4.8 mA) <

Fig. 3 Schematic diagram of the three-electrode test cell

Fig. 4 Cyclic potentiodynamic polarization curves of **a** SS316, **b** SS904L, **c** SS254sMo, **d** SS2507 stainless steel crevice corrosion specimen in two times concentrated artificial seawater at 70 °C

Table 3 Parameter values of cyclic polarization curves of SS316, SS904L, SS254sMo and SS2507 stainless steels (mV_{SCE})

	E_{corr}	E_b	E_{rp}
SS316	– 325	86	– 373
SS904L	– 307	114	– 242
SS254sMo	– 231	175	– 189
SS2507	– 213	178	– 165

SS2507 (2.9 mA) < SS904L (195.5 mA) < SS316 (521.9 mA), respectively, which mean that the corrosion damage to the surface of stainless steel is in the same order.

Figure 5 shows macromorphology of SS904L artificial crevice corrosion specimen and the depth measurement by laser microscope. Each artificial crevice specimen has 40 crevice sites, while not every crevice site was attacked during cyclic polarization testing. The number of crevice sites attacked by crevice corrosion reflects the crevice

corrosion sensitivity of the stainless steel. The more locations of crevice sites were attacked by crevice corrosion, the poorer performance of stainless steel to resist crevice corrosion. Figure 6 shows the statistical diagram of the number of crevice sites attacked by crevice corrosion on four kinds of stainless steel after cyclic polarization testing. As can be seen from Fig. 6, the number of crevice sites attacked by crevice corrosion can be described with the following sequence: SS904L (31) > SS316 (29) > SS254sMo (16) > SS2507 (14). In addition, the maximum depth of crevice corrosion can also reflect the resistance to crevice corrosion of stainless steel. Figure 7 shows the statistical diagram of the maximum depth of each crevice corrosion sites on four kinds of stainless steel. As shown in Fig. 7, the maximum depth of crevice corrosion sites can be described with the following sequence: SS904L > SS316 > SS254sMo > SS2507. The maximum crevice corrosion depths on SS904L and SS316 are larger than 140

Fig. 5 Macromorphology of SS904L artificial crevice corrosion specimen **a** and picture of crevice depth measurement **b**

Fig. 6 Statistical diagram of the number of crevice sites attacked by crevice corrosion on four kinds of stainless steel after cyclic polarization testing

and 120 μm , respectively, which are significantly greater than SS254sMo and SS2507.

Figure 8 shows the sketch map of depth distribution of crevice corrosion [17]. According to the IR drop theory, three regions extending from the boundary of the crevice to the center inside the crevice can be classified: (A) passive region, (B) active region and (C) variable region [17, 26–28]. Meanwhile, the active region of crevice corrosion site on stainless steel consists of three subregions: (B₁) severely attacked region, (B₂) commonly attacked

Fig. 7 Statistical diagram of the maximum depth of each crevice corrosion sites on four kinds of stainless steel

region and (B₃) lightly attacked region [17]. Surface morphologies of the four kinds of stainless steel crevice corrosion sites are further investigated by SEM, as shown in Figs. 9, 10, 11 and 12, respectively.

Figure 9a shows the morphology of a typical crevice corrosion site on SS316. Figure 9b shows the magnified morphology of the active region. Figure 9c–e is local microscopic morphologies of the active region, respectively, called severely attacked region, commonly attacked region and lightly attacked region. As can be seen from

Fig. 8 Sketch map of depth distribution of crevice corrosion [17]

Fig. 9 SEM micrographs of different regions on a typical crevice corrosion site of SS316 after cyclic polarization testing

Fig. 9c, d, in the severely and commonly attacked region, austenite grain boundary is preferentially corroded, and there is ladder-like corrosion morphology on the grain surface. As shown in Fig. 9e, there is no austenite grain boundary on the surface of lightly attacked region and the corrosion morphology is dense ravine-like.

Figure 10a shows the morphology of a typical crevice corrosion site of SS904L, which also includes passive

region, active region and variable region. It can be seen from the top right corner of the crevice corrosion site that there is a “cover” structure over the edge of the active region. Figure 10b is the high-magnification morphology of “cover” structure. According to the EDS results from the red area of Fig. 10b, the “cover” structure is thin oxide slice composed of Fe, Cr, Mo and Ni elements (Fig. 10c), which is similar to the “lace cover” structure formed in the

Fig. 10 SEM micrographs of different regions on a typical crevice corrosion site of SS904L after cyclic polarization testing

metastable pitting period of stainless steel [29]. As can be seen from Fig. 10d, the right and left parts of the picture should be the severely attacked region and the commonly attacked region, respectively. Their corrosion morphologies are similar to each other. Attributing to preferential corrosion, the austenite grain boundary is clearly visible, and the grain surface is relatively smooth. Figure 10e shows the corrosion morphology of lightly attacked region, and the friction scratches by sandpaper are still visible, illustrating that the corrosion is mild in this area. Some pits with the diameters of about 50 μm are found, illustrating that the surface is prone to pitting corrosion in this area.

Figure 11a shows the morphology of a typical crevice corrosion site on SS254sMo. Figure 11b is the high-magnification morphology of active region. Being similar to SS904L, there is also “cover” structure over the edge of the active region. Figure 11c shows the high-magnification morphology of “cover” structure, the size of which is significantly smaller than that of SS904L. According to the EDS results from the red area in Fig. 11c, the “cover” structure is also thin oxide slice composed of Fe, Cr, Mo and Ni elements (Fig. 11d). Figure 10e–g shows the corrosion morphologies of severely, commonly and lightly attacked region, respectively. As can be seen from Fig. 11e, in the severely attacked region, austenite grain boundary is preferentially corroded

and the grain surface is uneven. Figure 11f shows that the corrosion morphology of commonly attacked region is dense ravine-like, and the austenite grain boundary is also preferentially corroded. Figure 11g shows the corrosion morphology of lightly attacked region. Being similar to SS904L, the friction scratches by sandpaper are visible, and some small size pits are also found in this area.

Figure 12a shows the corrosion morphology of a corner on a typical crevice corrosion site of SS2507. As can be seen from Fig. 12b, there is also “cover” structure over the edge of the active region. Being different from the thin oxide slice structures of SS904L and SS254sMo, the “cover” structure of 2507 stainless steel consists of many micron-sized small holes, which is more like the “lace cover” structure of metastable pitting in stainless steel morphologically [29, 30]. Figure 12c–e corresponds to the corrosion morphologies of severely, commonly and lightly attacked region, respectively. The phenomena of galvanic corrosion were observed in the three corrosion region. SS2507 is duplex stainless steel, which consists of ferrite phase (α) and austenite phase (γ). Tsai and Chen [13] have fabricated single-phase electrodes of α -phase and γ -phase from duplex stainless steel and studied the galvanic corrosion between two phases. They found that, in an acid solution containing Cl^- , the corrosion potential of ferrite

Fig. 11 SEM micrographs of different regions on a typical crevice corrosion site of SS254sMo after cyclic polarization testing

phase (α) is normally lower than that of austenite phase (γ) and is preferentially corrosion as an anode phase [13]. The local environment of active region of crevice structure is also acid solution containing Cl^- , and passive film was partly dissolved attributing to the local acidification. As the passive film gradually loses its protective properties, galvanic corrosion occurs between the two phases, and the ferrite phase (α) preferentially corroded, while austenite phase (γ) remained. This result agrees with the previous related studies about corrosion resistance of duplex stainless steel [13, 31, 32]. As can be seen from Fig. 12c, the corrosion morphology on the surface of retained austenite phase (γ) is honeycomb in severely attacked region. Figure 12d shows that, in commonly attacked region, there is a great difference in corrosion depth between the two phases, which means that the galvanic corrosion is the most serious here. The corrosion damage is mild in lightly attacked region, and the friction scratches by sandpaper are also still visible, as shown in Fig. 9e.

Identifying from the statistical results of the number and depth of crevice corrosion pits, the most serious crevice corrosion occurred on SS904L followed by SS316, SS254sMo and SS2507 in the two times concentrated seawater at 70 °C. Attributing to the high amounts of alloying elements, namely chromium, molybdenum and nitrogen, SS254sMo and SS2507 are called supper stainless steels, and their crevice corrosion resistances are remarkably superior to ordinary stainless steel SS316. However, being known as supper stainless steel, the crevice corrosion resistance of SS904L is interestingly not as good as ordinary stainless steel SS316.

From the observation of crevice corrosion microscopic morphology, the “cover” structures exist over the edge of the active region of all the three kinds of supper stainless steels. The “cover” structure is often observed in metastable pitting of stainless steel, which is called “lace cover,” while rarely observed in the crevice corrosion process [29, 30]. The classical study shows that, in the

Fig. 12 SEM micrographs of different regions on a typical crevice corrosion site of SS2507 after cyclic polarization testing

process of transition from metastable to steady pitting of stainless steel, as an additional barrier for the ionic current passing through the pit anolyte and the exterior electrolyte, the “lace cover” could sustain pit growth [29]. According to the same principle, the “cover” structure can also accelerate the corrosion process of stainless steel in the crevice. Related studies show that the “cover” structure mainly consists of the passive film which had originally covered the unattacked metal [29]. It is known that alloying elements like chromium, molybdenum and nitrogen are favorable for the stabilization of passive film [8]. As the two primary forms of chromium, CrOOH can reduce the defects in the passive film, and CrO_4^{2-} was found to be beneficial as potent inhibitor [8, 33–35]. Molybdenum can induce the formation of a bipolar layer in the form of molybdate and promote the generation of Cr_2O_3 by accelerating the migration of O^{2-} [36–41]. Nitrogen can stabilize the passive film in the form of NO_3^- ; meanwhile, CrN which is incorporated of nitride and bulk chromium could impede the dissolution of metal ions [42–47]. The PREN calculated by formula (1) can be described as the following sequence: SS316 (22.665) < SS904L (33.432) < SS254sMo (45.86) $\approx$ SS2507 (47.36).

Therefore, the passive film of SS316 is the most unstable, and there are “cover” structures over the edge of the active region of all the three kinds of super stainless steel except SS316.

Taking into account the role of “cover” structure in accelerating crevice corrosion, the crevice corrosion resistance of the 316 stainless steel should be better than those of SS904L, SS254sMo and SS2507. However, the self-repairing ability of the passive film inside the crevice is another major factor affecting the development of crevice corrosion. According to the results of cyclic polarization curves, the order of self-repairing ability can be described with the following sequence: SS2507 > SS254sMo > SS904L > SS316. The self-repairing ability of the passive film also depends on the amounts of alloying elements chromium, molybdenum and nitrogen. Obviously, nitrogen in the form of NO_3^- plays the most important role in the process of repassivation [42–46]. Both SS254sMo and SS2507 contain trace amounts of nitrogen, and their PREN is much higher than those of SS316 and SS904L. That is why the resistances to crevice corrosion of SS254sMo and SS2507 are significantly better than those of SS316 and SS904L. The self-repair ability of passive

film inside the crevice for SS904L is slightly better than SS316. Meanwhile, due to the barrier mass transfer effect of large-size “cover” structure, the crevice gap of SS904L is more blocked than that of SS316. Between the two opposing factors, the barrier mass transfer effect of large-size “cover” structure plays a more prominent role; thus, the crevice corrosion of SS904L is more serious than that of the SS316.

4 Conclusions

1. In the two times concentrated seawater at 70 °C, the crevice corrosion occurred on the surface of all the three kinds of supper stainless steels. The crevice corrosion damage of SS904L is the most serious (even more serious than the ordinary stainless steel SS316), and those of SS254sMo and SS2507 stainless steel are the slightest.
2. Preferential corrosion in austenite grain boundary was observed in severely and commonly attacked regions of austenite stainless steels (SS316, SS904L and SS254s). The phenomena of galvanic corrosion were observed in the active crevice corrosion region of duplex supper stainless steel (SS2507), and the ferrite phase (α) was preferentially corroded, while the austenite phase (γ) was remained.
3. Due to the stable passivation film of high-alloyed supper stainless steel, there are “cover” structures over the edge of active crevice corrosion regions of SS904L, SS254sMo and SS2507, but SS316 is an exception.
4. Attributing to the barrier mass transfer effect of large-size “cover” structure, the crevice gap of SS904L is more blocked than that of SS316, and thus, the crevice corrosion of SS904L is more serious than SS316.

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